Interactive and non interactive multi-trap models in thermoluminescence: Closed expressions derived without approximations for analyzing glow curves

F. Ortega[a], P. Molina[b], J. Marcazzó[b], M. Santiago[b], M. Lester[b], E. Caselli[b],*

[a] *Facultad de Ingeniería - CIFICEN (CONICET – UNCPBA), Av. del Valle 5737, 7400 Olavarría, Argentina*
[b] *Instituto de Física Arroyo Seco - CIFICEN (CONICET – UNCPBA), Pinto 399, 7000 Tandil, Argentina*

A B S T R A C T

The kinetics usually employed for analyzing glow curves are either heuristic or derived from physical models resorting to approximations. In this article closed expressions for the interactive and non interactive multi-trap models are derived without approximations, which render the analysis of glow curves more reliable.

1. Introduction

Glow curve analysis is a frequently used procedure for investigating the kinetics involved in thermoluminescence (TL). Basically it relies on choosing a model, which is in accordance with experimental results at hand, and on deriving a theoretical expression for the emitted light $I_{th}(T,\alpha)$ from the set of differential equations describing the carrier traffic among traps and recombination centers. α stands for the set of parameters characterizing traps and recombination centers and T for the absolute temperature.

A reliable set of values for α can be found by best-fitting the theoretical expression $I(T,\alpha)$ to the set of experimental values $I_{exp}(T_s)$, namely, the light intensity measured at each sampling temperature T_s. The fitting is usually performed by minimizing the expression:

$$S(\alpha) = \sum_{s=1}^{Z} [I_{th}(T_s, \alpha) - I_{exp}(T_s)]^2$$

where Z is the number of sampling temperatures [1].

The most employed algorithm for finding the minimum of $S(\alpha)$ is the Levenberg-Marquardt algorithm (L-M) [1,2]. For the L-M algorithm to be employed a closed expression of the theoretical light intensity must be available, which is iteratively evaluated during the execution of the fitting routine.

The simplest model for TL, known as one trap-one recombination center (OTOR), is shown in Fig. 1.

For the model shown in Fig. 1 the set of parameters is $\alpha = [E, s, n_0, A_h, A_n]$, where n_0 stands for the initial concentration of trapped electrons. The initial concentration of holes h_0 is equal to the

initial concentration of trapped electrons n_0 according to Eq. (3) below because the initial concentration of electrons in the conduction $n_{c,0}$ band is zero.

If a sample is heated with a constant heating rate β, namely, $T(t) = T_0 + \beta t$, the equations describing the traffic of electrons are:

$$\frac{dn(T)}{dT} = -\frac{s}{\beta}n(T)\exp(-\frac{E}{kT}) + \frac{A_n}{\beta}(N - n(T))n_c(T) \quad (1)$$

$$\frac{dh(T)}{dT} = -\frac{A_h}{\beta}n_c(T)h(T) \quad (2)$$

$$h(T) = n(T) + n_c(T) \quad (3)$$

$$I(T) = A_h n_c(T)h(T) \quad (4)$$

Eq. (3) represents charge neutrality and $I(T)$ stands for the emitted light.

The first analytical expression for $I(T)$ has been put forward by Randall and Wilkins [3]. It is known as first order (FO) kinetics. It is derived from the set of coupled differential equations by resorting first to the quasi-equilibrium (QE) approximation. The QE approximation assumes that $\frac{dn_c}{dt} \cong 0$ and $n_c \ll n$. As shown in reference [2], the light intensity is given by:

$$I(T) = \frac{n(T) s \exp\left(\frac{E}{kT}\right)h(T)A_h}{(N - n(T))A_n + h(T)A_h} \quad (5)$$

FO kinetics results if it is assumed that retrapping is negligible against recombination, i. e., $(N - n(T))A_n \ll hA_h$,:
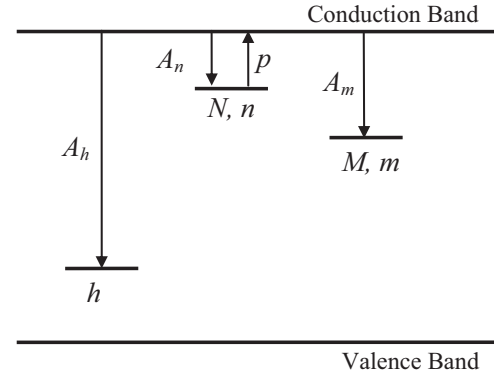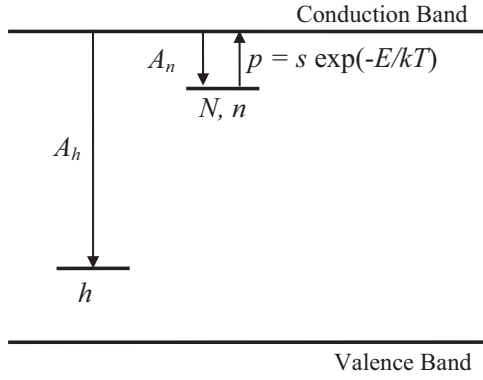

Fig. 1. OTOR model. A_h is the recombination probability, h is the concentration of holes in the recombination centers, A_n is the retrapping probability, N is the concentration of traps, n is the concentration of trapped electrons, s is the frequency factor, E the activation energy, and k is the Boltzmann constant. The product $s \exp(-E/kT)$ is the escape probability of an electron from a trap.

$$I(T) = n(T)\, s\, \exp\left(-\frac{E}{kT}\right) \tag{5-a}$$

Since the QE approximation entails that $n_c(T) \approx 0$ it results $h(T) = n(T)$. Taking into account that $I(T) = -\frac{dh(T)}{dT} = -\frac{dn(T)}{dT}$, this equation along with Eq. (5-a) yield:

$$I(T) = n_0 s \exp\left(\frac{-E}{kT}\right) \exp\left(-\frac{s}{\beta} \int_{T_0}^{T} \exp\left(\frac{E}{ku}\right) du\right) \tag{6}$$

where T_0 stands for the temperature at which the recording of a glow curve starts.

On the contrary, if retrapping prevails on recombination, i. e., $(N - n(T))A_n \gg hA_m$, the following equation results:

$$I(T) = s\left(\frac{A_n}{NA_h}\right) n^2 \exp\left(\frac{-E}{kT}\right) \tag{7}$$

From this equation one can obtain:

$$I(T) = n_0{}^2 s' \exp\left(-\frac{E}{kT}\right) \left[1 + \frac{n_0 s'}{\beta} T \int_{T_0}^{T} \exp\left(-\frac{E}{ku}\right) du\right]^2 \tag{8}$$

In Eq. (8) s' stands for $\frac{A_n}{NA_h}$. Garlick and Gibson put forward this kinetics, called second order (SO) kinetics [4].

Since a closed expression cannot be derived when recombination does not prevail on retrapping, or retrapping on recombination, May and Partridge put forward a heuristic expression with the aim of describing kinetics comprised between first and second order [5]:

$$I(T) = n0 s'' \exp\left(-\frac{E}{kT}\right) \left[1 + (b-1)\frac{s''}{\beta} \int_{T_0}^{T} \exp\left(-\frac{E}{ku}\right) du\right]^{\frac{-b}{b-1}} \tag{9}$$

In this equation s'' stands for $s.\, n0^{b-1}$, and b is a parameter loosely related to the retrapping probability. When $b \to 1$ Eq. (9) converges to FO kinetics, and when $b = 2$, to SO kinetics. This kinetics is known as the general order (GO) kinetics, and it is nowadays the most employed kinetics for analyzing glow curves.

Several authors have investigated the validity of the GO kinetics. According to the reported results the GO kinetics suffers from several flaws:

1) Investigations have been carried out to find a connection between **b** and physically meaningful models, but a clear relationship could not been established [6,7].
2) Moharil and Opanowicz found that the kinetic order b is usually not constant during thermal stimulation and should not be used for characterization of thermoluminescence [8,9].
3) It has been reported that the GO model has limitations for

determination of the activation energy [10].
4) By resorting to computer simulation Sakurai has shown that the GO kinetics can yield wrong parameters [11]. Furthermore he states that one of the defects of the GO kinetics arises from the fact that traffic of electrons (or holes) among traps is ignored [12].
5) Basun et al. have shown that the interaction among traps affects the shape of glow curves, thus affecting the parameters given by the GO kinetics [13].

2. Derivation of the light intensities for the IMTS and NMTS models

A more general model is that depicted in Fig. 2. It is known as the interactive multitrap system (IMTS). This model includes a deep trap (also known as thermally disconnected trap). This trap can capture electrons but cannot release electrons for the temperature range the glow curve is recorded. If the deep trap is fully occupied the model is called Non-interactive multitrap system (NMTS).

The equations describing the carrier traffic are:

$$\frac{dn(t)}{dt} = -n(t)\, s\, \exp\left(-\frac{E}{kT(t)}\right) + A_n[N - n(t)]n_c(t) \tag{10}$$

$$\frac{dm(t)}{dt} = A_m[M - m(t)]n_c \tag{11}$$

$$\frac{dh(t)}{dt} = -A_h n_c(t) h(t) \tag{12}$$

$$h(t) = n(t) + m(t) + n_c(t) \tag{13}$$

$$I(t) = -\frac{dh(t)}{dt} \tag{14}$$

A_m is the trapping probability at a deep trap, m is the concentration of occupied deep traps, and M is the concentration of deep traps.

Eq. (14) is the thermoluminescent light, and Eq. (13) represents charge conservation. If a probe is heated with a constant heating rate β, namely, $T(t) = T_0 + \beta t$, then Eqs. (10)–(14) read:

$$\frac{dn(T)}{dT} = -n(T)\frac{s}{\beta}\exp\left(-\frac{E}{kT}\right) + \frac{A_n}{\beta}[N - n(T)]n_c(T) \tag{15}$$

$$\frac{dm(T)}{dT} = \frac{A_m}{\beta}[M - m(T)]n_c(T) \tag{16}$$

$$\frac{dh(T)}{dT} = -\frac{A_h}{\beta}n_c(T)h(T) \tag{17}$$

$$h(T) = n(T) + m(T) + n_c(T) \tag{18}$$

$$I(T) = -\beta\frac{dh(T)}{dT} \tag{19}$$

Fig. 2. IMTS and NMTS are an extension of the model shown in Fig. 1 by adding a deep trap having concentration M.

Integration of Eq. (19) yields:

$$h(T) = C - \frac{1}{\beta} \int_{T_0}^{T} I(u)du \qquad (20)$$

For $T = T_0$ Eq. (18) reads:

$$h(T_0) = n_0 + m_0 = C \qquad (21)$$

where m_0 stands for the initial concentration of occupied deep traps.

Then Eq. (20) becomes:

$$h(T) = n_0 + m_0 - \frac{1}{\beta} \int_{T_0}^{T} I(u)du \qquad (22)$$

Eq. (15) can be rearranged as follows:

$$\frac{dn(T)}{dT} + \left[\frac{s}{\beta} \exp\left(-\frac{E}{kT}\right) + \frac{A_n}{\beta} n_c(T) \right] n(T) = \frac{A_n}{\beta} N n_c(T) \qquad (23)$$

From Eqs. (17) and (22) the following relationship can be derived:

$$n_c(T) = \frac{I(T)}{A_h \left[n_0 + m_0 - \frac{1}{\beta} \int_{T_0}^{T} I(u)du \right]} \qquad (24)$$

By inserting this relationship into Eq. (23) it results:

$$\frac{dn(T)}{dT} + \left[\frac{s}{\beta} exp\left(-\frac{E}{kT}\right) + \frac{A_n}{\beta} \frac{I(T)}{A_h \left[n_0 + m_0 - \frac{F(T)}{\beta} \right]} \right] n(t)$$
$$= \frac{A_n}{\beta} N \frac{I(T)}{A_h \left[n_0 + m_0 - \frac{F(T)}{\beta} \right]} \qquad (25)$$

where $F(T)$ stands for $F(t) = \int_{T_0}^{T} I(u)du$.

By defining $G(T)$ as follows:

$$G(T) = \frac{s}{\beta} \exp\left(-\frac{E}{kT}\right) + \frac{R_n}{\beta} \frac{I(T)}{\left[n_0 + m_0 - \frac{F(T)}{\beta} \right]} \qquad (26)$$

where $R_n = \frac{A_n}{A_h}$, then Eq. (25) becomes:

$$\frac{dn(T)}{dT} + G(T)n(t) = \frac{R_n}{\beta} N \frac{I(T)}{\left[n_0 + m_0 - \frac{F(T)}{\beta} \right]} \qquad (27)$$

If $U(T)$ is defined as:

$$U(T) = \exp\left(\int_{T_0}^{T} G(u)du \right) \qquad (28)$$

and multiplied by both sides of Eq. (27) the derivation of the following relationship is straightforward:

$$\frac{d[U(T)n(T)]}{dT} = \frac{U(T)R_n}{\beta} N \frac{I(T)}{n_0 + m_0 - \frac{F(T)}{\beta}} \qquad (29)$$

Integration of Eq. (29) yields:

$$U(T)n(T) - n_0 = \frac{R_n N}{\beta} \int_{T0}^{T} U(u) \frac{I(u)}{n_0 + m_0 - \frac{F(u)}{\beta}} du \qquad (30)$$

from which the following relationship results:

$$n(T) = \frac{n_0}{U(T)} + \frac{R_n N}{\beta U(T)} \int_{T_0}^{T} U(u) \frac{I(u)}{n_0 + m_0 - \frac{F(u)}{\beta}} du \qquad (31)$$

In a similar fashion, an expression for $m(T)$ can be found by defining the following expressions:

$$P(T) = \frac{A_m}{\beta} \frac{I(T)}{A_h \left[n_0 + m_0 - \frac{F(T)}{\beta} \right]} \qquad (32)$$

and:

$$V(T) = \exp\left(\int_{T_0}^{T} P(u)du \right) \qquad (33)$$

By resorting to Eq. (16), Eq. (32) and Eq. (33) it results:

$$m(T) = \frac{m0}{V(T)} + \frac{R_m M}{\beta V(T)} \int_{T_0}^{T} V(u) \frac{I(u)}{n_0 + m_0 - \frac{F(u)}{\beta}} du \qquad (34)$$

Now we can find an expression for the thermoluminescence intensity. Indeed, from Eqs. (17) and (19) the following relationship results:

$$I(T) = A_h n_c(T)h(T) \qquad (35)$$

If we take into account that $n_c(T) = h(T) - n(T) - m(T)$ and $h(T) = n_0 + m_0 - \frac{1}{\beta}F(T)$, it results:

$$I(T, \alpha) = A_h \left(n_0 + m_0 - \frac{1}{\beta}F(T) - n(T) - m(T) \right) \left(n_0 + m_0 - \frac{1}{\beta}F(T) \right) \qquad (36)$$

Where, as before, $F(T) = \int_{T_0}^{T} I(u)du$, and $n(T)$ and $m(T)$ are given by Eqs. (31) and (34) respectively. In this case, the set of parameters characterizing the model is:

$$\alpha_{IMTS} = [A_h, A_n, A_m, N, n_0, M, m_0, s, E].$$

For the NMTS model the deep traps are fully occupied, namely, $m = m_0 = M$. The equations describing the model are:

$$\frac{dn(T)}{dT} = -n(T)\frac{s}{\beta} \exp\left(-\frac{E}{kT}\right) + \frac{A_n}{\beta}[N - n(T)]n_c(T) \qquad (37)$$

$$\frac{dh(T)}{dT} = -\frac{A_h}{\beta} n_c(T)h(T) \qquad (38)$$

$$h(T) = n(T) + M + n_c(T) \qquad (39)$$

$$I(T) = -\frac{dh(T)}{dT} \qquad (40)$$

Following a similar procedure as that employed for deriving Eq. (36), it is easy to show that the light intensity $I(T, \alpha)$ is given by:

$$I(T, \alpha) = A_h \left(n_0 - \frac{1}{\beta}F(T) - n(T) \right) \left(n_0 + M - \frac{1}{\beta}F(T) \right) \qquad (41)$$

As before, $F(T) = \int_{T_0}^{T} I(u)du$, and $n(T)$ is given by:

$$n(T) = \frac{n_0}{W(T)} + \frac{R_n N}{\beta} \int_{T_0}^{T} \frac{W(u)}{W(T)} \frac{I(u)}{\left(n_0 + M - \frac{F(u)}{\beta} \right)} du \qquad (42)$$

where:

$$W(T) = \exp\left(\int_{T_0}^{T} X(u)du \right) \qquad (43)$$

and:

$$X(T) = \frac{s}{\beta} \exp\left(-\frac{E}{kT}\right) + \frac{R_n}{\beta} \frac{I(T)}{\left(n_0 + M - \frac{F(T)}{\beta} \right)} \qquad (44)$$

In this case, the set of parameters characterizing the model is:

$$\alpha_{NMTS} = [A_h, A_n, N, n_0, M, s, E]$$

As aforementioned most materials show more than one trap. For two or more traps Eq. (24) reads:

$$n_c(T) = \frac{I(T)}{A_h \left[\sum_{i=1}^{k} n_{i,0} + m_0 - \frac{1}{\beta} \int_{T_0}^{T} I(u)du \right]} \qquad (45)$$

And Eq. (36) turns into:

$$I(T, \alpha) = A_h \left(\sum_{i=1}^{k} n_{0,i} + \sum_{i=1}^{l} m_{0,i} - \frac{1}{\beta} F(T) - \sum_{i=1}^{k} n_i(T) \right.$$
$$\left. - \sum_{i=1}^{l} m_i(T) \right) \times$$
$$\times \left(\sum_{i=1}^{k} n_{0,i} + \sum_{i=1}^{l} m_{0,i} - \frac{1}{\beta} F(T) \right) \tag{46}$$

Following the same steps for deriving Eq. (42) we obtained:

$$n(T) \frac{n_0}{W(T)} + \frac{R_n N}{\beta} \int_{T_0}^{T} \frac{W(u)}{W(T)} \frac{I(u)}{\left(\sum_i n_{i,0} + M - \frac{F(u)}{\beta} \right)} du \tag{47}$$

and

$$X_i(T) = \frac{s_i}{\beta} \exp\left(-\frac{E_i}{kT}\right) + \frac{R_i}{\beta} \frac{I(T)}{\left(\sum_i n_{i,0} + M - \frac{F(T)}{\beta} \right)} \tag{48}$$

In Eq. (46) it is supposed that there are k traps and l deep traps. For the NMTS model Eq. (46) becomes:

$$I(T, \alpha) = A_h \left(N_0 - \frac{1}{\beta} F(T) - \sum_{i=1}^{k} n_i(T) \right) \left(N_0 + M - \frac{1}{\beta} F(T) \right) \tag{49}$$

where:

$$N_0 = \sum_{i=1}^{k} n_{0,i}; \quad M = \sum_{i=1}^{l} M_{0,i} \tag{50}$$

In this case, the set of parameters is given by:

$$\alpha = [A_h, A_{n,i}, N_i, n_{0,i}, M, s_i, E_i]$$

In Eq. (49) $n_i(T)$ is given by:

$$n_i(T) = \frac{n_{i,0}}{W_i(T)} + \frac{R_{n,i} N_i}{\beta} \int_{T_0}^{T} \frac{W_i(u)}{W_i(T)} \frac{I(u)}{\left(\sum_i n_{i,0} + M - \frac{F(u)}{\beta} \right)} du \tag{51}$$

where:

$$W_i(T) = \exp\left(\int_{T_0}^{T} X_i(u) du \right) \tag{52}$$

and:

$$X_i(T) = \frac{s_i}{\beta} \exp\left(-\frac{E_i}{kT}\right) + \frac{R_{n,i}}{\beta} \frac{I(T)}{\left(\sum_i n_{i,0} + M - \frac{F(T)}{\beta} \right)} \tag{53}$$

3. NMTS model applied to glow curve analysis

Let us suppose that a sample is successively irradiated with the same dose and that each time the glow curve is recorded. If the glow curve does not show changes of its shape and area, then we can assume that deep traps, if present, are fully occupied. In this case the NMTS model could hold. In what follows we will suppose that the glow curve is made up of a single peak. Under these conditions expression (41) can be employed as the theoretical expression $I(T,\alpha)$ to fit the glow curve by resorting to the L-M algorithm. Extrapolation to a multi-peak situation is straightforward by resorting to Eq. (49) as the fitting expression corresponding to $I(T,\alpha)$.

As mentioned, the L-M algorithm is an iterative procedure. For this reason, computation times should be as low as possible in order to make the fitting algorithm feasible. After substituting the term $\frac{W(u)}{W(T)}$ in Eq. (47) it results:

$$n(T) = \frac{n_0}{W(T)}$$
$$+ \frac{R_n N}{\beta} \int_{T_0}^{T} \left[\exp\left(\int_T^u X(u) du \right) \frac{I(u)}{\left[\sum_i n_{i,0} + M - \frac{F(u)}{\beta} \right]} \right] du \tag{54}$$

The double integration requires high accuracy, which means long computational times.

From Eq. (44) we have:

$$\int_{T_0}^{x} X(u) du = \int_{T_0}^{x} \left(\frac{s}{\beta} \exp\left(-\frac{E}{kT}\right) \right) + \int_{T_0}^{x} \left(\frac{R_n}{\beta} \frac{I(T) du}{\left(\sum_i n_{i,0} + M - \frac{F(T)}{\beta} \right)} \right) \tag{55}$$

According to Balarin [14], the first integral on the right side can be approached by:

$$\int_{T_0}^{T} \frac{s}{\beta} \exp\left(-\frac{E}{ku}\right) du = \frac{s}{\beta} \left(\frac{kT^2}{E} \frac{\exp\left(-\frac{E}{kT}\right)}{\sqrt{1 + \frac{4kT}{E}}} - \frac{kT_0^2}{E} \frac{\exp\left(-\frac{E}{kT_0}\right)}{\sqrt{1 + \frac{4kT_0}{E}}} \right) \tag{56}$$

In what follows the term on the right hand-side in Eq. (56) will be indicated by

$D(T, E)$.

If we take into account that $I(T) = \frac{dF}{dT}$ it is easy to show that:

$$\frac{R_n}{\beta} \int_{T_0}^{T} \frac{I(u) du}{\left(n_0 + M - \frac{F(u)}{\beta} \right)} = R_n \ln\left(\frac{n_0 + M}{\sum_i n_{i,0} + M - \frac{F(T)}{\beta}} \right) \tag{57}$$

Introducing this result into Eq. (55), and after minor algebra, one obtains:

$$n(T) = n_0 \exp(-D(T, E)) \left[\frac{\sum_i n_{i,0} + M - F(T)}{\sum_i n_{i,0} + M} \right]^{R_n} +$$
$$+ \frac{R_n N (\sum_i n_{i,0} + M - F(T))^{R_n}}{\beta} \int_{T_0}^{T} \exp(D(u, E)$$
$$- D(T, E)) \frac{I_{exp}(u)}{\left[\sum_i n_{i,0} + M - \frac{F(u)}{\beta} \right]^{R_n+1}} du \tag{58}$$

This relationship for each trap along with Eq. (49) allows the analysis of glow curves by assuming that the NMTS. It is worth mentioning that in Eq. (58) $I(T)$ is actually $I_{exp}(T)$, namely, the glow curve recorded experimentally. In the same way, $F(t) = \int_{T_0}^{T} I_{exp}(u) du$. Thus Eq. (49) is a self-consistent equation.

4. Analysis of the glow curve BeO (Thermalox 995) compounds

In order to evaluate the performance of the expressions derived above, a glow curve of BeO (Thermalox 995) was analyzed. The sample was irradiated with an ophthalmic 90-Sr beta source and its glow curve was recorded with a Harshaw 3500 TL reader at a heating rate of 1 K s^{-1}. The dose amounted to 0.27 Gy. (Fig. 3)

Fig. 4 shows three glow curves obtained with three heating rates, namely, 1, 3 and 5 K/s.

From Fig. 4 it is clear that the area of the glow curve decreases as the heating rate increases. This means that BeO is affected by thermal quenching. Indicating with A1, A2, and A3 the area of the glow curves recorded with heating rates of 1, 3 and 5 K/s respectively, the following relationships were found:

$$\frac{A2}{A1} = 0.774, \quad and \quad \frac{A3}{A1} = 0.663.$$

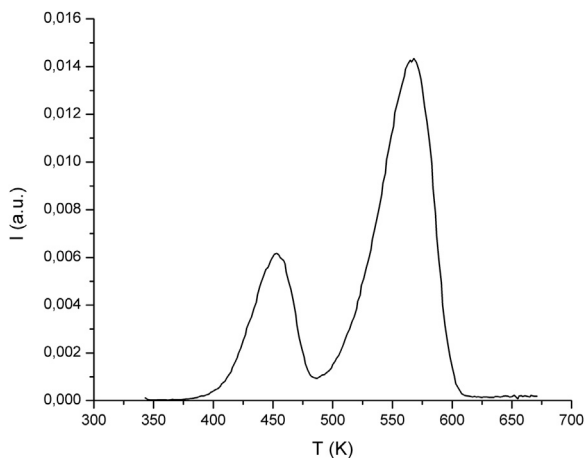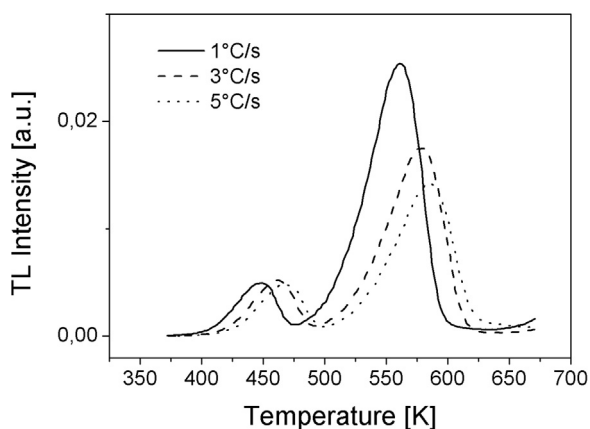**Fig. 3.** Recorded glow curve for a dose of 0.27 Gy.

Fig. 4. Glow curves obtained for the three heating rates indicated in the figure.

Yukihara reported the thermal quenching function [15]. The function, known also as luminescence efficiency, is given by:

$\eta(T) = \frac{1}{1 + C \exp(E_q / kT)}$ where $C = 2.66 \; 10^7$, and $E_q = 0.568$ eV.

The corrected glow curve is given by:

$$I_{corr}(T) = \frac{I_{meas}(T)}{\eta(T)}$$

Fig. 5 shows the corrected glow curve, and that resulting from the fit of Eq. (49) to the corrected glow curve.
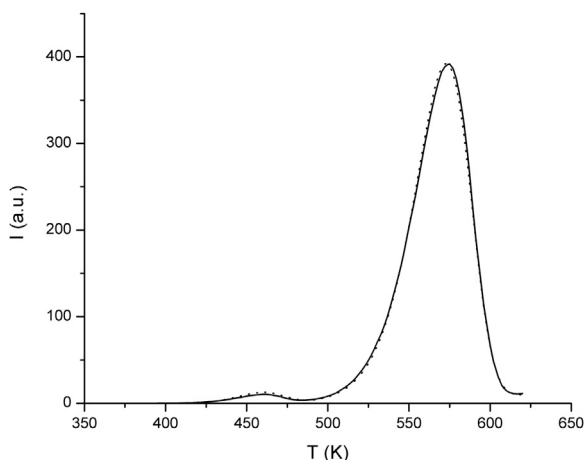

Fig. 5. Corrected glow curve (solid line), and fitted curve (dot line). Heating rate β = 1 K/s. FOM = 3.5%.

Table 1
Computed parameters. The concentration of active traps and deep traps are given in units of area (see Appendix A). FOM = 3.6%.

Trap	E (eV)	s (1/s)	$n_{,o}$	N	R	M
1	0.91	$6.0 \; 10^{11}$	360	$6.0 \; 10^6$	0.009	$1 \; 10^7$
2	1.23	$6.1 \; 10^{12}$	18190	$8.5 \; 10^5$	0.500	

ESR-TL correlation studies on BeO samples (Thermalox 995) show the presence of two traps giving rise to the glow curve [16]. There is a third trap but its contribution is negligible. Thus, two traps have been considered for the glow curve analysis (k = 2 in Eq. (49)). The resulting parameters are shown in Table 1.

Since most of the articles reporting analyses of glow curves employed the GO kinetics [2], in particular the expression put forward by Kitis et al. [17], it is worthwhile to compare the parameters obtained with the new equation with those obtained with the GO kinetics. For two traps the light intensity is given by [2]:

$$I(T) = \frac{s_1. \; n0_1. \; \exp(-\frac{E_1}{kT})}{\left[1 + (b_1 - 1)n0_1 \int_{T0}^{T} \exp(\frac{-E}{ku})du \right]^{\frac{b_1}{b_1 - 1}}}$$
$$+ \frac{s_2. \; n0_2. \; \exp(-\frac{E_2}{kT})}{\left[1 + (b_2 - 1)n0_2 \int_{T0}^{T} \exp(\frac{-E_2}{ku})du \right]^{\frac{b_2}{b_2 - 1}}}$$

In Table 2 the parameters computed with the GO kinetics are listed. From the comparison of the set of parameters shown in Tables 1 and 2 it can be concluded:

a) the energies differ significantly.
b) the new kinetics gives the concentration of active traps, and the concentration of deep traps, while the GO kinetics does not.
c) The new kinetics gives the parameter R of each trap.
d) For $b = 1$ the GO kinetics becomes first order kinetics, which means that retrapping is negligible, namely, $R_i = 0$ for i = 1,2, but according to the new kinetics for the second trap $R = 0.5$, i. e., the retrapping of trap 2 is not negligible against recombination.
e) For low doses the filling of the traps during irradiation is proportional to $N_i. \; R_i$. From Table 1 $N_1. \; R1 = 5.4 \; 10^4$, and $N_2. \; R_2 = 4.3 \; 10^5$. Therefore for low doses trap 2 will capture more electrons than trap 1. Thus, the second peak will be higher than the low temperature peak for low doses. For higher doses trap 2 approaches saturation ($N_2 < N_1$), and trap 2 starts to capture more electrons. As a consequence, the low temperature peak grows more than the high temperature peak at higher doses, as can be seen in Fig. 1 of reference [16]. This information cannot be obtained from the GO kinetics.
f) The new kinetics takes into account interaction among traps, while the heuristic GO kinetics does not.
g) In Table 1 the quotient N_1/N_2 is nearly 7, result that agrees with the concentration of traps 1 and 2 measured with ESR (see Figs. 7 and 8 of reference [16].

Finally, Azorin Nieto et al. report the energy for the highest temperature trap lying in the interval between 1.07 and 1.12 eV, and the frequency factor lying between $9 \; 10^{10}$ and $2.2 \; 10^{11}$ 1/s. These values

Table 2
Trap parameters computed with the GO kinetics. The concentrations of trapped electrons are given in units of area (see Appendix A). FOM = 2.0%.

Trap	E (eV)	S (1/s)	$n_{1,o}$	b
1	1.06	$2.4 \; 10^{10}$	397	1
2	1.62	$7.0 \; 10^{12}$	17930	1

differ from those obtained with the new kinetics [20].

5. Summary

The kinetics employed so far for analyzing glow curves are either heuristic (GO kinetics), or are derived from a physical model by resorting to the quasi-equilibrium approximation (first order, second order, mixed order). Recently an expression based on the OTOR model was derived, which resorts to approximations and for only one trap, i. e., ignoring the interaction among traps [18,19]. On the contrary, the closed expression put forward in this article was derived for the IMTS and NMTS models without resorting to any approximation and taking into account interaction among traps, which renders more reliable the parameters characterizing traps and recombination centers.

Appendix A. Some aspects of the glow curve analysis

A remark should be made about how Eq. (46) should be employed in order to find the set of parameters α. The mentioned equation renders the intensity in counts per second. Since usually the TL readers give the light intensity in Amperes, to perform a fitting Eq. (46) should be written as:

$$I(T, \alpha) = CA_h \left(N_0 - \frac{1}{\beta} F(T) - \sum_{i=1}^{k} n_i(T) \right) \left(N_0 + M - \frac{1}{\beta} F(T) \right) \tag{59}$$

The constant C takes into account the proportionality between the intensity given by counts per second and the recorded intensity given by the TL readers in Amperes.

Eq. (59) can be rewritten:

$$I(T, \alpha) = \lambda \left(CN_0 - \frac{1}{\beta} CF(T) - \sum_{i=1}^{k} Cn_i(T) \right) \left(CN_0 + CM - \frac{1}{\beta} CF(T) \right) \tag{60}$$

where $\lambda = A_h/C$.

Constant C can be chosen so that the concentration of trapped electrons just before the recording of a glow curve, namely, CN_0, be given in units of area. Thus CN_0 is the area of the glow curve. By the same token $Cn_{0,i}$ is the initial concentration of trapped electrons in trap #i, which is also given in units of area. Indeed, this is the parameter actually indicated as n_0 in Tables 1 and 2. Because of this change of units the concentration $\boldsymbol{M}$ of deep traps is also given in units of area, and $CF(T) = \int_{T_0}^{T} I_{\exp}(u) du$ is the area of the recorded glow curve $I_{\exp}(T)$ between the temperatures T_0 and T.

This metric for giving the concentration of carriers in units of area is not new. It has been reported in reference [21]. The advantage of this metric is that the initial concentration of trapped electrons n_0 is known, namely, it is given by the area of the glow curve.

It should also be mentioned that fitting algorithms, such as the L-M method, require that $I(T)$ and

$$CF(T) = \int_{T_0}^{T} I_{\exp}(u) du$$

be continuous and differentiable functions. TL readers give $I_{exp}(T)$ at sampling points. Continuous and differentiable functions for $I(T)$ and $C.F(T)$ can be obtained by approximating them with cubic spline, or another function, as for instance, the equations describing the GO kinetics.

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