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## CONTINUOUS MODEL FOR TL TRAPS\*

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### ABSTRACT

Analysis of glow curves in  $\text{CaF}_2$ :natural demonstrated that some glow peaks in this material do not obey the simple model of thermoluminescence proposed by Randall and Wilkins. In particular these peaks do not decay exponentially during isothermal heating; and, moreover, the peak positions change during such treatment. A model based on the superposition of two peaks was first proposed, wherein two closely spaced, unresolved peaks were assumed to add to give the observed behavior. The calculations revealed that this model can reasonably well explain the isothermal decay curves, but not the isothermal change in the peak position. As a result, a second model was developed based on a narrow, continuous distribution of trap depth, thus modifying the model of Randall and Wilkins which is based on only one distinct trap depth. This second model, which supposes a Gaussian distribution for the trap depths, was used to explain the behavior of two glow peaks in natural  $\text{CaF}_2$ , giving satisfactory fits for the glow curve, for the isothermal decay, and for the isothermal change in the peak position. The application of this model also enables calculation of the trap parameters, which had not been previously accomplished. Analysis based on this continuous model was also extended to similar behavior of 280°C and 370°C peaks in the glow curve of dosimetric  $\text{LiF:Mg}$ . Isothermal decay, and isothermal change in the position of these peaks were measured for this material.

### INTRODUCTION

Randall and Wilkins proposed a model which has been widely used to fit observed glow curves of thermoluminescent crystals. This model assumes a well defined depth for traps giving rise to a glow peak.

If we denote by  $E$  the trap depth, the probability per unit time at absolute temperature  $T$  for a trapped electron to escape can be written as

$$p = \zeta^{-1} = s \exp(-E/kT) \quad (1)$$

where  $k$  is the Boltzmann constant,  $\zeta$  is the mean-lifetime, and  $s$  is the frequency factor. If, further, we denote by  $\beta$  the heating rate of the TL reader, and by  $n(E, T_0)$  the number of trapped electrons at temperature  $T_0$ , then after heating the sample to temperature  $T$ , the number of remaining electrons is given by:

$$n(E, T) = n(E, T_0) \exp \left[ - \int_{T_0}^T \frac{s}{\beta(T)} \exp(-E/kT) dT \right] \quad (2)$$

Emitted light at temperature  $T$  is given by

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\* Based in part upon portions of a thesis submitted by Spero Penha Morato to the Institute of Physics, University of São Paulo, in partial fulfillment of the requirements for the Master's degree.

. 2 .

$$I(T) = s \exp(-E/kT) n(E,T) \quad (3)$$

Let us now consider an irradiated sample subjected to an isothermal annealing at temperature  $T_a$  during a time  $t_a$ . The number of electrons left in the traps is given by

$$n(E, T_a, t_a) = n(E, T_a, 0) \exp \left[ -st_a \exp(-E/kT_a) \right] \quad (4)$$

In the subsequent reading cycle this number acts as the initial number of trapped electrons in Eq.(3). It is, therefore, easy to recognize that at peak temperature  $T_p$ ,

$$\log I(T_p) = \text{const.} - st_a \exp(-E/kT_a) \quad (5)$$

This linear dependence of  $\log I(T_p)$  on  $t_a$  is a typical result of the Randall and Wilkins model.

Furthermore, the peak temperature  $T_p$  is a constant for each peak.

Measurements on fading or other isothermal decay carried out by several investigators<sup>1,2,3</sup> indicate, however, that  $\log I(T_p)$  does not always follow a linear law. Hence, the glow curve does not obey the Randall and Wilkins equation.

We extended post-annealing decay measurements to Brazilian fluorite (peaks I, II and III) and to the 280°C and 370°C peaks in LiF:Mg, again finding a behavior not predicted by the Randall and Wilkins model.

## EXPERIMENTAL METHODS

### a) Natural Calcium Fluoride

Green colored samples of natural calcium fluoride extracted from a mine located in Criciuma, Santa Catarina State, Brazil, were pulverized and sieved through 80 or to 200 mesh Tyler screens to be used in this experiment. To eliminate TL induced in the fluorite by natural surrounding radioactivity, the samples were heated for 10 minutes at 580°C and then at 400°C for 2 hours. Subsequently they were irradiated to 100 R with Cs-137 gamma-rays, to be annealed isothermally at temperatures listed in Table I for times varying between 15 and 180 minutes. Then the TL was read on a CON-RAD model 5100 reader.

TABLE I

PEAK	POST-ANNEALING TEMPERATURE $T_a$ (°C)				
I	18	26	38		
II	87	113	123	131	138
III	177	200	214	233	240

The error in oven temperature during post-annealings was estimated to be  $\pm 1^\circ\text{C}$ .

Figure 1 to 3 and Figures 6 to 9 show isothermal decay curves and peak displacements for peaks II, and III, as functions of annealing time  $t_a$ .

**b) The 280°C and 370°C peaks in LiF:Mg**

Peaks occurring at 280°C and 370°C in the glow curve of dosimetric LiF:Mg, known as TLD-100 and produced by Harshaw Chemical Company, were considered in this experiment. Samples annealed for one hour at 400°C were exposed to 30,000 R of Cobalto-60 gamma-rays\*, and subsequently divided into several groups for isothermal post-annealing at temperatures listed in Table II, for times  $t_a$  varying between 5 and 120 minutes.

TABLE II

PEAK	POST-ANNEALING TEMPERATURE $T_a$ (°C)			
280°C	210	223	235	249
370°C	290	298	305	320

The results for  $\log I(T_p)$  vs.  $t_a$  are presented in Fig. 11 for the 280°C peak and in Fig. 13 for the 370°C peak. Figures 12 and 14 show the displacement of the peak position as a function of  $t_a$  for the 280°C and 370°C peaks, respectively.

**Two peaks model**

Let us assume that there are two sets of traps giving rise to two closely spaced, unresolved peaks, which add to give the observed glow peak. Let us denote by  $E_1$  and  $E_2$  their depths and by  $s$  their common frequency factor. The probability of liberating an electron being given by Eq.(1), and assuming that the rate of emptying each trap is not affected by the presence of other one, we can write the following expression for the emitted light intensity:

$$I(T) = n'_0 s \exp(-A_1) + n''_0 s \exp(-A_2) \quad (6)$$

where

$$A_i = s t_a \exp(-E_i/kT_a) + E_i/kT + s \int_{T_0}^T \frac{\exp(-E_j/kT)}{\beta(T)} dT \quad (7)$$

$$i = 1, 2$$

\* The cobalto source we used belongs to Instituto Central de Câncer and Hospital A.C.Camargo - São Paulo.

. 4 .

Schulman et al.<sup>4</sup> considered a superposition of several closely spaced components to explain anomalous fading of CaF<sub>2</sub>:Mn. No quantitative discussion was reported though.

#### Continuous Model

Saddy<sup>5</sup>, Curie<sup>6</sup>, and Medlin<sup>7</sup> have discussed a model that assumes a Gaussian distribution of trap depth<sup>5</sup> around a value E<sub>0</sub> to explain decay curves in the phosphorescence of ZnS and calcite.

Let n(E, t<sub>a</sub>) denote the number of trapped electrons per unit interval of trap depth after time t<sub>a</sub> of isothermal post-annealing at temperature T<sub>a</sub>:

$$n(E, t_a) = \frac{dN(E, t_a)}{dE} \quad (8)$$

We now assume that

$$\frac{d}{dt} (dN) = -dN s \exp(-E/kT) \quad (9)$$

such that

$$dN = dN_0 \exp [ -st_a \exp(-E/kT_a) ] \quad (10)$$

Integrating over trap depth E we obtain

$$N(E, t_a) = \int n(E, 0) \exp [ -st_a \exp(-E/kT_a) ] dE \quad (11)$$

for the actual number of unemptied traps after time t<sub>a</sub>, where n(E, 0) is the distribution function of trap energies at t<sub>a</sub>=0.

Let us take a Gaussian form for n(E, 0) with half-width σ:

$$n(E, 0) = \frac{N_0}{\sqrt{2\pi}\sigma} \exp \left[ -\frac{(E-E_0)^2}{2\sigma^2} \right] \quad (12)$$

where N<sub>0</sub> is the initial density of filled traps. Thus the glow curve can be expressed as

$$I(T) = \frac{N_0 s}{\sqrt{2\pi}\sigma} \int_{E_1}^{E_2} \exp \left[ -\frac{(E-E_0)^2}{2\sigma^2} - st_a \exp(-E/kT_a) - \frac{E}{kT} - s \int_{T_0}^T \frac{\exp(-E/kT)}{\beta(T)} dT \right] dE \quad (13)$$

where  $E_1$  and  $E_2$  are values of  $E$  for which the integrand becomes negligible. If we denote by  $F(E, t_a, T_a, T)$  the integrand of Eq.(13) and let

$$G(E, t_a, T_a, T_p) = \left[ \frac{E}{k T_p^2} - \frac{s}{\beta(T_p)} \exp(-E/kT_p) \right] F(E, t_a, T_a, T_p) \quad (14)$$

where  $T_p$  is the peak temperature, the condition that  $I(T)$  is maximum at  $T=T_p$  can be expressed as

$$\int_{E_1}^{E_2} G(E, t_a, T_a, T_p) dE = 0 \quad (15)$$

The slope of  $\log I(T_p)$  at  $t_a=0$ , which will be denoted by  $m$ , is obtained expanding  $\log I(T_p)$  in a power series of  $t_a$ ;  $m$  is the coefficient of the linear term. So  $m$  can be expressed as

$$m = - \int_{E_1}^{E_2} H(E, 0, T_a, T_p) dE / \int_{E_1}^{E_2} F(E, 0, T_a, T_p) dE \quad (16)$$

where  $H(E, 0, T_a, T_p) = s \exp(-E/kT_a) F(E, 0, T_a, T_p)$ .

## NUMERICAL CALCULATIONS

### a) Two peaks model for natural CaF<sub>2</sub>

We set  $n'_0 = n''_0$  in Eq.(6) to simplify the computation. Initial values of  $s$ ,  $E_1$  and  $E_2$  were obtained solving Eq.(5) for  $s$  and  $E$  using decay data.  $E_1$  and  $E_2$  were then varied, and for each pair of  $E_1$  and  $E_2$ ,  $s$  was varied. Once a set of parameters which produces a glow curve and decay curve close to the observed ones was found, the numerical calculations were carried out in finer steps around that set of parameters. Starting with peak III and  $T_a=200^\circ\text{C}$  the best result obtained was for  $E_1=1.53$  eV,  $E_2=1.71$  eV and  $s=2 \times 10^{11} \text{ sec}^{-1}$ . These values were used to obtain decay curves for  $T_a=214, 233, 244^\circ\text{C}$ . The results of these calculations are plotted in Figures 6 to 10. Practically no displacement of peak position resulted.

A similar procedure of computation was applied to peak II. The results are shown in Figures 1 to 4. Best fits were found for  $E_1=0.94$  eV,  $E_2=0.96$  eV and  $s=2 \times 10^8 \text{ sec}^{-1}$ . No displacement of peak position was obtained.

### b) Continuous model for natural CaF<sub>2</sub>

Again the analysis was carried out for peak III and  $T_a=200^\circ\text{C}$ , varying  $E_0$  between 1.50 and 1.60 eV in a step of 0.02 eV. For each value of  $E_0$ ,  $s$  was varied from  $10^{11} \text{ sec}^{-1}$  to  $10^{12} \text{ sec}^{-1}$  in steps of  $2 \times 10^{11} \text{ sec}^{-1}$  while  $\sigma$  was kept at 0.1 eV. This analysis led to  $E_0=1.58$  eV and  $s=5 \times 10^{11} \text{ sec}^{-1}$  as best parameters. These values were then held fixed and  $\sigma$  varied, resulting in  $\sigma=0.09$  eV as the best value. This set of parameters was subsequently used to fit experimental data for different values of  $T_a$ . Similar calculations were carried out for peak II, and the results are represented in Figures 1 to 11 (except Figs. 4 and 10). For this model there was a clearcut displacement of peak position.

### c) Continuous model for 280 and 370°C peak of LiF:Mg

Following the same procedure as described in Section b, we searched for a set of parameters to fit glow curves and decay curves for the 280°C peak starting with  $T_a=223^\circ\text{C}$ :  $E_0=1.50\text{ eV}$ ,  $s=5 \times 10^{11}\text{ sec}^{-1}$ , and  $\sigma=0.09\text{ eV}$ . These values were used to compute decay data and displacement of peak position for  $T_a=210^\circ\text{C}$ , 235, and 249°C. The results are plotted in Figures 12 and 13.

For the 370°C peak the search for parameters was carried out for  $T_a=290^\circ\text{C}$  data, obtaining  $E_0=1.66\text{ eV}$ ,  $s=8 \times 10^{11}\text{ sec}$  and  $\sigma=0.09\text{ eV}$ . The results of calculation for  $T_a=298$ , 305, and 320°C are shown in Figures 14 and 15.

Since there is a slight possibility that the variation of oven temperature during the operation of inserting the sample for annealing might influence short-time decay data, we performed isothermal post-annealing measurements at 118, 132, and 140°C for peak 5 of LiF:Mg. This peak was chosen because its  $\sigma$  is very close to zero, so that its  $\log I(T_p)$  is linear in  $t_a$ , as shown in Fig.16. The experimental result agrees with this statement. Since the same oven was used in all the experiments of the present work, the curvatures in decay data are crystal's properties.

## CONCLUSIONS

Although extensive calculations were performed, a more complete numerical analysis can and will be carried out, especially for the 280°C and 370°C peaks in TLD-100. Nonetheless, the following conclusions can be drawn:

1. The two peaks model predicts too small a displacement of peak position found in isothermal decay data in fluorite and in LiF:Mg, although decay curves and glow curves can be fitted reasonably well.
2. The continuous model predicts the displacement observed experimentally in the TL peaks of fluorite. In TLD-100, it predicts well enough the displacement of the 280°C peak, but in the case of the 370°C peak, the model gives only qualitative agreement.

In the two peaks model one might try  $n'_0 \neq n''_0$ , but then we will be introducing one more parameter to be adjusted. The situations would be similarly complicated if we assume three or more peaks model. Furthermore, it is hard to understand why two peaks would explain every case that does not follow the Randall and Wilkins model.

The detailed study of the behavior of functions, F, G, H and m will be discussed elsewhere.

## ACKNOWLEDGEMENT

We would like to acknowledge Dra. Emico Okuno for taking data on natural calcium fluoride; Miss Rejane A. Nogueira for taking data on TLD-100; Miss Wanda C. Las for numerical calculations on TLD-100 data. Thanks are due to personnel at the computation section of the physics institute of the University of São Paulo, and also to Dr. Michael R. Mayhugh for discussions.

## RESUMO

A análise das curvas de emissão em  $\text{CaF}_2$  natural mostrou que alguns dos picos de emissão neste material não obedecem ao modelo simples de termoluminescência proposto por Randall e Wilkins. Em particular esses picos não decaem exponencialmente durante um aquecimento isotérmico e, além disso, as

posições dos picos se deslocam durante este aquecimento. Um modelo baseado na superposição de dois picos foi, primeiro, proposto, no qual dois picos pouco espaçados e não resolvidos se superpõem para dar o comportamento observado. Os cálculos revelaram que, este modelo pode explicar razoavelmente bem as curvas de decaimento isotérmico, porém, não a mudança isotérmica da posição do pico. Foi então proposto um segundo modelo, baseado numa distribuição contínua das profundidades das armadilhas, ao invés de um valor bem definido das mesmas, como no de Randall e Wilkins. Usando uma distribuição gaussiana foi feito um ajuste das curvas de emissão dos picos II e III no CaF<sub>2</sub> natural, obtendo-se um resultado satisfatório, também, para o decaimento isotérmico e mudança isotérmica da posição dos picos. A aplicação deste modelo nos permite a determinação dos parâmetros das armadilhas. Análise baseada neste modelo foi estendida aos picos de 280°C e 370°C na curva de emissão do LiF:Mg dosimétrico.

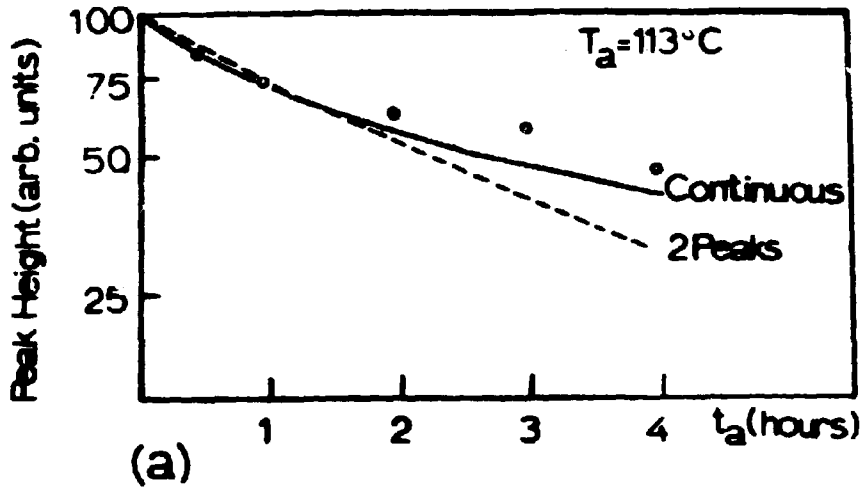
## RÉSUMÉ

L'analyse des courbes d'émission dans le CaF<sub>2</sub> naturel a montré que quelques pics d'émission de ce matériau n'obéissent pas au modèle simple de thermoluminescence proposé par Randall et Wilkins. En particulier ces pics ne décroissent pas exponentiellement pendant un chauffage isothermique et en outre la position des pics se déplace durant ce chauffage. Nous avons proposé un modèle dans lequel deux pics peu séparés se superposent pour donner lieu au pic expérimentalement observé. Les calculs ont montré que ce modèle peut expliquer raisonnablement les courbes de décroissement isothermique, mais pas le changement isothermique de position des pics. Il fut alors proposé un second modèle basé sur une distribution continue de la profondeur des pièges, au lieu d'une valeur bien définie de celle-ci comme Randall et Wilkins l'ont admise. En prenant une distribution de Gauss nous avons calculé théoriquement les courbes d'émission des pics II et III du CaF<sub>2</sub> naturel et nous avons obtenu un résultat satisfaisant aussi bien pour le décroissement isothermique que pour le changement isothermique de la position des pics.

L'application de ce modèle nous permet de calculer les paramètres du pièges. L'analyse basée sur ce modèle fut étendue aux pics de 280°C et de 370°C des courbes d'émission du LiF:Mg dosimétrique.

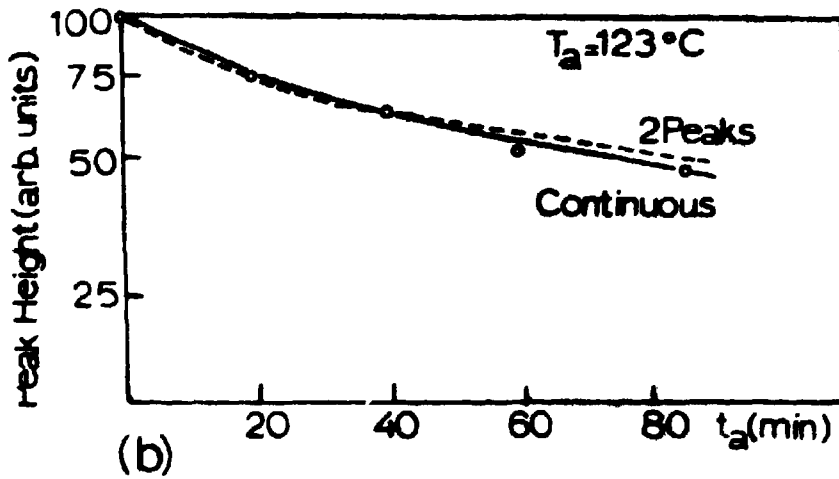
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(a) for  $T_a = 113^\circ\text{C}$ .

Fig. 1 Decay data for peak II in fluorite.



(b) for  $T_a = 123^\circ\text{C}$ .  $E_1 = 0.94$  eV,  $E_2 = 0.96$  eV,  $s = 2 \times 10^8 \text{ sec}^{-1}$ ;  $E_0 = 0.96$  eV,  $s = 2 \times 10^8 \text{ sec}^{-1}$ ,  $\sigma = 0.05$  eV.

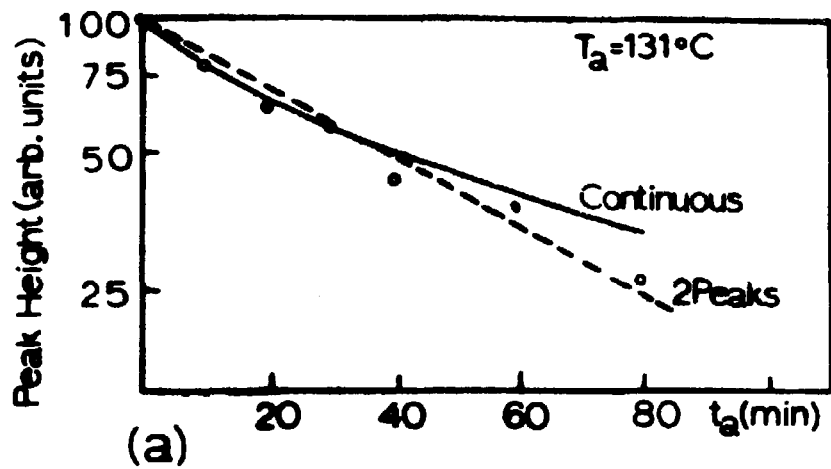


Fig. 2 (a)  
Decay data for peak II in fluorite for  $T_a = 131^\circ\text{C}$ . Same parameters as in Fig. 1 were used.

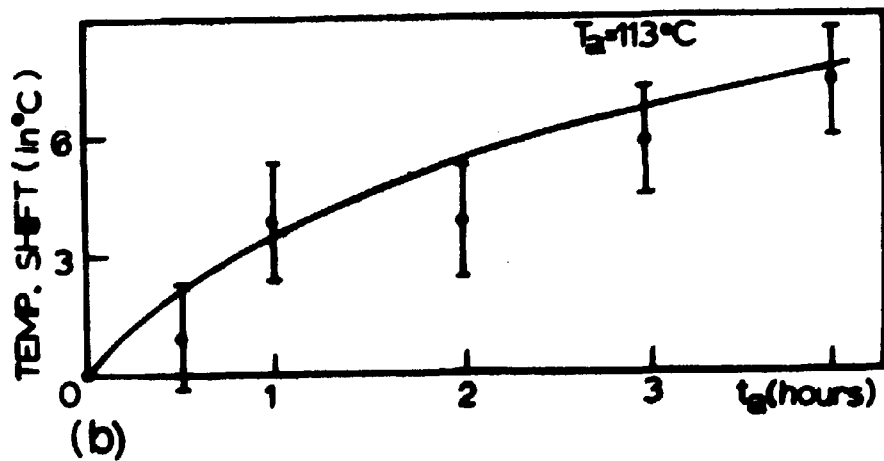
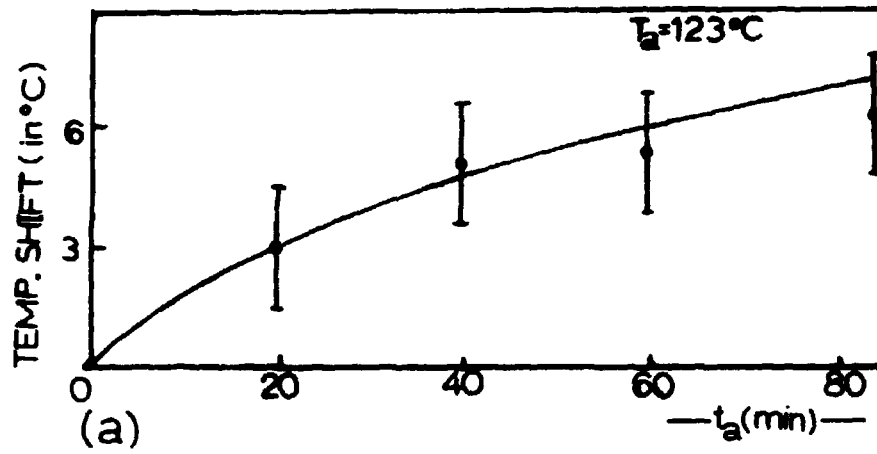
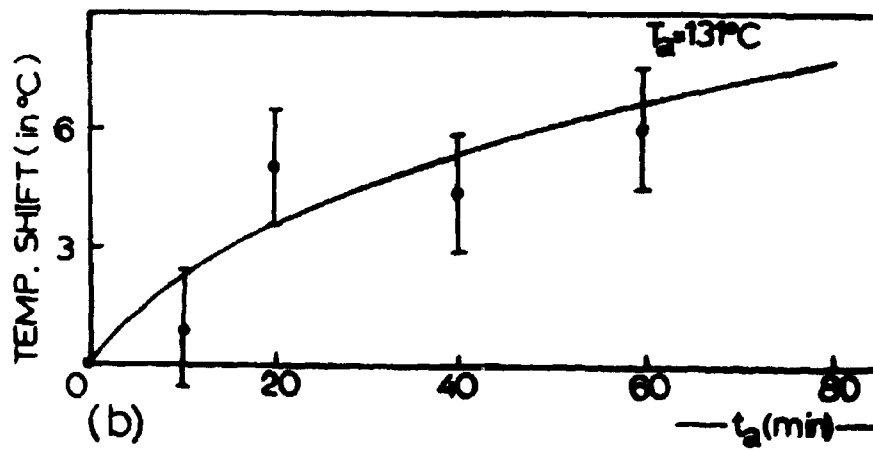


Fig. 2 (b)  
Displacement of peak II in fluorite for  $T_a = 113^\circ\text{C}$ . Solid line: Continuous model calculation.



(a) for  $T_a = 123^\circ\text{C}$ .

Fig. 3 Displacement of peak II in fluorite.



(b) for  $T_a = 131^\circ\text{C}$ . Solid line: Continuous model calculation.

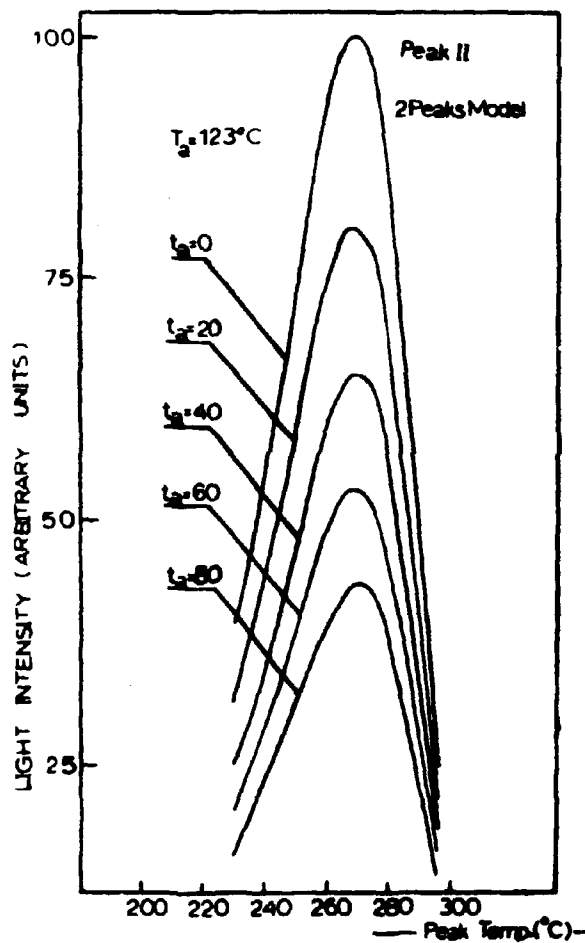
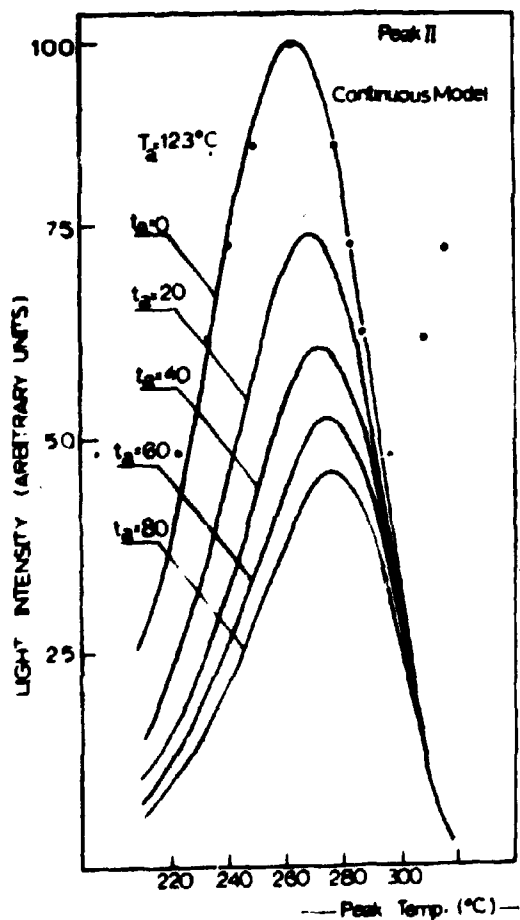
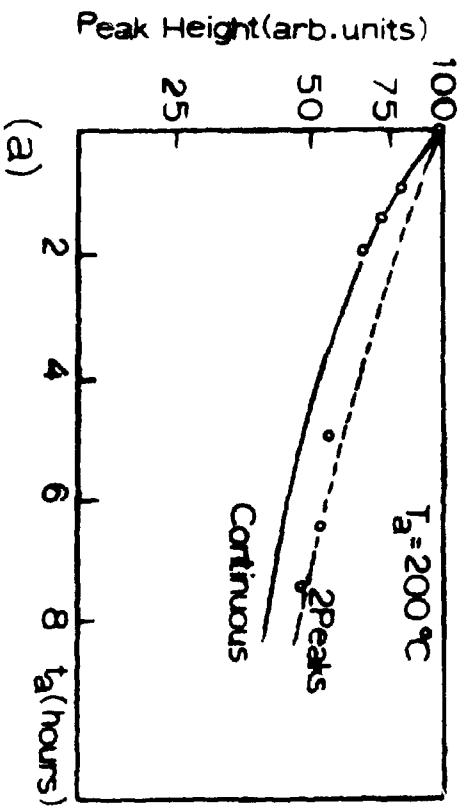


Fig. 4  
Theoretical glow curves for peak II using 2 peaks model.  $E_1 = 0.94$  eV,  $E_2 = 0.96$  eV,  $s = 2 \times 10^8$  sec $^{-1}$ .

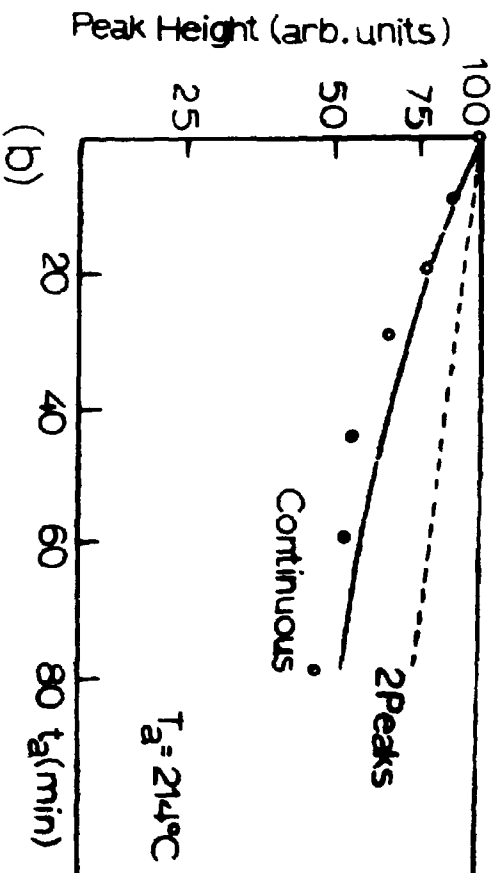
Fig. 5  
Solid lines: Theoretical glow curves for peak II using continuous model.  $E_0 = 0.96$  eV,  $s = 2 \times 10^8$  sec $^{-1}$ ,  $\sigma = 0.05$  eV Circles: Experimental points.



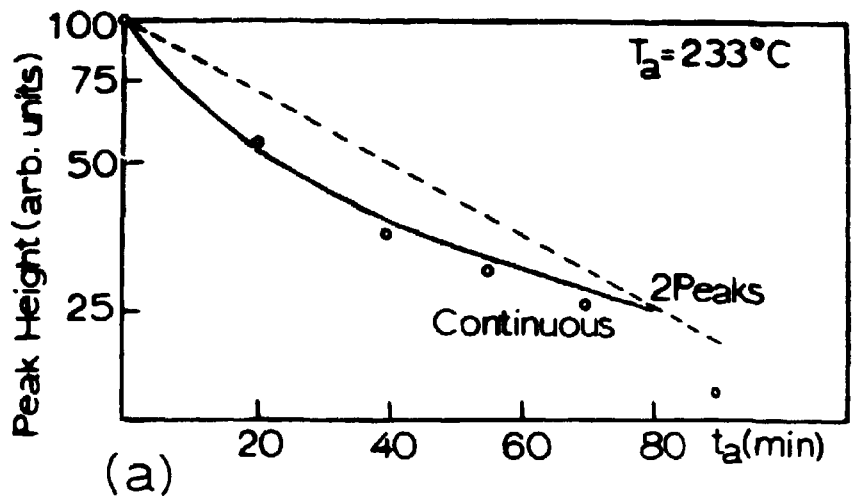


(a) for  $T_g = 200^\circ\text{C}$ .

Fig. 6 Decay for peak III in fluorite.

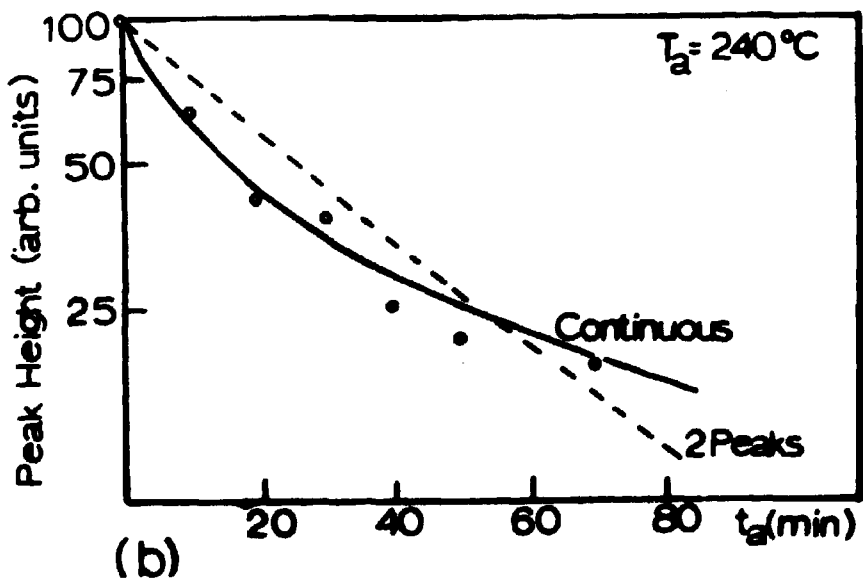


(b) for  $T_g = 214^\circ\text{C}$ .  $E_0 = 1.58 \text{ eV}$ ,  $s = 5 \times 10^{11} \text{ sec}^{-1}$ ,  $\sigma = 0.09 \text{ eV}$ ;  $E_1 = 1.53 \text{ eV}$ ,  $E_2 = 1.71 \text{ eV}$ ,  $s = 5 \times 10^{11} \text{ sec}^{-1}$ .

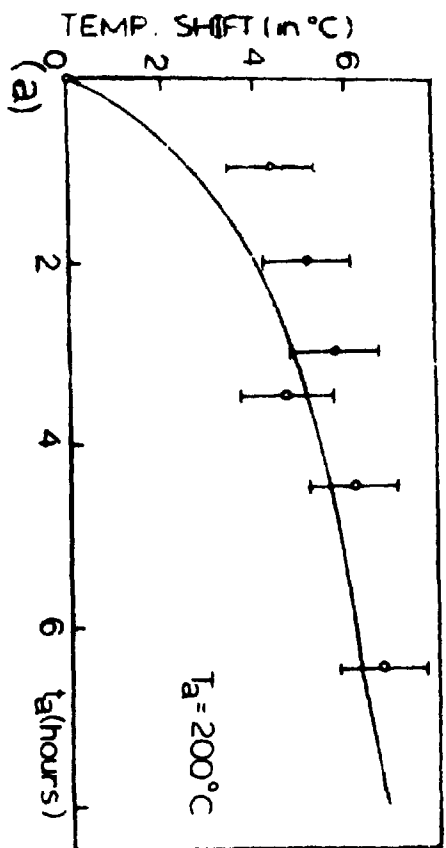


(a) for  $T_a = 233^\circ\text{C}$ .

Fig. 7 Decay data for peak III in fluorite.

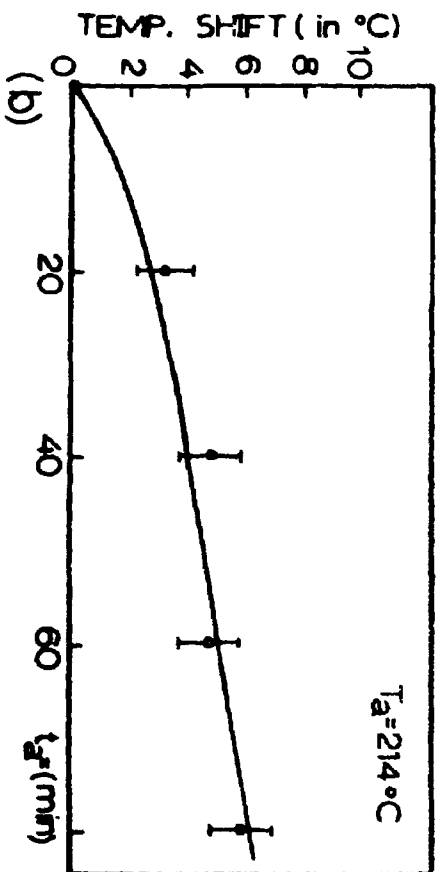


(b) for  $T_a = 240^\circ\text{C}$ . Same parameters as in Fig. 6 were used.



(a) for  $T_a = 200^\circ\text{C}$ .

Fig. 8 Displacement of peak III in fluorite.



(b) for  $T_a = 214^\circ\text{C}$ .

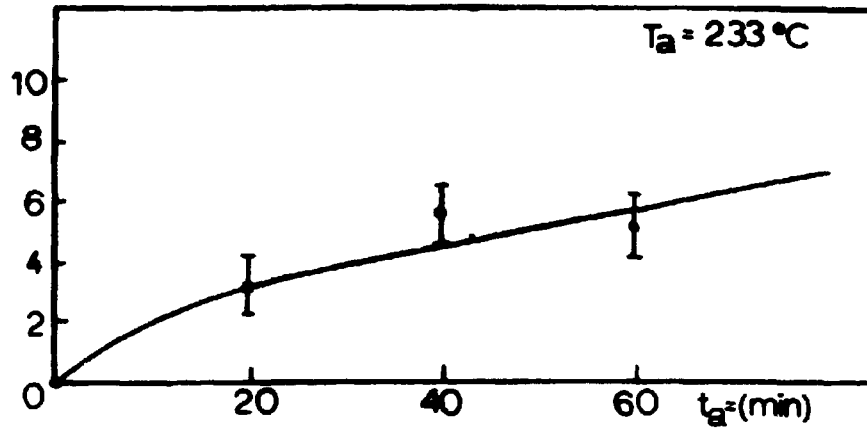


Fig. 9  
Displacement of peak III in fluorite for  $T_a = 233^\circ\text{C}$ .

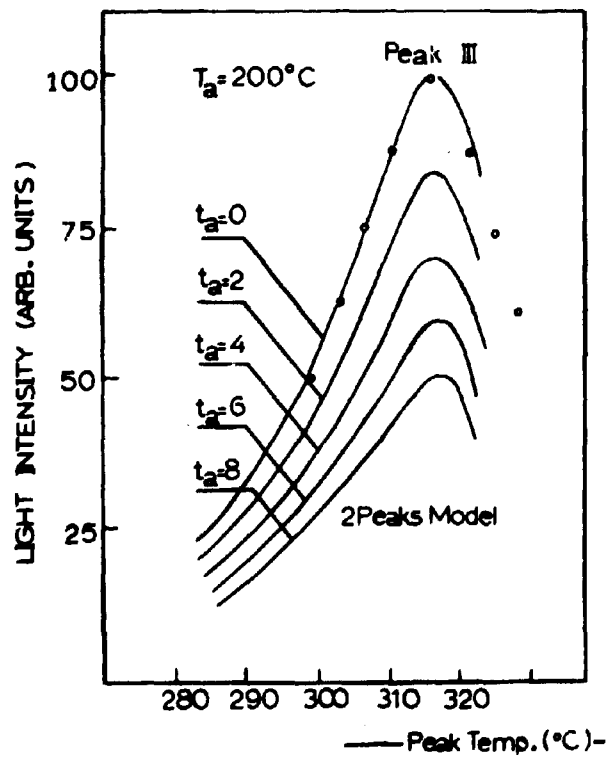


Fig. 10  
Theoretical glow curves for peak III using 2 peaks model  $E_1 = 1.53\text{ eV}$ ,  $E_2 = 1.78\text{ eV}$  and  $s = 2 \times 10^{11}\text{ sec}^{-1}$ .

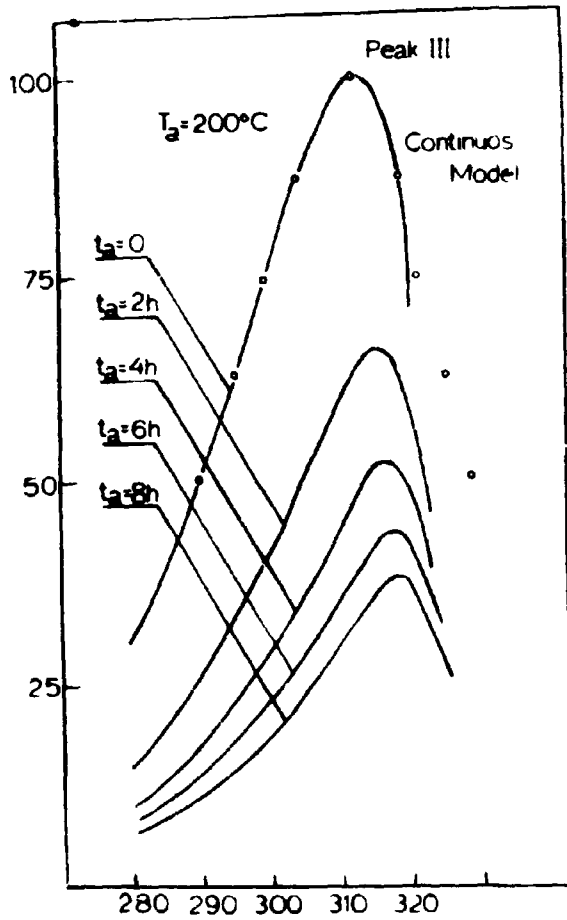


Fig. 11  
 Solid Lines: Theoretical glow curves for peak III using Continuous model.  $E_0 = 1.58\text{ eV}$ ,  $s = 5 \times 10^{11}\text{ sec}^{-1}$ ,  $\sigma = 0.09\text{ eV}$ .  
 Circles: Experimental points.

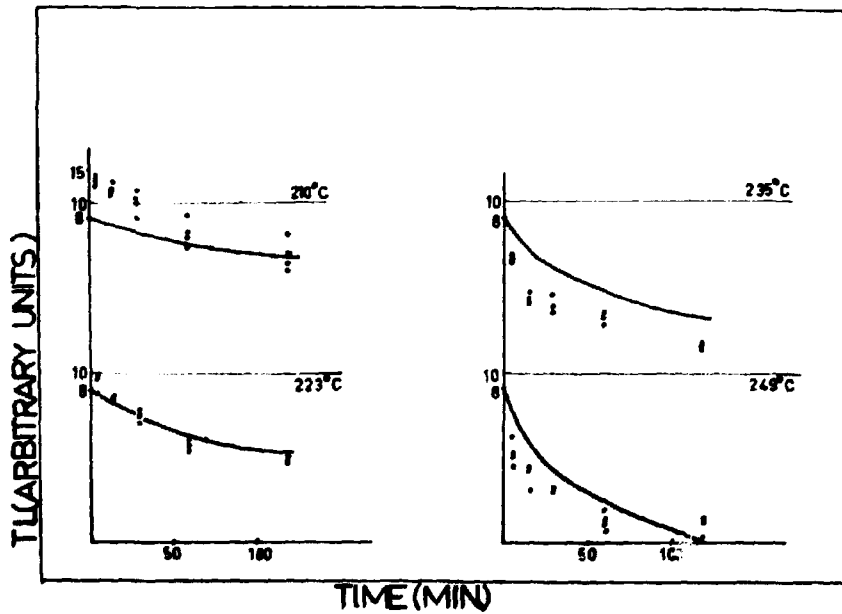


Fig. 12  
 Decay data for 280°C peak in TLD-100. Solid Lines: Continuous model calculation for  $E_0 = 1.50\text{ eV}$ ,  $s = 5 \times 10^{11}\text{ sec}^{-1}$ ,  $\sigma = 0.09\text{ eV}$ .

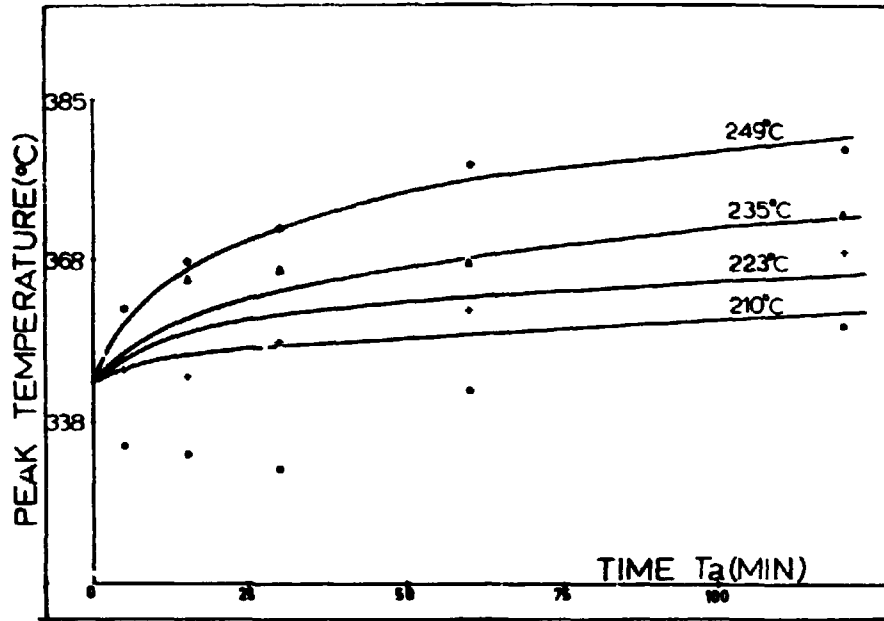


Fig. 13  
Displacement of 280°C peak in TLD-100. Solid Lines: Continuous model calculation for same parameters listed in Fig. 12.

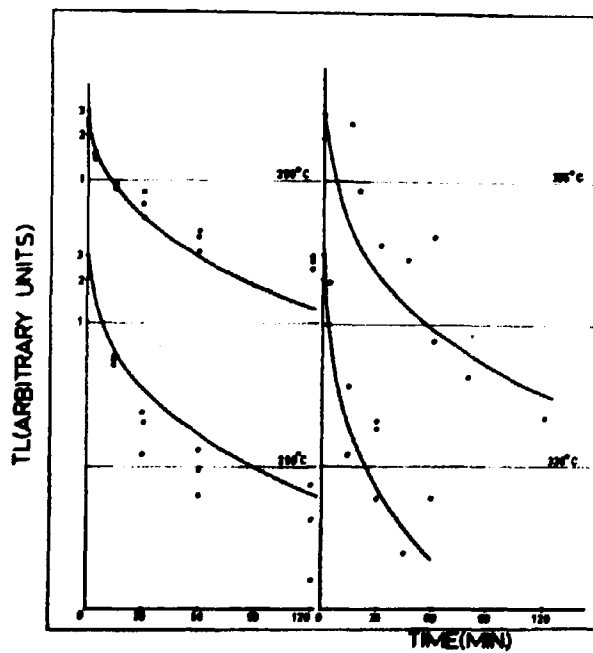


Fig. 14  
Decay data for 370°C in TLD-100. Solid lines: Continuous model calculation for  $E_0 = 1.66$  eV,  $s = 8 \times 10^{11} \text{ sec}^{-1}$ ,  $\sigma = 0.09$  eV.

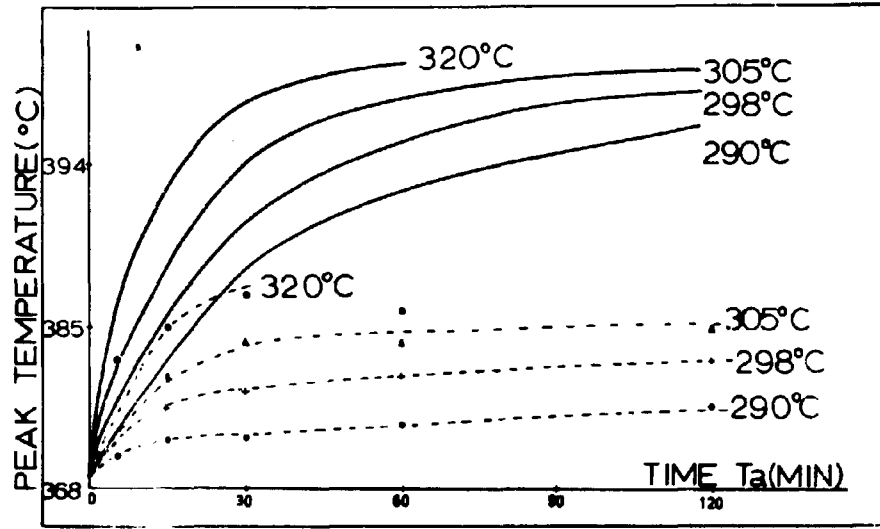


Fig. 15  
Displacement of 370°C peak in TLD-100. Solid Lines: Continuous model calculation for same parameters as in Fig. 14.

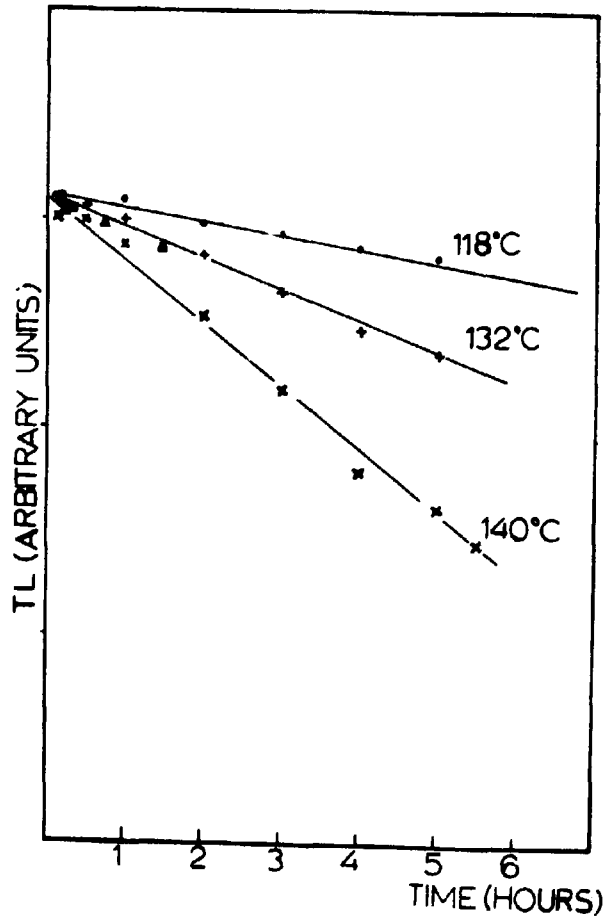


Fig. 16  
Experimental decay data for peak 5 in TLD-100.