

DEPENDENCE OF GLOW CURVE STRUCTURE ON THE CONCENTRATION OF DOPANTS IN LiF:Mg,Cu,Na,Si PHOSPHOR

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Abstract — The dependence of dopants concentration on glow curve structure of newly developed LiF:Mg,Cu,Na,Si phosphor has been investigated. LiF samples with different dopants concentration were prepared by varying the concentrations of Mg (0–1.0 mol%), Cu (0–1.0 mol%) and NaSi (0–2.4 mol%) and their glow curve shapes were analysed after ^{137}Cs γ irradiation. The intensity of the main peak has a strong influence on Mg concentration. The intensity of the high temperature peak tends to be reduced with increasing Cu concentration; Cu inhibits the growth of the high temperature peak. NaSi was an effective dopant to obtain the high intensity of the low temperature peak and main peak. The optimum composition of dopants was found to be Mg 0.6 mol%, Cu 0.8 mol% and NaSi 1.8 mol%. This phosphor has about two times higher sensitivity than that of LiF:Mg,Cu,P.

INTRODUCTION

A new TL material, LiF:Mg,Cu,Na,Si phosphor with high sensitivity and good fading characteristics was developed by Doh *et al.*^(1,2). LiF:Mg,Cu,Na,Si has a similar glow curve shape to LiF:Mg,Cu,P, but the relative TL sensitivity of LiF:Mg,Cu,Na,Si is higher by about 2 times than that of LiF:Mg,Cu,P⁽³⁾. Hence it can be used for the measurement of low doses effectively, but the glow curve structure and the dosimetric characteristics are not yet well known. Doh *et al.*⁽¹⁾ had selected the dopants composition of LiF:Mg,Cu,Na,Si phosphor, but this was not an optimum composition because Mg and Cu contents were determined by varying the two concentrations simultaneously.

In the present work the effects of individual dopants (Mg, Cu, NaSi) in LiF:Mg,Cu,Na,Si phosphor have been investigated by analysis of the glow curve structures and the TL intensities as a function of dopants concentration. The optimum dopants composition for this material having the maximum TL intensity has been also determined.

MATERIALS AND METHODS

The set of samples used in this experiment was prepared in two ways: (1) by varying the concentration of one dopant while fixing the concentrations of the others; (2) by subtracting one or two dopants in the LiF:Mg,Cu,Na,Si of Doh *et al.* (LiF:Mg,Cu,Na,Si material doped with the fixed concentrations of the previous study⁽¹⁾ as Mg and Cu 0.6 mol%, NaSi 1.8 mol%). NaSi was considered as a single dopant because the compound NaSiO₃·9H₂O was added. The former samples were fabricated by varying dopant concentrations in following ranges: Mg 0.2–1.0 mol%, Cu 0.2–1.0 mol% and NaSi 1.6–2.4 mol%. The last has two sets of samples, one set (LiF:Cu,Na,Si, LiF:Mg,Na,Si and

LiF:Mg,Cu) was fabricated by subtracting one dopant in LiF:Mg,Cu,Na,Si of Doh *et al.*, the other set (LiF:Mg, LiF:Cu and LiF:Na,Si) was made by adding only one dopant in the host LiF material. Otherwise, the concentration of dopants was the values used by Doh *et al.*⁽¹⁾.

The fabricating procedure for LiF:Mg,Cu,Na,Si phosphor was as follows. A host material LiF was thoroughly mixed with dopants of the following compounds: MgSO₄·7H₂O, CuSO₄·5H₂O and NaSiO₃·9H₂O in distilled water. The solution was mixed by a magnetic stirrer and dried in an oven at 353 K. The wet mixture was sintered in a muffle furnace at 1073 K for 30 min under a controlled nitrogen atmosphere. The sintered material in the crucible was quickly quenched to room temperature and then pulverised in a mortar by hand to give a grain size about 200 mesh.

These samples were irradiated to 10 mGy with ^{137}Cs γ rays at Korea Atomic Energy Research Institute (KAERI) except for one impurity doped sample. Because these samples had low TL sensitivity, the higher dose irradiation of ^{137}Cs γ ray was needed. The glow curves of the samples were subsequently measured using a commercial TLD reader (System 310, Teledyne Brown Engineering) controlled by a personal computer. Measurements were carried out with a linear heating rate 5 K.s⁻¹ to 573 K in a nitrogen flow. Individual TL peak intensity was given by the light output and was estimated by the maximum TL peak heights.

The glow curve of LiF:Mg,Cu,Na,Si phosphor could be divided into three parts by a thermal bleaching method as shown in Figure 1: low temperature peaks, main peak having maximum amplitude in the glow curve and high temperature peak at the end of the main peak. The high temperature peak height was estimated as TL peak height at 518 K. Only one TL peak of the two in the low temperature region was considered because of the difficulty in separating the peaks.

RESULTS AND DISCUSSION

Variation of one dopant concentration in LiF:Mg,Cu,Na,Si

When Mg, Cu and NaSi contents in LiF:Mg,Cu,Na,Si were varied one by one, the dependence found on dopants concentration is presented in Table 1. The TL peak heights of Table 1 were obtained from the measured glow curve. For the purpose of analysis of individual peaks, the values of Table 1 are displayed as the

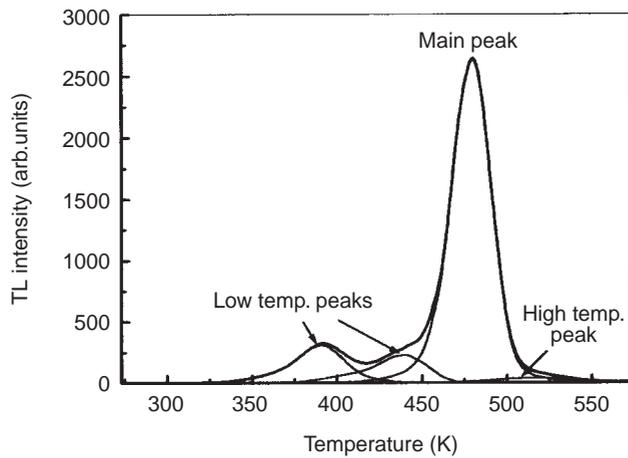


Figure 1. The typical glow curves of LiF:Mg,Cu,Na,Si phosphor. Samples are irradiated with 0.1 Gy of ¹³⁷Cs γ rays. Heating rate is 5 K.s⁻¹.

dependence of the TL intensity of individual peaks on dopants concentration in Figures 2, 3 and 4.

Figure 2 shows low temperature peak heights of one dopant variable. The TL intensities of Mg and Cu variables grow with slowly increasing the concentration, reaching a maximum value and then decrease. In the

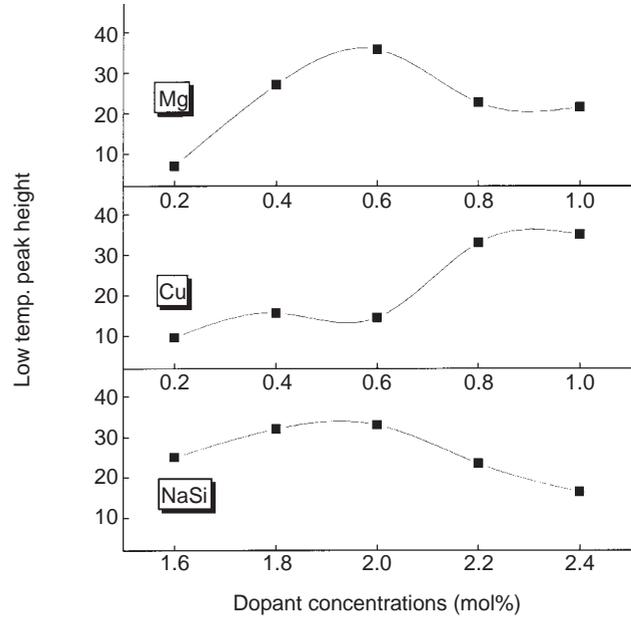


Figure 2. The relative peak heights of the low temperature peaks of LiF:Mg,Cu,Na,Si with one variable dopant, Mg, Cu and NaSi, respectively.

Table 1. The individual peak heights of the glow curve are presented with a variation of dopant concentrations in LiF:Mg,Cu,Na,Si phosphor.

Dopants	Concentration (mol%)	TL peak heights (arb. units)		
		Low temp. peak	Main peak	High temp. peak
Variable: Mg (Fixed: Cu 0.6 mol%, NaSi 2.0 mol%)	0.2	7.0	55.3	20.5
	0.4	27.0	441.4	26.9
	0.6	35.7	565.9	17.8
	0.8	22.7	391.6	19.0
	1.0	21.5	392.6	12.5
Variable: Cu (Fixed: Mg 0.6 mol%, NaSi 2.0 mol%)	0.2	9.6	407.6	73.7
	0.4	15.7	510.5	33.0
	0.6	14.5	520.5	16.7
	0.8	14.5	576.2	15.5
	1.0	35.1	433.3	18.4
Variable: NaSi (Fixed: Mg 0.6 mol%, Cu 0.6 mol%)	1.6	25.1	425.6	9.1
	1.8	32.0	632.1	8.7
	2.0	33.0	632.1	8.7
	2.0	33.0	576.0	13.4
	2.2	23.5	467.8	12.5
	2.4	16.5	310.7	25.4

case of the Cu variable, low temperature peak height successively increases as the Cu content rises; this fact agrees with the study on LiF:Mg,Cu,P^(4,5).

In general, the low temperature peaks are subject to fading and the high temperature peak signal is difficult to distinguish between the phosphor luminescence and the infrared black-body radiation. For this reason, the

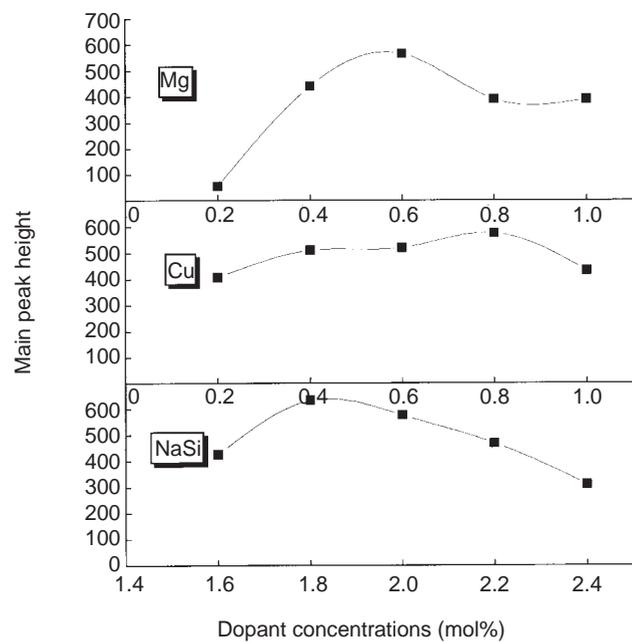


Figure 3. The relative peak heights of the main peaks of LiF:Mg,Cu,Na,Si with one variable dopant, Mg, Cu and NaSi, respectively.

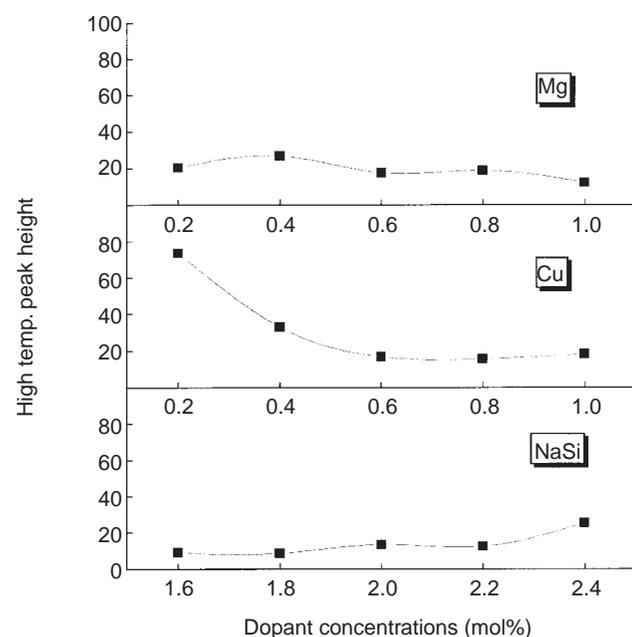


Figure 4. The relative peak heights of the high temperature peaks of LiF:Mg,Cu,Na,Si with one variable dopant, Mg, Cu and NaSi, respectively.

main peak is taken as a relevant signal for practical applications while the other peaks are excluded. In Figure 3, the main peak heights have a maximum for a certain amount of dopant in all cases. Hence one can determine the optimum dopants composition which maximises the main peak TL intensity: this is found to be: Mg 0.6 mol%, Cu 0.8 mol% and NaSi 1.8 mol%. The main peak heights were strongly influenced by the Mg and NaSi concentrations but showed small variation in the Cu concentration. It can be seen that Mg and NaSi dopants are strongly related to the main peak, but Cu does not contribute to the main peak. This result is somewhat different from that of Wang⁽⁴⁾ and Bilski *et al.*⁽⁶⁾. It may be thought that the difference in the results is caused by the amount of dopant concentration. In this experiment, the Cu concentration of LiF:Mg,Cu,Na,Si was varied between 0.2 and 1.0 mol%, these values are larger than the optimum Cu concentration (0.05 mol%) of LiF:Mg,Cu,P.

In Figure 4, high temperature peak height is rapidly reduced with an increasing Cu concentration, which agrees with other authors⁽⁴⁻⁶⁾. However LiF:Mg and LiF:Na,Si have a flat low intensity on the whole. This means that Mg and NaSi do not contribute to the high temperature peak trap, which is somewhat unlike the study of LiF:Mg,Cu,P⁽⁴⁻⁶⁾.

Variation of the doped impurity: fixed dopants concentration

The glow curves of LiF:Cu,Na,Si, LiF:Mg,Na,Si and LiF:Mg,Cu are shown in Figure 5, those of LiF:Mg, LiF:Cu and LiF:Na,Si are shown in Figure 6. In Figure 5, the glow curve of LiF:Cu,Na,Si that showed the effect of Mg absence shows the reduced main peak. The main peak intensity of LiF:Mg in Figure 6 is relatively high in the glow curve. This means that Mg particularly contributes to an increase in the main peak traps. LiF:Mg,Na,Si without Cu has a high intensity in the

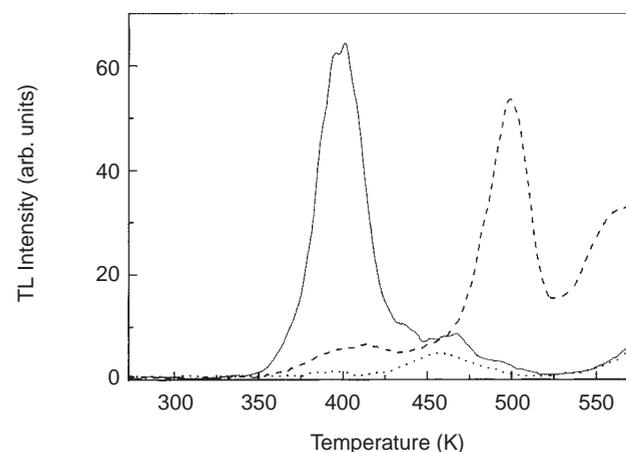


Figure 5. The glow curves of doubly doped samples: (—) LiF: Cu,Na,Si, (---) LiF: Mg,Na,Si and (····) LiF: Mg,Cu.

high temperature peak and no effect on the other peak intensities. The glow peak of LiF:Cu was dominant only in the low temperature region. From these results it could be seen that the intensity of the high temperature peak tends to be reduced with an increasing Cu concentration, that is, Cu inhibits the growth of the high temperature peak. The glow curve of LiF:Mg,Cu had almost no TL intensity and that of LiF:Na,Si has the highest TL intensity. This means that NaSi is a very effective dopant in the construction of traps in the low temperature peaks and main peak.

TL glow curve and sensitivity

Figure 7 shows the typical TL glow curves of MCP-N, GR-200P and LiF:Mg,Cu,Na,Si phosphors in powder form. MCP-N and GR-200P were annealed at 240°C for 10 min and LiF:Mg,Cu,Na,Si was used as virgin phosphor.

In the shapes of the glow curves, one can see that both side peaks of the main peak of LiF:Mg,Cu,Na,Si are lower than the others. The TL sensitivities of MCP-N and GR-200P have nearly the same value. It is shown that the TL sensitivity of LiF:Mg,Cu,Na,Si is about two

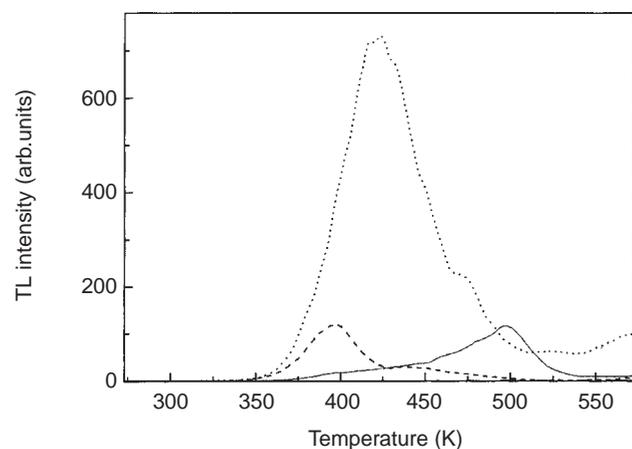


Figure 6. The glow curves of one dopant sample: (—) LiF: Mg, (---) LiF:Cu and (····) LiF: Na,Si.

times higher than that of LiF:Mg,Cu,P, and the main peak temperature of the LiF:Mg,Cu,Na,Si occurs at a slightly higher temperature than that of LiF:Mg,Cu,P.

CONCLUSIONS

The analysis of the glow curve structure of LiF:Mg,Cu,Na,Si permits the following conclusions about the correlation between TL peaks and particular dopants. The change of the Mg content has a strong influence on the intensity of the main peak. The role of Cu concentration is to raise the low temperature peak and to reduce the high temperature peak. NaSi was a very effective dopant in producing the high intensity of the low temperature peak and the main peak.

The optimum composition of dopants concentration was found to be Mg 0.6 mol%, Cu 0.8 mol% and NaSi 1.8 mol%. This phosphor has about two times higher sensitivity than that of LiF:Mg,Cu,P, it can be used for the measurement of low doses effectively.

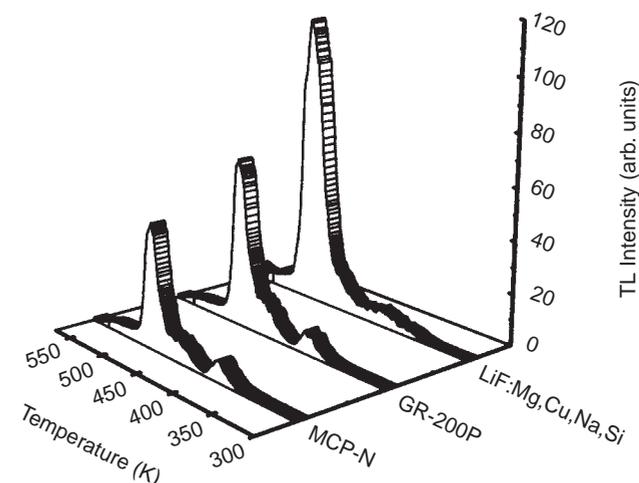


Figure 7. Typical TL glow curves of phosphors for a constant heating rate of 5 K.s⁻¹. MCP-N (LiF:Mg,Cu,P: Poland); GR-200P (LiF:Mg,Cu,P: China) and LiF:Mg,Cu,Na,Si. Irradiation dose was 10 mGy by a ¹³⁷Cs γ source.

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