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Analysis of the main dosimetric peak of Al_2O_3:C compounds with a model of interacting traps

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

[a] Universidad Nacional del Centro, 7000 Tandil, Argentina
[b] Consejo Nacional de Investigaciones Científicas y Técnicas (CONICET), Buenos Aires, Argentina
[c] Dresden University of Technology, Institute of Nuclear and Particle Physics, Radiation Physics Group, Germany
[d] Comisión de Investigaciones Científicas de la Provincia de Buenos Aires, 1900 La Plata, Argentina

HIGHLIGHTS

- Glow curves of Al_2O_3 :C for two doses have been analysed taking into account interactions among traps.
- The system of differential equations describing the kinetics has been uncoupled.
- The new system of equations takes into account equations without derivatives.
- The algorithm used will not become stiff.
- The kinetics parameters obtained do not depend on the dose.

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ABSTRACT

The glow curve of Al_2O_3:C compounds has been analyzed by employing a model consisting of two active traps, thermally disconnected traps and one recombination centre. The analysis takes into account interaction among traps and the thermal quenching of the thermoluminescent emission.

1. Introduction

The Al_2O_3:C compound is an intensively investigated material because of its thermoluminescent, radioluminescent and optical stimulated luminescent properties (Akselrod and Gorelova, 1993; Akselrod et al., 1998, 2006; Emfietzoglou and Moskovitch, 1996; Kitis et al., 1994; McKeever et al., 1999; Moskovitch et al., 1993; Polf et al., 2004; Santiago et al., 2012; Weinstein and Pelenyov, 2004; Xing-Bo et al., 2008; Xing-Bo et al., 2010; Yukihara et al., 2003). A frequently used procedure for investigating the mechanisms involved in the thermoluminescence of materials is the deconvolution of glow curves. Basically deconvolutions are employed for finding parameters such as trapping probabilities of electrons (or holes) by trap and recombination centres, activation energies and frequencies factors. The last two are related to the escape probability of an electron (or hole) from a trap centre (Chen and McKeever, 1997). Deconvolutions consist of finding a theoretical expression for the glow curve. The theoretical expression contains the trap parameters. The parameters are given different guess values until a satisfactory fit is achieved. Instead of randomly changing the parameters, an algorithm, such as the Levenberg–Marquardt (Horowitz and Yossian, 1995), is usually employed to find the parameters that yield the best fit between the theoretical and the experimental glow curve. As for Al_2O_3:C the parameters characterizing its thermoluminescent kinetics were found by deconvolution of glow curves employing a kinetics known as the general order kinetics (GO) (Nemecddin Yazici et al., 2003; Zahedifar et al., 2012). The GO kinetics is a heuristic model, and it was put forward by May and Patridge (1964). The GO kinetics suffers from several flaws:

1) It does not yield recombination and trapping probabilities. Instead it yields a parameter, usually denoted as b, which is loosely related to trapping probabilities.

* Corresponding author at: Universidad Nacional del Centro, 7000 Tandil, Argentina. Tel.: +54 249 4439660; fax: +54 249 4439669.
E-mail addresses: ecaselli@exa.unicen.edu.ar, caselli44@gmail.com (E. Caselli).

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2) Investigations have been carried out to find a connection between b and physically meaningful models, but a clear relationship could not been established (Sunta et al., 1997, 2002).
3) It has been reported that the GO model has limitations for determination of the activation energy (Sunta et al., 1999).
4) Sakurai has shown by resorting to computer simulation that the GO kinetics can yield wrong parameters (Sakurai and Gartia 1996;Sakurai, 2001). Further 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 (Sakurai and Gartia, 2003).
5) Marcazzó et al. (2007) have shown that it is incorrect to assume beforehand that a peak is related to a specific trap, as the GO model does.
6) Basun et al. (2003) have shown that the interaction among traps affects the shape of glow curves, thus affecting the parameters yielded by the GO kinetics.

Nonetheless the GO kinetics is nowadays by far the most employed kinetics for analyzing glow curves. The rationale behind is, as pointed out by Lewandowski and McKeever (1999), that in practice the system of coupled differential equation describing correctly the thermoluminescent kinetics usually become intractable, so that the exact analytical solutions are unobtainable for even the simplest of systems. Thus physically meaningful models can only be described by a set of coupled differential equations that has to be integrated for a given set of guess values of the parameters to find the theoretical value of the light intensity at a given temperature. Therefore so many integrations are necessary to obtain the theoretical glow curve as the number of sampling temperatures for which the digitized experimental glow curve has been recorded. Later, starting with the set of guess values, the Levenberg–Marquardt algorithm changes the parameters in an iterative way until the fit between the experimental and theoretical glow curve is satisfactory. The goodness of the fit is evaluated by means of the factor of merit (*FOM*) given by

$$FOM = \frac{\sum_{j=1}^{N} |I_{th}(t_j,\alpha) - I_{\exp}(t_j)|}{\sum_{j=1}^{N} |I_{\exp}(t_j)|} 100\%$$

A set of parameters is acceptable if the *FOM* is less than 5% (Horowitz and Yossian, 1995).

On occasions the set of coupled differential equations becomes stiff for the set of the chosen guess values. Recently an algorithm has been reported to avoid this problem (Chung et al., 2012). Nonetheless, computational times remain huge if several traps are included in the adopted model, which makes the deconvolution intractable.

In this article we report the parameters obtained by analyzing glow curves of Al_2O_3:C (Landauer). For the analysis of the glow curves we resorted to the procedure described in Caselli et al. (2011), which includes interaction among traps. The algorithm features an advantage: the set of coupled differential equations is replaced by a set of uncoupled equations without derivatives. Thus the problem of choosing a huge number of points for performing the integrations in order to avoid the equations becoming stiff is solved. As a consequence deconvolutions become tractable.

2. Model and algorithm

As stated above TL kinetics is described by a set of coupled differential equations. For the model shown in Fig. 1, composed of three active traps, one thermally disconnected trap and a recombination centre, the equations read

$$\frac{dn_1(t)}{dt} = -s_1 n_1(t) exp\left(-\frac{E_1}{kT}\right) + A_{n1}(N_1 - n_1(t)) n_c(t) \quad (1)$$

$$\frac{dn_2(t)}{dt} = -s_2 n_2(t) exp\left(-\frac{E_2}{kT}\right) + A_{n2}(N_2 - n_2(t)) n_c(t) \quad (2)$$

$$\frac{dn_3(t)}{dt} = -s_3 n_3(t) \exp\left(-\frac{E_3}{kT}\right) + A_{n3}(N_3 - n_3(t)) n_c(t) \quad (3)$$

$$\frac{dh(t)}{dt} = -(A_m n_c(t))(h(t)) \quad (4)$$

$$h(t) = n_1(t) + n_2(t) + n_3(t) + n_c(t) + M \quad (5)$$

After solving the set of coupled differential equations the theoretical glow curve is given by:

$$I(t) = -C\frac{dh(t)}{dt} \quad (6)$$

C is a constant which takes into account the light collection by a phototube, and its response.

With N_i and n_i the concentration of traps ♯i and the number of trapped electrons in them are respectively indicated. n_c stands for the concentration of electrons in the conduction band. E_i and s_i are the activation energy and the frequency factor respectively related to the trap ♯i. $p_i = s_i \exp(-E_i/kT)$ is the probability that a trapped electron being freed from trap ♯i. k stands for the Boltzmann constant and T is the absolute temperature. $A_{n,i}$ and $A_{m,i}$ denote the retrapping and recombination probabilities respectively of trap ♯i. M stands for the concentration of thermally disconnected traps, i.e., traps that retain the trapped electrons (or holes) for the temperatures the sample is subjected to. The thermally disconnected trap is supposed to be fully occupied.

The system of coupled differential equations has to be integrated taking into account the initial values $n0_1 = n_1(T = T0)$, $n0_2 = n_2(T = T0)$, $n0_c = n_c(T = T0)$ and $h0 = h(T = 0)$. $T0$ is the lowest temperature of the interval considered for deconvolving the glow curve. As shown by Caselli et al. (2011) the differential equations can be decoupled, and an expression for the theoretical glow curve is obtained, which does not contain derivatives. The algorithm is valid for models made of one recombination centre, any number of fully occupied thermally disconnected traps and of active traps, and if $n_i \ll N_i$.

For the model shown in Fig. 1 the expression for the theoretical glow curve is

$$I(T) = \gamma\left(MM + \frac{A}{\beta} - \frac{F(T)}{\beta}\right)\left(\frac{A}{\beta} - \frac{F(T)}{\beta} - NN_1 x_1(T) - NN_2 x_2(T) - NN_3 x_3(T)\right) \quad (7)$$

and

$$x_i(T) = x0_i \exp\left(-\frac{s_i}{\beta}\int_{T0}^{T} \exp\left(-\frac{\varepsilon_i T0}{u}\right) du\right) + \alpha_i \int_{T0}^{T} \left[\frac{I(z)}{MM + (A/\beta) - (F(z)/\beta)} \exp\left(\frac{s_i}{\beta}\int_{z}^{T} \exp\left(-\frac{\varepsilon_i T0}{u}\right) du\right)\right] dz \quad (8)$$

where $x_i(T) = n_i(T)/N_i$ and $x0_i$ is the fraction of occupation of trap ♯i at $T = T0$, i.e. $x0_i = (n_i(T = T0))/N_i$.

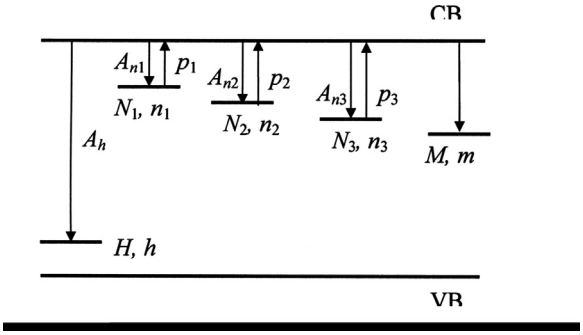

Fig. 1. Model employed for analyzing the glow curve.

The parameter in the foregoing equations are defined as follows: β is the heating rate employed for recording the glow curve, $\gamma = A_m/C$, $\alpha_i = A_{ni}/A_m$, $NN_i = C.N_i$, $MM = C.M$, $F(T) = \int_{T0}^{T} I_{exp}(u)du$, $I_{exp}(T)$ is the experimental glow curve and A its area. $\varepsilon_i = E_i/kT0$ is the activation energy normalized to the energy $kT0$.

The nine parameters NN_i, $x0_i$ and α_i, $i=1,2,3$, can be reduced to six parameters by defining: $Nx0_i(T) = NN_i x0_i$ and $N\alpha_i = NN_i\alpha_i$. Thus the equations read

$$NN_i x_i(T) = Nx0_i \exp\left(-\frac{s_i}{\beta}\int_{T0}^{T}\exp\left(-\frac{\varepsilon_i T0}{u}\right)du\right) + N\alpha_i\int_{T0}^{T}\left[\frac{I(z)}{MM+(A/\beta)-(F(z)/\beta)}\exp\left(\frac{s_i}{\beta}\int_{z}^{T}\exp\left(-\frac{\varepsilon_i T0}{u}\right)du\right)\right]dz \quad (9)$$

so that

$$I(T) = \gamma\left(MM+\frac{A}{\beta}-\frac{F(T)}{\beta}\right)\left(\frac{A}{\beta}-\frac{F(T)}{\beta}-Nx_1(T)-Nx_2(T)-Nx_3(T)\right) \quad (10)$$

where the following variable change has been made: $Nx_i(T) = NN_i x_i(T)$.

The reduction of parameters yields a reduction of the computational times.

Further reduction of the number of parameters can be achieved by noting that if $n_i \ll N_i$ the rate of trapped electrons when a sample is irradiated is given by $dn_i(t)/dt = A_{ni}N_i n_c(t)$. Thus the concentration of trapped electrons when the irradiation stops at $t=T$ is given by $n0_i = A_{ni}N_i\int_0^T n_c(t)dt$. From this expression the following equations are derived:

$$\frac{Nx0_i}{Nx0_j} = \frac{N\alpha_i}{N\alpha_j}. \quad Thus \quad Nx0_2 = \frac{Nx0_1 N\alpha_2}{N\alpha_1} \quad and \quad Nx0_3 = \frac{Nx0_1 N\alpha_3}{N\alpha_1}. \quad (11)$$

Therefore the parameters $Nx0_2$ and $Nx0_3$ drop in Eq. (9).

An additional time reduction is achieved by resorting to the expression reported by Balarin (1977) for the integrals in the foregoing equations.

$$\int_0^T \exp\left(-\frac{\varepsilon_i T0}{u}\right)du \simeq \frac{T^2 e^{-T0\varepsilon_i/T}}{\sqrt{1+(4T/T0\varepsilon_i)}} \quad (12)$$

3. Deconvolution

The first step for deconvolving a glow curve is the adoption of a model which is in agreement with experimental findings. We have considered the following findings:

(1) Walker et al. (1996) concluded from the dependence of the main dosimetric peak with dose that it is made up of the superposition of several peaks related to traps having close energy levels.
(2) Yukihara et al. (2003) reported that additionally to the dosimetric traps there are deep traps which are active at temperatures higher than 800 K and deep electron traps, which are active at temperatures higher than 1100 K.
(3) Thermal quenching affects the glow curve of Al_2O_3:C compounds (Akselrod et al., 1998). The expression which gives the thermal quenching, known as the Mott–Seitz equation, is $\eta(T) = 1/1+a\exp(-W/kT)$, which depends on the parameters a and W characteristic of each material.
(4) The TL emission features a peak at about 420 nm, which indicates the presence of only a radiative recombination centre.
(5) Zahedifar et al. (2012) reported T_m–T_{stop} measurements showing that for α-Al_2O_3:C supplied by Harshaw–Bicron (TLD-500) the number of traps is three. From similar measurements Nemecddin Yazici et al. (2003) concluded that two traps give rise to the glow curve of α-Al_2O_3:C supplied by Victoreen Inc.
(6) Resorting to the Initial Rise procedure we found that the activation energies of the traps giving rise to the glow curve are around 1.3 eV. Similar results are reported by Zahedifar et al. (2012).

Taking into account the foregoing remarks deconvolutions were performed with (1) a model of one recombination centre, two active traps and thermally disconnected traps, which are included through the net concentration of charge, i.e., the concentration of electrons minus the concentration of holes represented by the parameters *MM*, and (2) a model of one recombination centre, three active traps and thermally disconnected traps. For the analysis Eq. (10) has been multiplied by $\eta(T) = 1/1+a\exp(-W/kT)$ in order to include thermal quenching. The employed parameters are $a = 10^{11}$ and $W = 1.1$ eV(Pagonis et al., 2007).

Two glow curves were obtained by irradiating a sample for 10 s and 1000 s with a Sr-90 source. The estimated doses are 0.4 and 40 cGy correspondingly. For these doses the approximation $n_i \ll N_i$ holds.

Deconvolutions with two active traps yielded activation energies which differ significantly from the values obtained by means of the IR method. Thus the model with three traps was employed to analyze two glow curves. First the glow curve with the low dose was analyzed, and later it was checked whether the resulting parameters from the analysis of the low dose glow curve yield a glow curve that coincides with the high dose glow curve. To this end the parameter $Nx0_1$ obtained for the low dose glow curve was multiplied by 77.2, i.e., the quotient of the areas of the high dose curve to the low dose curve. Calculations were performed until a set of parameters was found, which yielded *FOM*'s lower than 5% for both doses. Table 1 shows the parameters for both glow curves, and Fig. 2(a) and (b) shows the experimental glow curves and those resulting from the fitting. As can be seen in Table 1 the agreement among the parameters for the two doses is excellent.

If the concentration of trapped electrons in each trap as function of the temperature is multiplied by $\eta(T)$ it results the contribution of each trap to the glow peak, i.e., $nni(T) = NN_i(T)\eta(T)$. Fig. 3 shows the result. As can be seen the trap with the highest activation energy makes the largest contribution to the glow peak.

The activation energy of trap ♯1 amounts to 1.19 eV, a value 8% lower than the value found from IR calculations we performed, i.e., 1.29 eV. Activation energies computed from glow curves are frequently lesser than the values found from initial rise (Jose et al., 2011; Zahedifar et al., 2012). In our case we believe that the small difference in percentage terms is due to the overlap of the contributions of traps ♯1 and ♯2 giving rise to the rising part of the glow peak.

Table 1
LD=low dose, HD=high dose. The value of MM is 7×10^7.

Parameters obtained from deconvolution	s_1 (s^{-1})	ε_1 (eV)	$N\alpha_1$	$Nx0_1$	s_2 (s^{-1})	ε_2 (eV)	$N\alpha_2$	s_3 (s^{-1})	ε_3 (eV)	$N\alpha_3$	*FOM* (%)
LD	1.4×10^{12}	1.19	110.0	3.2	8.4×10^{12}	1.33	165.0	4.0×10^{12}	1.37	355.7	3.6
HD	1.6×10^{12}	1.19	110.4	249.0	1.1×10^{13}	1.33	166.4	5.0×10^{12}	1.37	356.3	3.8

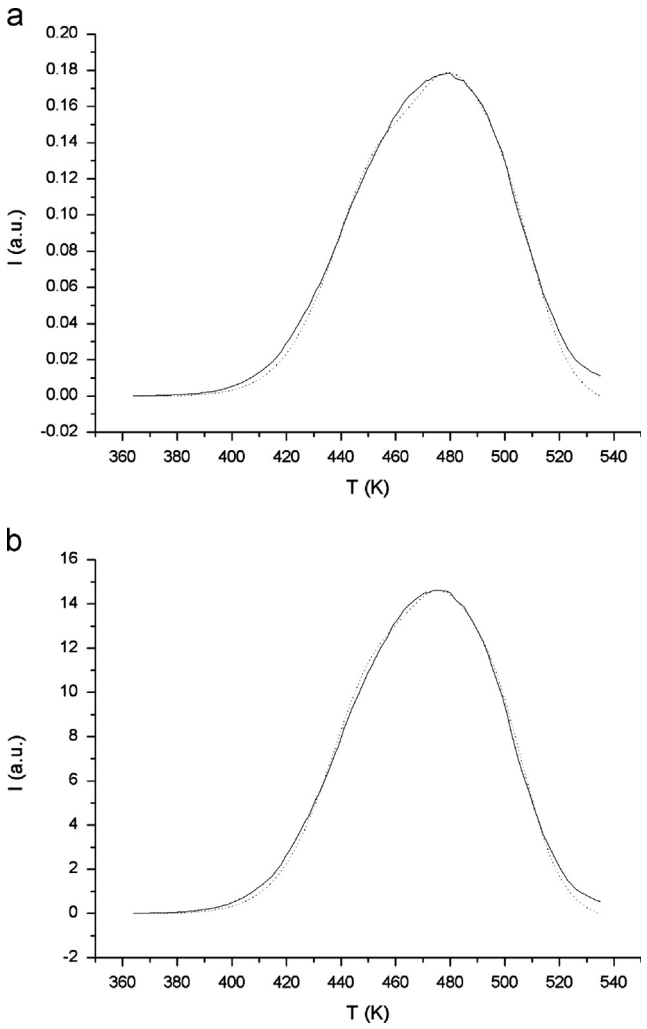

Fig. 2. (a) Solid line: experimental glow curve obtained after having irradiated the sample 10 s , dot line: fitted curve. (b) Solid line: experimental glow curve obtained after having irradiated the sample 1000 s, dot line: fitted curve.

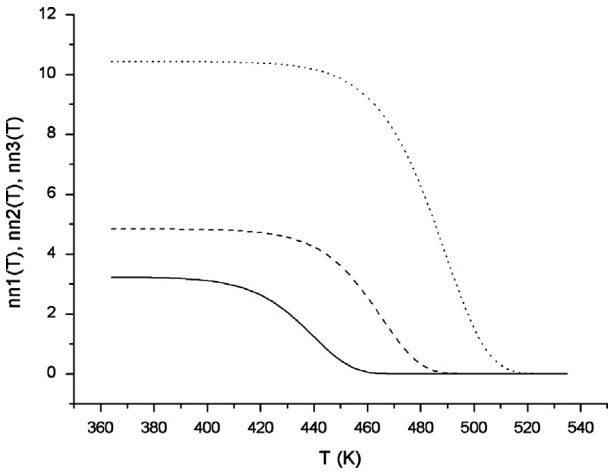

Fig. 3. Concentration of trapped electrons in each trap multiplied by $\eta(T)$. $nn1$ (line), $nn2$ (dash), and $nn3$ (dot).

4. Conclusions

The algorithm given by Eqs. (7) and (8) allows finding the theoretical intensity of the light as a function of the set of parameters related to traps and recombination centres, avoiding the integration of the set of coupled differential equations. In order to reduce computational times expression 12 has been employed. Further, new parameters have been defined, the number of which is less than the number of the original parameters. The new set of parameters also contributes to reduce computational times, but information is lost. For example, the relative concentrations of traps NN_i and the filling factors $x0_i$ are not obtained, but the product of both parameters $Nx0_i$ is obtained. The same happens with the parameter $N\alpha_i$, i.e., α_i is not found. The rationale behind is that the approximation $n_i \ll N_i$ has as a consequence that the rate of electron trapping be $N\alpha_i$ instead of $(N_i - n_i)\alpha_i$. In the first case the rate depends only on the product of the concentration of traps N and the relative trapping probability α, while in the second case the rate depends not only on the product $N\alpha_i$, but also on n_i. Nonetheless the analysis of glow curves taking into account interaction among traps yields more reliable parameters, such as frequency factors and activation energies, since contrary to the GO kinetics, the model is physically meaningful.

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