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INFLUENCE OF HEATING CONDITIONS ON THE THERMOLUMINESCENCE SENSITIVITY OF TLD – LiF PHOSPHORS

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INFLUENCE OF HEATING CONDITIONS ON THE THERMOLUMINESCENCE SENSITIVITY OF TLD – LiF PHOSPHORS [1]

Toshiyuki Nakajima[2] and Shigueo Watanabe [3]

ABSTRACT

In order to obtain some information on the mechanism of the thermoluminescence phenomena of TLD – LiF phosphors the heating-rate effects on the dose dependence of the sensitivity were studied The heating rate effect was scarcely found from the NTL 50p LiF phosphor but from the TLD – 100 and – 700 LiF phosphor such effect was found when the intensity of the thermoluminescence was measured by means of the integrating method However such effect was hardly obtained from the TLD – LiF series by the glow-peak height method The causes of the effect were discussed It has been proposed that the dosimetric properties obtained by various workers have to be compared with each other under the same thermal conditions

INTRODUCTION

Thermoluminescence dosimeters (TLD) of LiF material have the following advantages 1) dependence of the thermoluminescence sensitivity (the thermoluminescence intensity per unit absorbed dose) on the radiation energy is small 2) thermal neutrons as well as X γ and β rays can be measured using a small amount of the material 3) the material can be used repeatedly and 4) the procedure required for the dose measurement is very simple Therefore the dosimetric properties of the LiF material have been studied by many workers For example the non linear sensitivity against the irradiation dose [2] the variation in the sensitivity due to linear energy transfer (LET) of radiation to the irradiated material [3] variation in energy dependence of the TLD material due to the irradiation dose [6] and the LET dependence of the sensitization factor[1] have been observed

To explain these properties of the TLD sensitivity, Claffy and her co workers[4] proposed a track model from an observation of the correlation between the color centers and the glow peaks of the LiF thermoluminescence

Suntharalingam and his co workers[9] also proposed a competing trap model to explain the dosimetric properties of the LiF – TLD In the competing trap model it is assumed that the competing trap has a larger cross section of electron capture than that of the dosimetric trap that there are fewer competing traps than the other traps and that an activation energy of the dosimetric trap is less than that of the competing trap However it is uncertain whether or not the dosimetric properties of TLD have been obtained under the same thermal conditions by all the workers

According to the Randall and Wilkins[8] model of the thermoluminescence the intensity is strongly affected by such thermal conditions as the heating rate and the maximum heating temperature Therefore the results obtained under the same thermal condition should be compared when the thermoluminescence mechanism model is discussed however, the effects of these conditions on the sensitivity have not yet been fully investigated

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The present report will cover the influence of the heating rate and the maximum heating temperature on the thermoluminescence sensitivity

EXPERIMENTAL

In this work TLD 100 LiF, TLD 700 LiF, CaF_2 (Dy) which had been purchased from the Harshaw Chem Co and NTL 50p LiF prepared by the Aloka Co Tokyo were used as thermoluminescence phosphors All of these phosphors were small crystalline in form

To ensure an uniform build up of the secondary electrons over the entire volume of the phosphor, the phosphor was enclosed in a black polyethylene capsule with a wall thickness of 2 mm and was irradiated at room temperature in the dark at an exposure of 200 to 70 000 r with γ rays from a ^{137}Cs or ^{60}Co source

To obtain the glow curves, the irradiated phosphor was uniformly spread in a thin layer on about 1 cm^2 of a heater pan and then heated from room temperature up to 450°C at a heating rate of 10°C/min which was controlled with an automatic temperature controller Emitted light was passed through a filter of a $CuSO_4$ solution to cut off the infrared emission from the heater pan, and detected by means of a Toshiba photomultiplier tube of 7696(S 11) Output current of the photomultiplier tube was measured with a micromicroammeter and recorded with the heating temperature by means of a two pen recorder

The influence of the heating rate and the maximum heating temperature on the thermoluminescence sensitivity is ascertained as follows The total intensity of the thermoluminescence and the glow-peak height were measured for all cases, after the phosphor had been kept for at least 24 hrs, by using an EG and G TLD reader or Dai Nippon Toryo TLD 1200 SD reader With these readers it is possible to vary both the heating rate and the maximum heating temperature freely In this work the heating rate was varied from 5°C/sec to 27°C/sec and the maximum temperature from 200°C to 450°C

RESULTS

1 – INFLUENCE OF HEATING RATE ON THE SENSITIVITY

The changes in the relative thermoluminescence sensitivity of the TLD 100 LiF phosphor as a function of the exposure are shown in Figure 1 In this figure the vertical axis is the relative integrated thermoluminescence intensity of the phosphor which is heated up to 300°C after irradiation at room temperature

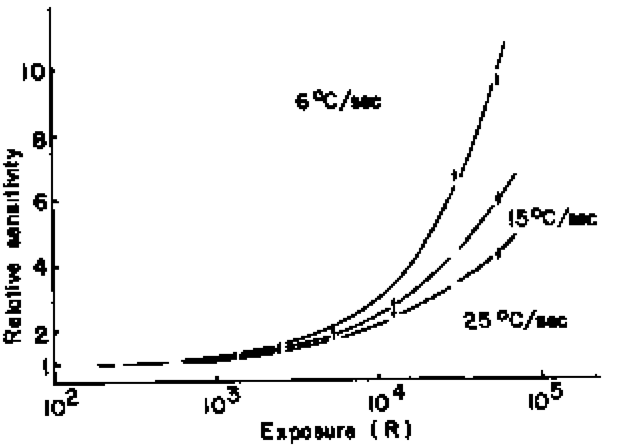

Figure 1 — Heating rate effect on dose dependence of the sensitivity of TLD 100 LiF phosphor heated at the maximum heating temperature of 300°C as a function of exposure and as a parameter of heating rate (thermoluminescence intensity was evaluated with intergrating method)

As is shown in Figure 1, the dose dependence of the sensitivity was increased by decreasing in the heating rate Especially, in the case of a high dose-irradiated phosphor the dose dependence is extremely influenced by the heating rate On the other hand in the case of a low dose irradiated phosphor it seems that the difference in the sensitivity resulting from a change in the heating rate is small For example when the irradiated phosphors were heated at the heating rate of 6°C/sec a ratio between the sensitivities of the 3×10^4 r irradiated and the 63 r irradiated phosphors was 6 75 but at 25°C/sec the ratio between them was 3 20 Measurements made in the present work on the Harshaw TLD 700 LiF phosphor showed that the influence of the heating rate on the dose dependence of the sensitivity was roughly similar to that on the the TLD 100 LiF phosphor

Figure 2 and Table I present the results of such measurements for an NTL 50p LiF the host crystal of which is similar to those of the TLD 100 and TLD 700 LiF phosphors CaF_2(Dy) phosphor In Figure 2 the vertical axis is the integrated thermoluminescence intensity of the phosphors which are heated to 300°C after irradiation at room temperature

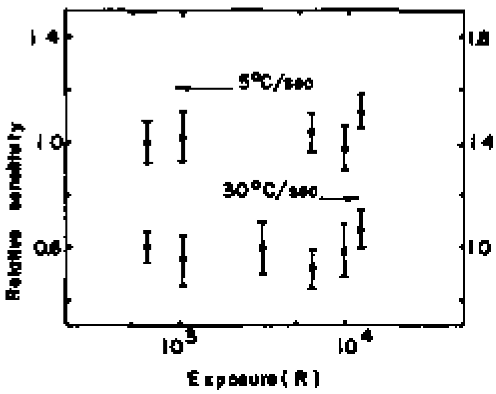

Figure 2 — Heating rate effect on dose dependence of the sensitivity of NTL 50p LiF phosphor heated at the maximum heating temperature of 300°C as a function of exposure and as a parameter of heating rate (thermoluminescence intensity was evaluated with integrating method)

Table I

Heating rate effect on the dose dependence of the sensitivity of CaF_2(Dy) phosphor irradiated at room temperature (maximum heating temperature is 300°C)

Exposure (R)	Relative thermoluminescence intensity		
	Heating rate		
	5°C/sec	15°C/sec	25°C/sec
$2 4 \times 10^3$	0 053 ± 0 0050	0 057 ± 0 0029	0 057 ± 0 0025
$3 0 \times 10^3$	0 064 ± 0 0036	0 066 ± 0 0058	0 075 ± 0 0042
$1 7 \times 10^4$	0 37 ± 0 031	0 42 ± 0 0058	0 37 ± 0 016
$2 7 \times 10^4$	0 38 ± 0 033	0 43 ± 0 022	0 41 ± 0 010
$4 2 \times 10^4$	0 60 ± 0 032	0 62 ± 0 018	0 62 ± 0 022
$1 0 \times 10^5$	1 00 ± 0 014	1 00 ± 0 028	1 00 ± 0 025

The relative thermoluminescence intensity of CaF_2(Dy) as measured by the integrating method is given in Table I. As can be seen in Table I and Figure 2, the dose dependence of the sensitivity on the heating rate is very small with the NTL 50p LiF and CaF_2(Dy). A difference in the heating rate effect on the sensitivity between the TLD and NTL 50p LiF phosphors has been found from the results shown in Figures 1 and 2. The causes of this difference will be discussed in the Discussion section.

The effects of the heating rate on the sensitivity with the samples had been observed by measuring the integrated thermoluminescence intensity. However, the use of the glow peak height method for evaluating the thermoluminescence intensity is also important in searching for the causes of the difference in the heating rate effects on the sensitivity between the TLD LiF series and NTL LiF phosphors, because different effects may be expected with different methods.

Figure 3 shows the changes in the relative sensitivity of the TLD 100 LiF phosphor as a function of the exposure as determined using the glow peak height method.

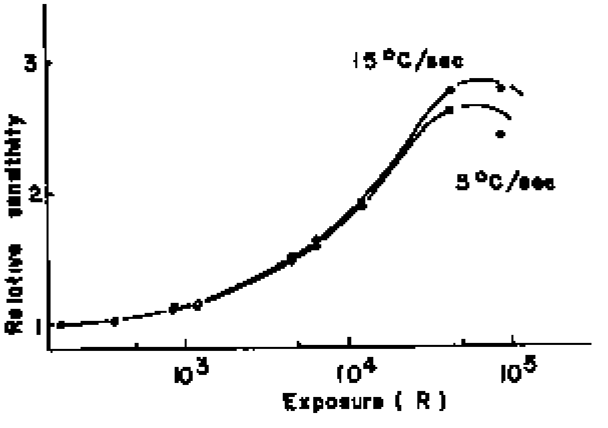

Figure 3 – Heating rate effect on dose dependence of the sensitivity for dominant glow peak height of TLD 100 LiF phosphor heated at 300°C as a function of exposure

It may be seen from Figure 3 that the heating rate effect on the dose dependence of the relative sensitivity in the TLD 100 LiF phosphor is hard to find using the glow peak height method, and the effect is varied with means of the termoluminescence measuring method. For example the sensitivity of the 3×10^4 r irradiated phosphor obtained using the integrating method was about 1.3 times that of the 4.4×10^4 r irradiated one using the glow peak height method when the relative sensitivity of the irradiated phosphors was compared with the measuring methods at 15°C/sec. This result suggests that the glow curve shape changes with an increase in the absorbed dose.

2 – MAXIMUM HEATING TEMPERATURE EFFECT ON THE SENSITIVITY

Next, a study of the glow curves and of the maximum heating temperature effect on the thermoluminescence sensitivity of the phosphor is undertaken in order to obtain some information on the causes of the different heating rate effects due to the difference in the measuring method.

Figure 4 shows the dose dependence of the sensitivity of the TLD 100 LiF phosphor with a parameter of the maximum heating temperature. The vertical axis is the integrated thermoluminescence intensity of the phosphor heated at the heating rate of 15°C/sec.

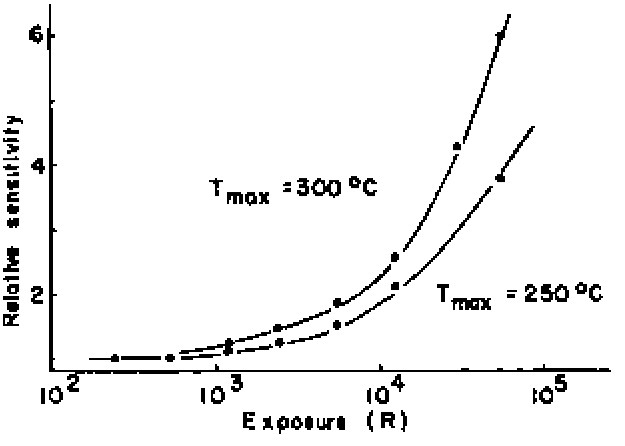

Figure 4 — Dose dependence of the sensitivity of TLD 100 LiF phosphor as a function of exposure and as a parameter of heating rate (the intensity was evaluated with the integrating method)

As is shown in Figure 4 it is evident that the dose dependence of the sensitivity is increased by increasing the maximum heating temperature If the glow curve shape is not changed with the absorbed dose no changes in the dose dependence of the sensitivity will be observed Then the influence of the absorbed dose on the glow curve shape of the TLD 100 and NTL 50p LiF phosphors and the CaF_2 (Dy) phosphor was studied at a constant heating rate of 10°C/min

Figure 5 presents the changes in the glow curve shape of TLD 100 LiF due to the irradiated dose When this phosphor was irradiated to a dose of a few hundred rad its glow curve contained five glow peaks while no glow peaks in the high temperature region of about 270°C or more could be found Moreover a predominant glow peak appeared at 250°C However as shown by Curves B and C in Figure 5, when the phosphor was exposed to a relatively high dose of 3000 r or more some glow peaks were found on the high temperature side of the glow curve these newly appearing glow peak grow with an increase in the exposure Figure 6 shows changes in the height of the new glow peaks which appear on the high temperature side of the glow curve In this figure the vertical axis is the relative glow peak height per unit exposure normalized to the peak height at 250°C per unit exposure When the 250°C peak height is assumed to be proportional to the exposure the glow peak height except the 250°C peak increase monotonously with the exposure, and after the increase the peak height is saturated On the other hand in order to compare the changes in the glow curve with exposure the NTL 50p LiF phosphor was used for the present experiment as well as TLD 100 LiF one Figure 7 shows the glow curve of NTL 50p LiF irradiated with 10^3 r It shows a glow curve shape similar to that of TLD 100 LiF phosphor at a low dose the predominant glow peak appeared at about 260°C However no new glow peak could be found from the phosphor on the high temperature side of the glow curve even if the phosphors was irradiated with 10^6 r The influence of the dose on the glow-curve shape is thus entirely different between these phosphors

It had been described that the changes in the dose dependence of the sensitivity due to the heating rate could not be obtained from NTL 50p LiF though they could be obtained from the TLD 100 LiF using the integrating method It may be inferred that the different effect of the heating rate is caused by a change in the glow curve shape with the exposure This suggestion is supported by the following experimental results

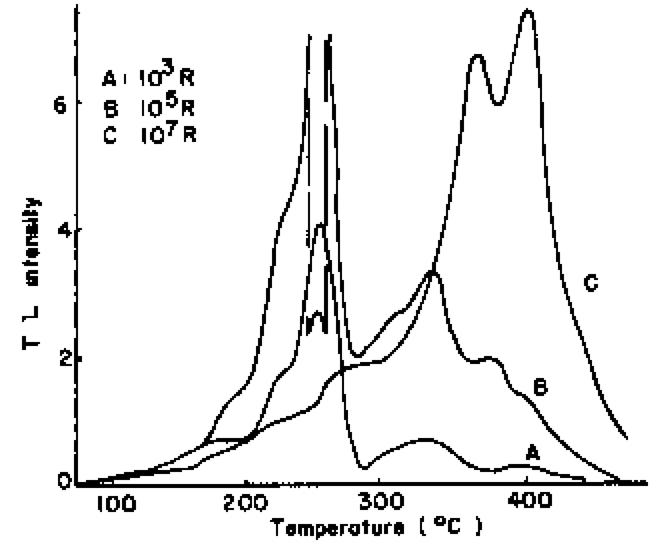

Figure 5 — Changes in glow curve shape of TLD 100 LiF phosphor as a parameter of exposure

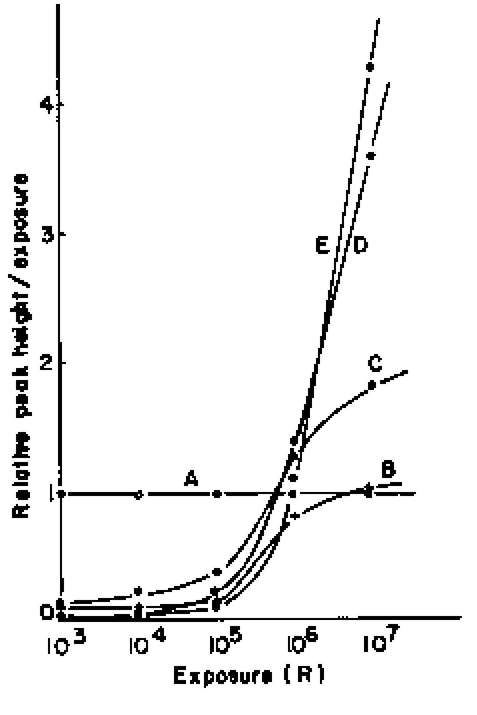

Figure 6 — Ratios of glow peak heights to 250°C peak per exposure as a function of exposure Curve A is glow peak at 250°C curve B at 310°C curve C at 340°C, curve D at 370°C and curve E is glow peak at 400°C

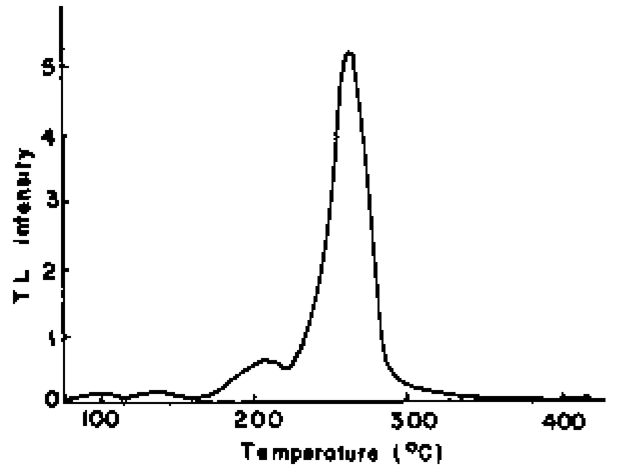

Figure 7 — Glow curve of NTL 50p LiF phosphor irradiated at room temperature with 10^3r

Figure 8 shows the sensitization phenomena of NTL 50p LiF irradiated with a test dose of 50r after having been sensitized with both irradiation with γ rays of ^{60}Co source and thermal treatment at 300°C for 5 min These phenomena so well known in the Harshaw TLD – LiF series and the Mg_2SiO_4(Tb) phosphor[1 7] were not observed from the NTL 50p LiF phosphor Furthermore the relationship between the thermoluminescence intensity of the NTL 50p LiF and the exposure maintains the linearity of the response in the dose range from 30 to 2000r, as shown in Figure 9, but a supralinearity of the thermoluminescence response of the phosphor to the exposure which has been observed in the series of Harshaw TLD – LiF phosphors, could not be observed in the NTL 50p LiF phosphor rather a sublinearity was observed Although the TLD – LiF series and NTL 50p LiF are from the same host crystal with the two LiF different relationship between the response and dose the sensitization phenomena and the heating rate effects on the dose dependence of the sensitivity were obtained The causes of the differences will be discussed below

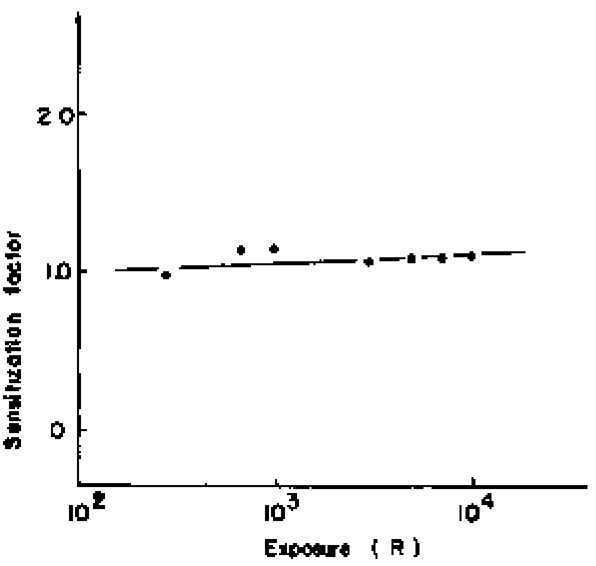

Figure 8 — Sensitization factor of NTL 50p LiF phosphor irradiated with test dose of 50r, after annealed at 300°C for 5 min and irradiated with γ rays from ^{60}Co source for sensitizing

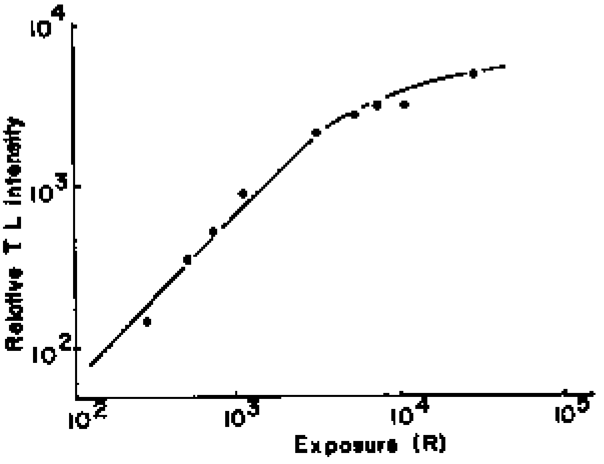

Figure 9 — Changes in the integrated thermoluminescence intensity of NTL 50p LiF phosphor as a function of exposure

DISCUSSION

The TLD phosphors may be classified into two groups One of them consists of the phosphors which have the properties of supralinearity and sensitization phenomena of the response to the irradiation dose while the other consists of those with sublinearity and non sensitization phenomena The TLD – LiF series ordinary LiF and the Mg_2SiO_4(Tb) phosphors are typical of the first group while NTL 50p LiF belongs to the latter group It has been considered that the difference is caused by a different tendency of changes in the glow curve to the absorbed dose of the phosphor That is in the former group the glow curve shape in the high temperature region changes with the absorbed dose For example as shown in Figure 6 the new glow peaks are born and grow in the high temperature region with an increase in the irradiation dose On the other hand in the latter group the glow-curve shape hardly changes with the dose and also no newly born glow peaks are observed in the high temperature region

When the thermoluminescence intensity was evaluated by the integrating method the heating rate effect on the dose dependence of the sensitivity was not found for the phosphor in the latter group but it was found in the former This will be considered below with due regard to the influence of the dose on the glow curve shape

According to Halperin and Braner s TL model [5] the integrated thermoluminescence intensity under a given glow curve is proportional to the number of the released or recombined hole m_g

$$m_g = (1/\beta) \int_{T_g}^{\infty} I \, dT = (1/\beta) \, I_g \, \delta \qquad (1)$$

where β is the heating rate T_g the glow peak temperature I_g the thermoluminescence intensity at the peak position where $\delta = T_2 - T_g$ T_2 being the temperature at a half intensity at the fall off point of the peak If it is assumed that the re trapping probability of a thermally released electron from the trap is negligible in comparison with the probability of recombination, it can be shown that m_g is proportional to the number of the trapped electron in the trap n and that equation (1) is rewritten as follows

$$m_g = (1/\beta) \int_{T_0}^{\infty} \gamma n \, dT \qquad (2)$$

where γ is the probability of thermal excitation

This equation reveals that the thermoluminescence intensity is represented as a multiplication of the number of trapped electron and the kind of transition or recombination probability $(1/\beta) \int_{T_0}^{\infty} \gamma \, dT$ and that if the glow curve shape of the phosphor does not change with the irradiation dose and if the maximum heating temperature is given, the dose dependence of the sensitivity is not affected by the heating rate because there are no changes in ratios among the trapped carrier densities in each trap as a result of changes in the dose and the absence of changes in the ratios among the thermally released carriers from the traps due to changes in the heating rate Therefore, the heating rate effect on the sensitivity could not be obtained from NTL 50p LiF phosphor the glow curve shape of which does not change with the dose This corresponds to the above assumption

In the case of the TLD – LiF series, the glow curve shape changes with the dose the number of the trapped carriers per unit of the dose in the deeper traps increases with the absorbed dose and the dose dependence of the sensitivity varies with the heating rate The increase in the dose dependence of the sensitivity due to the decrease in the heating rate will be discussed below

According to the Randall and Wilkins s model the glow peak temperature, T^* is a function of the heating rate as in equation (3)

$$(\beta E)/(ks) = T^{*2} \exp(-E/kT^*) \qquad (3)$$

where E is the activation energy k the Boltzmann constant, s the frequency factor and β the heating rate

It may be concluded from equation (3) that the decrease in the heating rate causes the transfer of the glow peak temperature to a lower temperature region Furthermore according to both the Halperin and Braner model and the Randall and Wilkins model more trapped carriers are released from the trapping levels at a given maximum heating temperature with a decrease in the heating rate Therefore it seems that the thermoluminescence intensity caused by the thermally released carriers from the deeper traps is added to the thermoluminescence intensity for the dosimetry when the integrating method is used for evaluating the irradiation dose

If the trapped carrier density per unit the dose in each trap is randomly changed with the dose, the dose dependence of the sensitivity is also randomly changed with the heating rate Furthermore the thermoluminescence intensity is represented as the product of the multiplication of n and the probability as is given in equation (2) In case of the TLD – LiF series when the phosphor is warmed at a high heating rate a large amount of the thermoluminescence intensity is occupied by the thermally released carriers from the so called dosimetric traps because the thermoluminescence intensity is hardly affected by releasing the trapped carriers in the deep traps and because of the transfer of the peak temperature to the high temperature region at a high heating rate However when the phosphor is warmed at a lower heating rate the thermoluminescence intensity is consistent with emission from the thermally released carriers from both the dosimetric traps and the deeper traps because of the transfer of the glow-peak temperature to the lower temperature region with a low heating rate Therefore, the dose dependence of the sensitivity at a low heating rate is more than that at a high heating rate

From the present study we expect that the heating rate effect on the energy dependence of the sensitivity as well as the dose dependence of the sensitivity of the irradiated phosphor can be observed, that is the energy dependence of the thermoluminescence sensitivity of the irradiated phosphor may decrease under some maximum heating temperatures with an increase in the heating rate In the near future we will report on this subject

It may be considered that the different effect of the heating rate on the dose dependence between the integrating method and the glow peak height method are caused by the following factors The glow peak height is given as a function of both the number of the trapped carriers and the heating rate as is shown in equation (2) but no changes in the glow peak height are brought about with the transfer of the glow peak temperature due to changes in the heating rate because the temperatures of all the peaks are transferred with changes in the heating rate as given in equation (3) Therefore, the dose dependence of the sensitivity is hardly affected by changes in the heating rate

Heat conduction from the heater to the phosphor must be considered in discussing the heating rate effect but it has been neglected in this discussion

CONCLUSION

When the heating rate effects on the dose dependence of the thermoluminescence sensitivity of the TLD – LiF series and NTL 50p LiF were measured by the integrating method for evaluating the irradiation dose the tendency of the heating rate effects were not in agreement between different phosphors For example in the case of the TLD – LiF series the dose dependence of the sensitivity was found to increase with a decrease in the heating rate while in the case of the NTL 50p LiF phosphor no effect was found at all in the dose range from about 600 to 20 000 r This difference is caused by changes in the glow-curve shape due to the irradiation dose The effect obtained in the TLD – LiF series and the difference between the TLD and NTL 50p LiF phosphors could not be explained by either of the thermoluminescence models proposed by the Randall and Halperin groups

It may be concluded from these results that in the case of the evaluation of the radiation dose by means of the integrating method the comparison of the dosimetric properties of the high dose irradiated phosphors obtained by each worker has to be carried out using results obtained under the same thermal conditions If in addition both the lower-dose irradiated phosphor and the glow peak height method are used in evaluating the radiation dose the comparison of the results will become more exact

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RESUMO

A fim de obter algumas informações sobre o mecanismo de termoluminescencia de fósforos TLD LiF efeitos da taxa de aquecimento sobre a dependencia com a dose de sensibilidade foram estudados O efeito da taxa de aquecimento foi pouco observado no material NTL 50p LiF mas em TLD 100 LiF e TLD 700 LiF tal efeito é observado quando a intensidade da termoluminescencia foi medida por meio do método de integração Entretanto tal efeito é dificilmente encontrado na série de TLD LiF pelo método da altura do pico As causas desse efeito foram discutidas Foi proposta a necessidade de comparar em iguais condições técnicas as propriedades dosimétricas encontradas por diferentes autoridades

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