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ESTIMATION AND CORRECTION FOR THE TEMPERATURE LAG IN THERMOLUMINESCENT MEASUREMENTS FOR LiF:Mg,Cu,Na,Si PHOSPHOR

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Abstract. *In the current work the glow curves for LiF:Mg,Cu,Na,Si thermoluminescent material were measured at various heating rates in the range from 1 K/s to 30 K/s. In the thermoluminescent measurements a contact heating was used to heat the powder samples. The temperature lag between the heating element and the dosimeter was estimated and corrected by applying Kitis-Tuyn's method. Some kinetic parameters of the traps in LiF:Mg,Cu,Na,Si were evaluated using a variable-heating-rate method.*

I. INTRODUCTION

Nowadays, it is well-known that thermoluminescence dosimetry (TLD) is a dosimetric technique with applications in areas such as personnel, environmental and clinical dosimetry. TLD is based on thermoluminescence (TL) materials which after exposure to ionising radiation emit light when they are heated. It is found that the impurities doped in TL material create localized energy levels (trapping levels) in the forbidden energy band gap and that these are crucial to the TL process. The study of the TL glow curve, which is a plot TL intensity versus temperature, is used to determine the trapping parameters. However, the TL response strongly depends on heating rate [1–10]. In addition, in TL measuring experiment the temperature is usually that of the heating element measured by the thermocouple fixed on it, but it is not exactly the temperature of the TL material. However, in order to extract physical information from TL glow curve, it is essential to know the sample temperature rather than that of the heating element. Contact heating is the most commonly used type of heating. In this case, the temperature of the sample usually differs from the temperature of the heating element measured by thermocouple placed on it. This difference is caused by temperature gradients in the heating element, non-ideal thermal contact between the heater element and the sample (temperature lag (TLA)), temperature gradients (TG) across the sample. The difference between the actual temperature of the sample and the measured temperature obtained from the heating element has been the object of study by different authors [5, 11–15]. The TLA and the TG could influence the values of the trapping parameters of a glow peak [12, 13]. Kitis and Tuyn [14, 15] proposed a simple method to correct for the temperature lag based on TL measurements only.

A powder-type of LiF doped with four dopants: Mg, Cu, Na and Si was first developed by Kim et al in 1989 [16, 17]. The LiF:Mg,Cu,Na,Si material, being a promising material for thermoluminescent dosimetry, has attracted the researchers' attention in the last ten years. The effect of the dopant concentrations on the TL glow curve in LiF:Mg,Cu,Na,Si phosphor was investigated [18–21]. The influence of the heating rate on TL response for LiF:Mg,Cu,Na,Si phosphor was reported by Ha et al [22]. However, to the best of our knowledge, up to now, there is almost no work about the evaluation of the trapping parameters in the LiF:Mg,Cu,Na,Si phosphor.

The aim of the present work is to estimate and correct the temperature lag between the heating element and the dosimeter by applying Kitis-Tuyn's method. Some kinetic parameters of the traps for the main TL glow peak in LiF:Mg,Cu,Na,Si were evaluated using a variable-heating-rate method.

II. EXPERIMENTAL

Phosphor preparation

Synthesis process for LiF:Mg,Cu,Na,Si TL powders described elsewhere [20, 21]. The host LiF material was prepared by using precursor chemicals such as lithium chloride (LiCl) and fluohydric acid (HF). The doping with Mg, Cu, Na and Si was performed by mixing the compounds containing required activator: $MgCl_2.6H_2O$, $CuCl_2.H_2O$ and $Na_2SiO_3.9H_2O$ in appropriate ratio. Our results of analysis of the dependence of the intensity for the TL glow curves on the dopant concentration [21] indicated that the optimum dopant concentrations were 0.2 mol% Mg, 0.6 mol% Cu and 2.0 mol% NaSi, which in good agreement with data reported by Nam et al [18].

Thermoluminescence measurements

The LiF:Mg,Cu,Na,Si powders were irradiated by X-ray radiation. All the samples were irradiated to the same dose of 0.1 Gy from an X-ray tube (RORIX, RFT, Germany) 1.5 kW in power with a cobalt target, operated at 20 kV and 1-5 mA. The total irradiation duration was 3 minutes. The TL glow curves of the powders were subsequently measured by using a commercial TLD reader (Harshaw TLD-3500) controlled by a computer. In the TL readout, the 5 mg of the phosphor powders were filled in a hole of 3 mm in diameter on a copper plate. The powder layer had a thickness below 1 mm. The TL glow curve measurements were carried out with linear heating rates from 1 K/s up to 30 K/s in temperature range from 323 K to 633 K. The TL measurements at a given heating rate were repeated 5 times and the results are represented by the average values with the mean deviation. The measurement data were processed using the software ORIGIN.

III. RESULTS AND DISCUSSION

As mentioned above, an important effect that has to be taken in to consideration to avoid large error in the trapping parameter determination is the temperature lag (TLA). There are various types of sample heating in the TL measurements: contact heating, hot gas heating, laser heating and microwave heating. Then the TLA may be caused by temperature gradients in the heating element, non-ideal thermal contact between heater element and sample, temperature gradients across the sample. These processes are determined by three physical heat transfers: conduction, convection, radiative loss. In the case of contact heating, the heat transfer mechanism by conduction plays a dominant role [12, 13] and the non-ideal thermal contact is the main cause for the temperature differences between the heating element and the sample. In addition, the authors of the works [13, 15] have found that for both LiF:Mg,Ti and Al_2O_3:C samples less than 1 mm thick, the temperature gradient across the sample is relatively small.

The TLA, as well as the thermal gradient (TG) across the dosimeter itself, can strongly influence on the trapping parameter evaluation. Kitis and Tuyn [14,15] proposed a simple method to correct for the temperature lag based on TL measurements only. This method is based on the following equation:

$$T_j = T_i - c\ln\left(\frac{\beta_i}{\beta_j}\right) \tag{1}$$

where T_j and T_i are the maximum temperatures of a glow peak with heating rates β_j and β_i, respectively, and c is a constant, which is determined by using two very low heating rates where the TLA can be considered a negligible. Kitis et al. [14] emphasized that the measurements at the low rates of heating are reference measurements, so they need a special attention. This could be achieved in two ways: first, using silicon oil of high thermal conductivity between the heating element and sample; and second, for these reference measurements one can use loose powder instead of chips, with or without silicon oil, improving the accuracy further.

Using the equation (1) and the calculated value of c, the new maximum temperatures $T_{m,corr}$ with correction for the TLA at high heating rates are evaluated relative to the lowest heating rate. The TLA (ΔT) in the position of the peak maximum is given by

$$\Delta T = T_{m,raw} - T_{m,corr} \tag{2}$$

where $T_{m,raw}$ is the glow-peak maximum temperature including the TLA.

The analysis results for the main glow-peak maximum are presented in Table 1. Column 2 shows the main glow-peak maximum temperatures of the raw data at different heating rates. Constant c was calculated by using the following expression

$$c = \frac{T_{m,raw,\beta=2} - T_{m,raw,\beta=1}}{\ln 2} \tag{3}$$

where $T_{m,raw,\beta=1}$ and $T_{m,raw,\beta=2}$ are the values of $T_{m,raw}$ for heating rates of 1 K/s and 2 K/s. The value of c was found to be 11.48. Column 3 shows the values of the maximum temperatures calculated by using equation (1), i.e. corrected $T_{m,corr}$. The temperature lags at the positions of the peak maxima are shown in column 4. In our case, like as [1], the samples are powder layers having less than 1 mm thickness, so the temperature gradient across the sample is relatively small. Therefore, the TLA is mainly caused by the non-ideal thermal contact between the heater element and the sample.

The effective heating rates β_{eff} were calculated using the expression proposed by Kitis and Tuyn [14]:

$$\beta_{eff} = \frac{T_{m,raw} - T_o - \Delta T}{T_{m,raw} - T_o}\beta \tag{4}$$

Table 1. Results of the analysis for LiF:Mg,Cu,Na,Si.

β (K/s)	$T_{m,raw}$ (K)	$T_{m,corr}$ (K)	ΔT (K)	β_{eff} (K/s)	$T_{m,corr,eff}$ (K)
1	490.4±1.0	490.4±1.0	0.0	1	490.4±1.0
2	498.4±1.0	498.4±1.0	0.0	2	498.4±1.0
5	510.9±1.0	508.9±1.0	2.0	4.94	508.7±1.0
10	522.6±1.0	516.8±1.0	5.8	9.74	516.5±1.0
15	529.8±1.2	521.5±1.2	8.3	14.39	521.0±1.2
20	536.0±1.7	524.8±1.7	11.2	18.91	524.1±1.7
30	546.2±1.0	529.4±1.0	16.8	27.82	528.6±1.0

where T_o is the starting temperature, ~293 K. The values of β_{eff} are listed in column 5. Finally, the maximum temperatures $T_{m,corr,eff}$ calculated with the effective heating rates are listed in column 6.

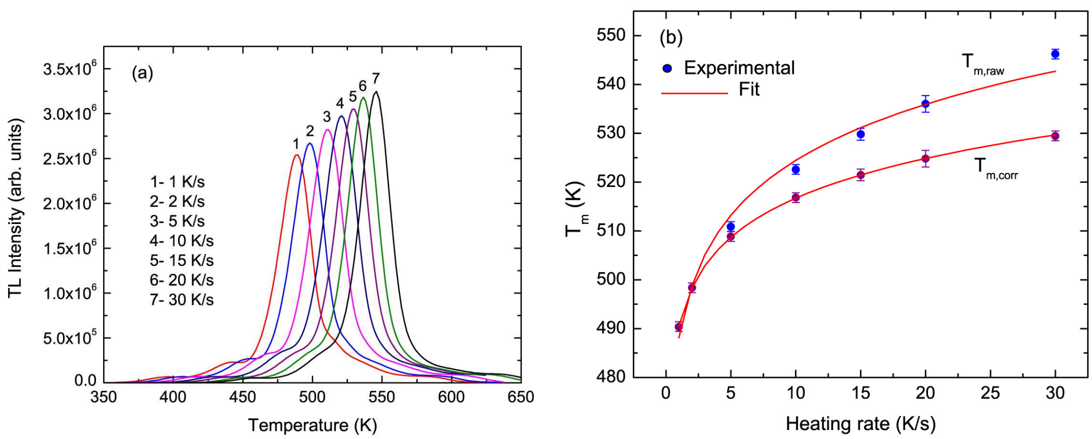

Fig. 1. (a) Glow curves of the LiF:Mg,Cu,Na,Si powders measured at different heating rates.(b) The main glow-peak maximum temperature as a function of heating rate for LiF:Mg,Cu,Na,Si phosphor, curve $T_{m,raw}$ is plotted from the data including the TLA and curve $T_{m,corr}$ is plotted from the data after correction for the TLA.

Figure 1(a) shows typical glow curves of LiF:Mg,Cu,Na,Si powder measured at some heating rates. As seen from the figure, the whole TL glow curve is shifted to higher temperatures and the height of the main glow peak increases with increasing heating rate in the range from 1 K/s to 30 K/s. We focused only on the main glow peak because it usually used in the dosimetric measurements. Figure 1(b) shows the plot of the main glow-peak maximum temperature (T_m) before and after correction for the TLA against heating rate.

Like as [1], we suppose that the change of the T_m against heating rate follows an equation of the form $\ln(T_m \text{ or } I_m) = A + B\ln(\beta)$, where A and B are constants and β is the heating rate. For the glow-peak maximum temperature including the TLA, denoted by $T_{m,raw}$, $A = 6.1906 \pm 0.0034$; $B = 0.0312 \pm 0.0015$; for the glow-peak maximum temperature corrected for the TLA, denoted by $T_{m,corr}$, $A = 6.19559 \pm 0.00024$; $B = 0.0225 \pm 0.0001$. Using these constants, we calculated the dependence of T_m on β. The results are shown by the solid lines in figures 1(b). It can be noted that the fitted lines are in rather good agreement with experimental data.

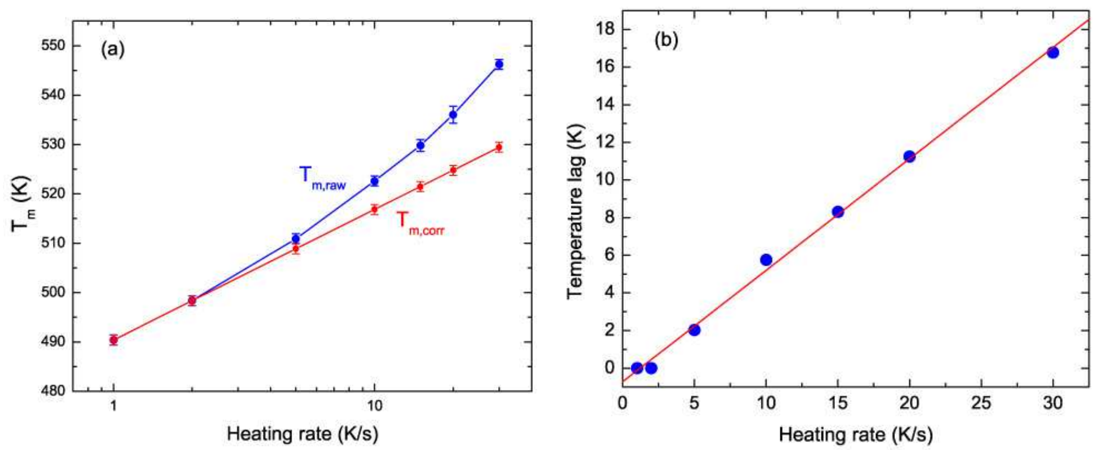

Fig. 2. (a) Peak maximum temperature of the main glow peak as a function of logarithm of the heating rate for LiF:Mg,Cu,Na,Si. Curve $T_{m,raw}$ is plotted from the data including the TLA and curve $T_{m,corr}$ is plotted from the data after correction for the TLA. (b) The temperature lag as a function of the heating rate for the main glow peak of LiF:Mg,Cu,Na,Si.

The behaviour of the main glow-peak temperature at the maximum plotted as a function of logarithm of the heating rates is illustrated in figure 2(a). The results indicated that a TLA of 2.0 K is already evident at a heating rate of 5 K/s. This TLA reaches a value of 16.8 K at a heating rate of 30 K/s.

In addition, it is found that the TLA has a good linear relation to the heating rate (see figure 2(b)), like that observed early by other authors [12–14] for LiF:Mg,Ti and Al_2O_3:C. For our LiF:Mg,Cu,Na,Si samples the linear relationship is $\Delta T = (-0.72 \pm 0.22) + (0.59 \pm 0.01)\beta$.

The variable heating rate method is based on the shift position of the glow-peak maximum temperature as a function of the heating rate. From expression (4) of Ref. [22], one obtains

$$\ln\left(\frac{T_m^2}{\beta}\right) = \ln\left(\frac{E}{sk}\right) + \frac{E}{kT_m} \tag{5}$$

It can be noted that plot of $\ln(T_m^2/\beta)$ versus $1/(kT_m)$ must be a straight line with slope E, and the intercept of this plot is $\ln(E/sk)$, which allows evaluating the frequency factor s.

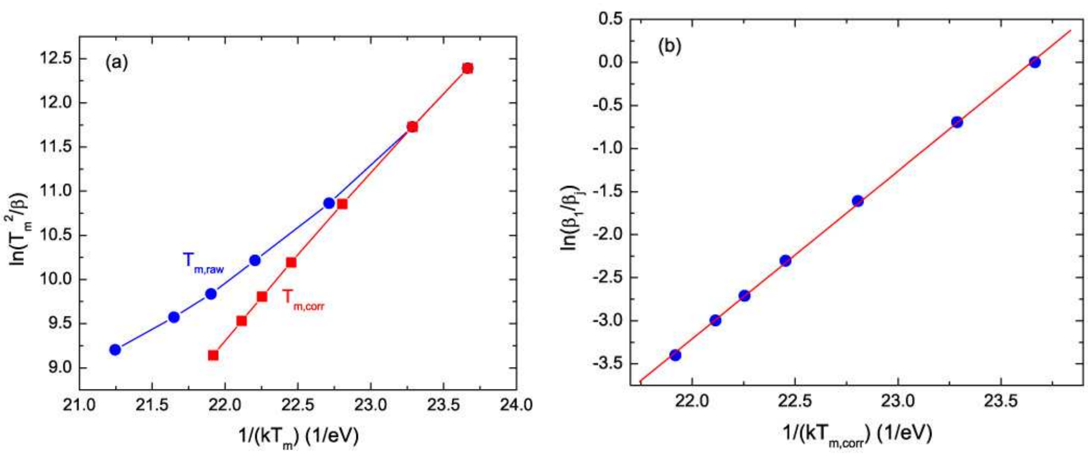

Fig. 3. a) Plot of $\ln(T_m^2/\beta)$ against $1/(kT_m)$ for LiF:Mg,Cu,Na,Si. Curve $T_{m,raw}$ is plotted from the data including the TLA and curve $T_{m,corr}$ is plotted from the data after correction for the TLA. (b) Plot of $\ln(\beta_i/\beta_j)$ versus $1/(kT_{m,corr})$.

Figure 3(a) shows a plot of $\ln(T_m^2/\beta)$ as a function of $1/(kT_m)$ for LiF:Mg,Cu,Na,Si. As seen from the figure, curve $T_{m,raw}$ plotted with the data including the TLA is not a straight line, but as expected, curve $T_{m,corr}$ plotted with the corrected data is a very good straight line with slope $E = 1.86 \pm 0.02$ eV. The intercept of this straight line is found to be -31.6 ± 0.4 from which one obtains $s = 1.14{\times}10^{18}$ s^{-1}.

From the equation proposed by [14, 23]

$$T_j = T_i - (T_jT_i)\frac{k}{E}\ln\Big(\frac{\beta_i}{\beta_j}\Big) \tag{6}$$

one finds out:

$$\ln\Big(\frac{\beta_i}{\beta_j}\Big) = -\frac{E}{kT_i} + \frac{E}{kT_j} \tag{7}$$

Thus, plot of $\ln(\beta_i/\beta_j)$ versus $1/(kT_m)$ must be a straight line with slope E, and the intercept of this plot is $-E/(kT_m)$ which allows to evaluate the maximum temperature T_m corresponding to the heating rate β_i. The relationship between $\ln(\beta_i/\beta_j)$ and $1/(kT_m)$ is described in figure 3(b). It can be seen from the figure, the plot of $\ln(\beta_i/\beta_j)$ against $1/(kT_m)$ is a very good straight line with slope $E = 1.95 \pm 0.02$ eV. The intercept of this plot is -46.07 ± 0.04, from which one finds out the glow-peak maximum temperature corresponding to the heating rate of 1 K/s is 491.2 K, which is in good agreement with the experimental data.

It must be noticed that the use of the data listed in columns 5 and 6 for the effective heating rates gives the same results as well.

In the Table 2 are listed the values of the trapping parameters resulting from the deconvolution of the glow curve in our previous report [22] and from the variable-heating-rate method for the main glow peak in LiF:Mg,Cu,Na,Si in the current work. All errors

Table 2. Evaluated trapping parameters of the main glow peak in LiF:Mg,Cu,Na,Si.

Method	E (eV)	**Intercept**	s (s^{-1})
Glow-curve deconvolution*, $\beta = 1$ K/s	1.89 ± 0.04	—	2.59×10^{18}
Variable-heating-rate method			
$\ln(T_m^2/\beta)$ versus $1/(kT_m)$	1.86 ± 0.02	-31.6 ± 0.4	1.14×10^{18}
$\ln(\beta_i/\beta_j)$ versus $1/(kT_m)$	1.95 ± 0.02	-46.07 ± 0.04	—

*Data from our previous work [22]

were obtained by fitting the measured data. It must be noted that our values of activation energy are lower than a value of 2.33 eV for the glow peak 4 in LiF:Mg,Cu,Na,Si (KAERI) dosimeter [24], but the work [24] did not indicate which method was used to evaluate the activation energy. The values of the trapping parameters obtained in the current work are approximate to that from the work [14] for the glow peak 5 in LiF:Mg,Ti ($E = 1.96 - 2.07$ eV; $s = 10^{18}$–10^{21} s^{-1}).

IV. CONCLUSION

The influence of the heating rate in the range from 1 K/s to 30 K/s on the TL response was investigated. It was found that the glow-peak maximum temperature T_m increased in the whole range of heating rates studied. The change of the T_m against heating rate follows an equation of the form $\ln(T_m) = A + B\ln(\beta)$. The TLA between the heating element and the dosimeter reached a value of 16.8 K at a heating rate of 30 K/s. This TLA was corrected by using Kitis-Tuyn's method. The variable-heating-rate method was as well applied to evaluate the kinetic parameters of the traps for the main glow-peak in the TL of LiF:Mg,Cu,Na,Si.

REFERENCES

[1] G. Kitis , M. Spiropulu , J. Papadopoulos, S. Charalambous, *Nucl. Instrum. Methods Phys. Res. B*, **73** (1993) 367.

[2] N.A. Sooner, A.D. Franklin, *Radiation Measurements*, **35** (2002) 59.

[3] A.S. Pradhan, *Radiat. Prot. Dosim.*, **58** (1995) 205.

[4] A. Abtahi, P. Braunlich, T. Haugan, P. Kelly, *Radiat. Prot. Dosim.*, **17** (1986) 313.

[5] G.C. Taylor, E. Lilley, *J. Phys. D: Appl. Phys.*, **15** (1982) 2053.

[6] A.J.J. Bos, R.N.M. Vijverberg, T.M. Piters, S.W.S. McKeever, *J. Phys. D: Appl. Phys.*, **25** (1992) 1249.

[7] A.J.J. Bos, T.M. Piters, *Radiat. Prot. Dosim.*, **47** (1993) 41.

[8] F.O. Ogundare, F.A. Balogun, L.A. Hussain, *Radiation Measurements*, **40** (2005) 60.

[9] L.Z. Luo, K.J. Velbeck, M. Moscovitch, J.E. Rotunda, *Radiat. Prot. Dosim.*, **119** (2006) 184.

[10] A.S. Pradhan, J.I. Lee, J.L. Kim, K.S. Chung, H.S. Choe, K.S. Lim, *Radiation Measurements*, **43** (2008) 361.

[11] D.S. Betts, L. Couturier, A.H. Khayrat, B.J. Luff, P.D. Townsend, *J. Phys. D: Appl. Phys.*, **26** (1993) 843.

[12] D.S. Betts, P.D. Townsend, *J. Phys. D: Appl. Phys.*, **26** (1993) 849.

[13] T.M. Piters, A.J.J. Bos, *J. Phys. D: Appl. Phys.*, **27** (1994) 1747.

[14] G. Kitis, J.W.N. Tuyn, *J. Phys. D: Appl. Phys.*, **31** (1998) 2065.
[15] G. Kitis, J.W.N. Tuyn, *Radiat. Prot. Dosim.*, **84** (1999) 371.
[16] S.H. Doh, M.C. Chu, W.H. Chung, H.J. Kim, D.S. Kim, Y.H. Kang, *Korean Appl. Phys.*, **2** (1989) 425.
[17] H.J. Kim, W.H. Chung, S.H. Doh, M.C. Chu, D.S. Kim, Y.H. Kang, *J. Korean Phys. Soc.*, **22** (1989) 415.
[18] Y.M. Nam, J.L. Kim, S.Y. Chang, *Radiat. Prot. Dosim.*, **84** (1999) 231.
[19] J.L. Kim, J.I. Lee, S.Y. Chang, K.S. Chung, H.S. Choe, *Radiation Measurements*, **38** (2004) 435.
[20] Vu Thi Thai Ha, Nguyen Thi Quy Hai, Nguyen Ngoc Long, Le Van Vu, *Hanoi National University Journal of Science, Mathematics - Physics*, **23** (2007) 225.
[21] Vu Thi Thai Ha, Nguyen Thi Quy Hai, Nguyen Ngoc Long, *Hanoi National University Journal of Science, Mathematics - Physics*, **25** (2009) 83.
[22] Vu Thi Thai Ha, Nguyen Thi Quy Hai, Nguyen Ngoc Long, *Communications in Physics* **21** (2011) 43.
[23] Shachar B. Ben, Y.S. Horowitz, *J. Phys. D: Appl. Phys.*, **25** (1992) 694.
[24] M. Budzanowski, J.L. Kim, Y.M. Nam, S.Y. Chang, P. Bilski, P. Olko, *Radiation Measurements*, **33** (2001) 537.

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