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A unified presentation of thermoluminescence (TL), phosphorescence and linear-modulated optically stimulated luminescence (LM-OSL)

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Abstract

It has been shown in the past that a thermoluminescence (TL) peak has a relatively simple shape if the heating function is hyperbolic. Also, two different ways have been given for the transformation of the featureless decaying phosphorescence and optically stimulated luminescence (OSL) functions into peak-shaped curves. In the former case, one plots $L \cdot t$ versus $\ln(t)$, where L is the phosphorescence intensity and t the time. In the latter, linear-modulated OSL (LM-OSL) is reached by changing the stimulated light intensity linearly with time, and pseudo-LM-OSL by manipulating the regular CW-OSL curve. In this work, a modified presentation of LM-OSL is suggested and a unified presentation of these phenomena is offered, which may help in analysing the results and elucidating the details of the relevant kinetics process. The implications on the use of LM-OSL for dosimetry are also discussed.

1. Introduction

Thermoluminescence (TL) and optically stimulated luminescence (OSL) are two related phenomena, broadly used in radiation dosimetry and in the dating of archaeological and geological samples. TL was first explained with a sound theoretical model by Randall and Wilkins (1945). These authors introduced the ‘first-order kinetics’ while assuming that a freed carrier has negligible probability of retrapping. The governing first-order equation can be written as

$$L = -\frac{dn}{dt} = s \cdot n \cdot \exp(-E/kT), \quad (1)$$

which determines the simplest TL single peak if the heating function is given. Here, E (eV) is the activation energy, k (eV K⁻¹) the Boltzmann constant, s (s⁻¹) the frequency factor, T (K) the temperature, t (s) the time and n (cm⁻³)

the concentration of electrons trapped in the relevant trapping states with n_0 its value when the heating begins. The single TL peak in this simple situation behaves like the solution of equation (1),

$$L(t) = sn_0 e^{-E/kT} \exp\left[-s \int_0^t e^{-E/kT'} dt'\right]. \quad (2)$$

Note that this expression is the intensity as a function of time. The ‘conventional’ expression where L is given as a function of temperature, T , for a linear heating function is reached by writing $dt' = dT'/\beta$, where β (K s⁻¹) is the constant heating rate, and taking the lower and upper limits of the integral as T_0 and T , respectively. Garlick and Gibson (1948) extended the theory to cases where retrapping is substantial, and got the second-order governing differential equation

$$L = -\frac{dn}{dt} = s' \cdot n^2 \cdot \exp(-E/kT), \quad (3)$$

where s' ($\text{cm}^3 \text{s}^{-1}$) is the pre-exponential factor. The solution of equation (3) is

$$L(t) = \frac{n_0^2 s' \exp(-E/kT)}{\left[1 + n_0 s' \int_0^t \exp(-E/kT') dt'\right]^2}. \quad (4)$$

Here too, the expression for linear heating function is written by replacing dt' by dT'/β and changing the integral limits to the appropriate temperatures.

May and Partridge (1964) suggested the use of 'general-order' kinetics, which can be written as

$$L = -\frac{dn}{dt} = s' \cdot n^b \cdot \exp(-E/kT). \quad (5)$$

Here, b is the effective order, usually $1 \leq b \leq 2$, and s' is a constant with dimensions of $\text{cm}^{3(b-1)} \text{s}^{-1}$, which is the generalization of the pre-exponential factor s' mentioned above in relation to the second-order case where $b = 2$. Rasheedy (1993) proposed the use of $s' = s/N^{b-1}$ where N is the concentration of the relevant trapping states. Although entirely heuristic, the general-order approach has become very popular in the study of TL and related phenomena. The solution of equation (5) is

$$L(t) = s'' n_0 \exp(-E/kT) \times \left[1 + (b-1)s'' \int_0^t \exp(-E/kT') dt'\right]^{-b/(b-1)}, \quad (6)$$

where $s'' = s' n_0^{b-1}$. As in the cases mentioned above, the temperature dependent intensity $L(T)$ is reached when the specific heating function $T(t)$ is inserted in equation (6), the simple case being the linear heating function mentioned.

2. Presentations of the different luminescence phenomena

When the 'conventional' linear heating function is utilized, the exponential integral appearing in equations (2), (4) and (6) changes into $\int_{T_0}^T \exp(-E/kT') dT'$. This is not an elementary integral, and a number of numerical methods for its evaluation have been given in the literature. An alternative has been suggested (see e.g. Arnold and Sherwood (1959), Stammers (1979), Chen and McKeever (1997)), namely the use of a hyperbolic heating function, which transforms the integral into an elementary one. In this case, the relation between time and temperature is $1/T = 1/T_0 - t/\beta'$ where β' (K·s) is a constant. Therefore, one has $dt = (\beta'/T^2) dT$, which can be inserted in equations (2), (4) and (6). The resulting integral is

$$\int_0^t \exp(-E/kT') dt' = \beta' \int_{T_0}^T (1/T'^2) \exp(-E/kT') dT' \\ = (k\beta'/E) [\exp(-E/kT) - \exp(-E/kT_0)]. \quad (7)$$

Since the function $\exp(-E/kT)$ increases very fast with temperature, the second term in the square brackets is negligible for T larger than T_0 even by a few degrees. The use of this expression will be elaborated below.

Another important case is that of phosphorescence, namely, where the temperature of the sample is held constant

following excitation. The integral in the above equations is of a constant, and therefore, of course,

$$\int_0^t \exp(-E/kT') dt' = \exp(-E/kT) \cdot t. \quad (8)$$

When inserted in equations (2), (4) and (6), different kinds of phosphorescence decay are found. Since in all these cases the functional form of phosphorescence is a rather featureless decay, it is quite difficult to tell them apart. In their seminal work, Randall and Wilkins (1945) suggested plotting $L \cdot t$ as a function of $\ln t$. The result is a peak-shaped curve, quite similar to the first-order TL peak. Visocekas (1978) and Chen and Kristianpoller (1986) have extended this presentation to general-order peaks.

OSL has been developed as an important tool for dosimetry and archaeological and geological dating (see e.g. Huntley *et al* 1985). In fact, for a given sample, the equations governing OSL and phosphorescence are very similar. Whereas in phosphorescence, trapped electrons are released thermally into the conduction band prior to their recombination in a centre, in OSL the release is by photons of the stimulating light. Thus, equations very similar to (2), (4) and (6) can be used when a term proportional to the intensity of the stimulating light replaces the product of s (or s'' , depending on the specific case) and $\exp(-E/kT)$ which, in the case of phosphorescence is a constant. Bulur (1996) suggested changing the featureless OSL decay curve into a peak-shaped function by increasing the intensity of the stimulating light linearly with time. Using the linear-modulated OSL (LM-OSL), the governing equation is

$$L(t) = -dn/dt = \alpha(I_0/\theta)tn(t), \quad (9)$$

where θ (s) is the duration of the observation and αI_0 (s^{-1}) is the probability of escape of electrons from the traps at a light intensity of stimulation I_0 . The stimulating light intensity here varies with time as $I_0 t/\theta$. The solution of this equation yields the intensity measured under these circumstances as a function of time

$$L(t) = n_0 I_0 \alpha (t/\theta) \exp[-(\alpha I_0/2\theta)t^2]. \quad (10)$$

This is a peak-shaped function. As shown by Bulur, the maximum intensity here is given by

$$L_{\max} = n_0 (\alpha I_0/\theta)^{1/2} \exp(-1/2), \quad (11)$$

which is very useful in the applications in dosimetry since the maximum intensity is proportional to the initial concentration of carriers in traps, n_0 . This, in turn, is assumed to carry the information, which concerns the absorbed dose. On the other hand, the peak-shaped function (10) does not resemble the first-order kinetics TL peak mentioned above. A simple method discussed below will remedy this problem, and methods previously developed for the determination of the kinetics order in TL will be easily utilized here as well. As is quite obvious, integration of equation (10) from 0 to infinity yields n_0 . Thus, if one takes as the LM-OSL signal either the maximum intensity or the whole area under the curve, in the

first-order case, the signal is proportional to the concentration following the excitation, n_0 .

Bulur (1996) gave a similar treatment to LM-OSL governed by general-order kinetics:

$$L(t) = -dn/dt = (\gamma I_0/\theta)tn^b, \quad (12)$$

where γ is a constant with different dimensions (similar to s' in equation (5)) and b is the kinetics order. Here, Bulur (1996) assumed that $\gamma = \alpha/n_0^{b-1}$, where α is the frequency factor. Bulur and Göksu (1999) changed this assumption to $\gamma = \alpha/N^{b-1}$, similar to the assumption by Rasheedy (1993) for general-order TL. Using the definition $\gamma = \alpha/N^{b-1}$, the solution of this differential equation is

$$L(t) = n_0^b(\alpha I_0) \left[t/(N^{b-1}\theta) \right] \times \left\{ 1 + (b-1) \left[\alpha I_0 n_0^{b-1}/(2\theta N^{b-1}) \right] t^2 \right\}^{-b/(b-1)}. \quad (13)$$

An important special case is that of second order kinetics, where $b = 2$ and the intensity is given by

$$L(t) = \frac{n_0^2[\alpha I_0/(N\theta)]t}{\left\{ 1 + [\alpha I_0 n_0/(2N\theta)] t^2 \right\}^2}. \quad (14)$$

Bulur and Göksu (1999) have shown that for the general-order situation, by equating the derivative of equation (13) to zero, the condition for the maximum of the LM-OSL signal is

$$t_{\max} = \sqrt{\frac{2}{(b+1)} \frac{\theta}{\alpha I_0} \left(\frac{N}{n_0} \right)^{b-1}}. \quad (15)$$

For the special important case of second order kinetics, this reduces to

$$t_{\max} = \sqrt{\frac{2\theta}{3\alpha I_0} \frac{N}{n_0}}. \quad (16)$$

Inserting from equation (15) into (13) yields

$$L_{\max} = \frac{2n_0}{(b+1)} \frac{1}{t_{\max}} \left[\frac{2b}{b+1} \right]^{-b/(b-1)}. \quad (17)$$

In the special case of $b = 2$, this reduces to

$$L_{\max} = \frac{3}{8} \frac{n_0}{t_{\max}}. \quad (18)$$

It is quite obvious that since t_{\max} depends on n_0 , L_{\max} is not proportional to n_0 , but is rather superlinear with the initial carrier concentration. In ranges where n_0 is proportional to the dose, L_{\max} will be expected to be superlinear with the dose. If so, using L_{\max} as the signal measuring the applied dose may be rather problematic. It should be noted here that if we consider as the measured signal the area under the LM-OSL curve, taken from $t = 0$ to infinity, the situation is different. Since $L(t)$ is defined as $-dn/dt$, this integral must be n_0 . The fact that the total area and the maximum intensity do not behave in the same way with n_0 , and therefore with the dose, indicates that the shape of the peak distorts with increasing n_0 . Several investigators have used the LM-OSL for dosimetry and dating purposes (see e.g., Schlapp *et al* (2003), Adamiec (2005)).

3. The unified presentation

In this work, we show that the TL measured under hyperbolic scheme, phosphorescence and LM-OSL can be cast into the same form for any given kinetics order. Therefore, any method of analysis developed for one of these presentations can very easily be carried over to the other. For example, if the symmetry factor of TL measured with a hyperbolic heating function can be used for the evaluation of the effective kinetics order, it can just as well determine the kinetics order in the new unified presentation of LM-OSL.

Let us start with the unified presentation in the first-order case. Taking equation (2) for TL and using the hyperbolic heating function as given by equation (7) and without the term including T_0 which, as explained above is negligible, one gets

$$L(T) = sn_0 \exp(-E/kT) \exp[-(s\beta'k/E) \exp(-E/kT)]. \quad (19)$$

If we denote $-E/kT$ by x , we get

$$L(x) = sn_0 e^x \exp[-(s\beta'k/E)e^x], \quad (20)$$

which is practically the same as the expressions mentioned before in the literature (see e.g. Kelly and Laubitz (1967)). Note that, as defined, x is negative, a point that will be discussed below while comparison is made with the two other phenomena. It should be mentioned that a slightly different presentation could be made, namely, with $x = E/kT$. However, in the former, an increasing temperature T is associated with an increasing x , and therefore we prefer it over the latter, where it is the other way around.

In the case of phosphorescence, where the temperature is fixed, equation (2) along with (8) gives

$$L(t) = sn_0 \exp(-E/kT) \exp[-s \cdot \exp(-E/kT) \cdot t]. \quad (21)$$

As pointed out above, the way to transform this to a peak-shaped curve is plotting $L(t) \cdot t$ as a function of $\ln t$. We can write

$$y = L(t) \cdot t = \phi n_0 t e^{-\phi t}, \quad (22)$$

where $\phi = s \exp(-E/kT)$. If we denote $\ln t$ by x , we have $t = e^x$, and get

$$y(x) = \phi n_0 e^x \exp(-\phi e^x), \quad (23)$$

which, apart from the meaning of the constants, is exactly of the same form as equation (20).

As for the LM-OSL, let us multiply both sides of equation (10) by t , and we get

$$y = t \cdot L(t) = n_0 I_0 \alpha (t^2/\theta) \exp \left[- \left(\frac{\alpha I_0}{2\theta} \right) t^2 \right]. \quad (24)$$

If we now take $t^2 = e^x$ ($x = 2 \ln t$), we get

$$y(x) = 2n_0 \delta e^x \exp(-\delta e^x), \quad (25)$$

where $\delta = \alpha I_0/(2\theta)$. The shape of the first-order peak in equation (25) is exactly of the same form as in (20) and (23).

It should be noted that whereas in the TL case, equation (20), x is always negative, in equations (23) and (25),

x may be either positive or negative, depending on the relevant values of t ; for $t < 1$ it is negative and for $t > 1$ it is positive. This, however, is not important since changing the units of time may change x from positive to negative or vice versa. Making such a change simply means that instead of $\ln t$, we have $\ln(ct)$ where c is the conversion factor from, say, minutes to seconds ($c = 60$). Since $\ln(ct) = \ln c + \ln t$, this simply means a shift of the whole relevant curve by $\ln c$ along the x -axis.

A very similar treatment can be made in the three mentioned phenomena for the general-order kinetics, which includes, of course, the second order with $b = 2$. Inserting the value of the integral with the hyperbolic heating function (7) in equation (6), we get

$$L(T) = s''n_0 \exp(-E/kT)[1 + (b - 1)(s''k\beta'/E) \times \exp(-E/kT)]^{-b/(b-1)}, \quad (26)$$

and by using the same substitution $x = -E/kT$, we get

$$L(x) = s''n_0 e^x [1 + (b - 1)(s''k\beta'/E)e^x]^{-b/(b-1)}. \quad (27)$$

Obviously, in the second-order case, $b = 2$ and one gets

$$L(x) = s''n_0 e^x [1 + (s''k\beta'/E)e^x]^{-2}. \quad (28)$$

For the TL-like presentation of phosphorescence, the parallel expression is (see, Chen and Kristianpoller (1986))

$$y = \phi n_0 e^x [1 + (b - 1)\phi e^x]^{-b/(b-1)}, \quad (29)$$

where, again, $\phi = s \exp(-E/kT)$. Obviously, the second-order function is

$$y = n_0 \phi e^x [1 + \phi e^x]^{-2}. \quad (30)$$

As for the new way of presenting the LM-OSL, we multiply both sides of equation (13) by t , and take, as before, $y = t \cdot L(t)$, $t^2 = e^x$ and define $\delta' = \alpha I_0 n_0^{b-1} / (2\theta N^{b-1})$. We get

$$y = 2n_0 \delta' e^x [1 + (b - 1)\delta' e^x]^{-b/(b-1)}. \quad (31)$$

Here too, the second-order expression is simply

$$y = 2n_0 \delta' e^x [1 + \delta' e^x]^{-2}, \quad (32)$$

where $\delta' = \alpha I_0 n_0 / (2\theta N)$.

It is evident that equations (20), (23) and (25) have the same functional form for the three phenomena and first-order kinetics. The same is true for equations (27), (29) and (31) for general order, and equation (28), (30) and (32) for the second-order case.

4. Applications of the unified presentation

Chen and Kristianpoller (1986) have shown that for the TL-like presentation of phosphorescence, there is a one-to-one correspondence between the kinetic order and the symmetry factor μ_g (See e.g. figure 3.11 in Chen and McKeever (1997)). It should be mentioned that this figure resembles the one given by Chen (1969b) for TL with a linear heating function, except that in the present case, μ_g is, indeed, a unique function of

b , whereas in the linear heating function case, there is an apparent 'error bar' which, in fact, represents a relatively weak dependence of μ_g on the activation energy and the pre-exponential factor. Thus, somewhat different values of μ_g can be associated with a certain value of b , depending on the values of E and s (or s''). The one-to-one correspondence between b and μ_g given in the case of TL-like phosphorescence, can automatically be applied in the other mentioned phenomena since the underlying equation is the same. This includes TL with hyperbolic heating function, OSL decay transformed to the $L \cdot t$ versus $x = \ln t$, as well as the new way of presenting LM-OSL mentioned above. Once this presentation is used in any of these phenomena, the effective order of kinetics can be evaluated from the mentioned curve of μ_g versus b by using the experimentally determined value of μ_g .

Let us consider the signal dependence of the new presentation of LM-OSL on the initial concentration n_0 . For the first-order case, if we set the derivative of equation (25) with respect to x to zero, we get the condition for the maximum as

$$e^{x_m} = \frac{1}{\delta}, \quad (33)$$

where x_m is the maximum point. Re-inserting this into (25), we get

$$y_m = 2n_0 e^{-1}. \quad (34)$$

Here, the maximum intensity of the signal is proportional to n_0 , in a similar manner to that shown by Chen and Kristianpoller (1986), in the case of TL-like phosphorescence, where the maximum intensity in the first-order case is $y_m = n_0/e$. The factor of 2 in equation (34) arises from the same factor appearing in equation (25). As for the area under the curve, integration of equation (25) for x going from $-\infty$ to ∞ (t goes from 0 to ∞) yields

$$\int_{-\infty}^{\infty} y dx = 2n_0. \quad (35)$$

The significance of this is that while using this method for dosimetry, the maximum intensity of the curve as well as the area under the curve are proportional to n_0 , which, as pointed out, is usually the measure of the dose. It should be noted here that whereas the integral under the $L(t)$ curve is, by definition, equal to n_0 , this is not the case for the $y(x)$ curve. However, for the applications, it is important that the area is proportional to n_0 .

A similar treatment can be given to the general-order case. Setting the derivative of equation (31) to zero, we get again, as in equation (33) the condition for the maximum, $\exp(x_m) = 1/\delta$. Re-inserting this in equation (31), we get

$$y_m = \frac{2n_0}{b^{b/(b-1)}}. \quad (36)$$

Here too, the maximum intensity is proportional to n_0 , as opposed to the original presentation by Bulur (see equation (17)). For the second-order case, equation (36) reduces to $y_m = n_0/2$. Chen and Kristianpoller (1986) have shown that the maximum intensity for the TL-like phosphorescence is expressed by a similar equation, excluding the factor of 2.

As for the integral under the $y(x)$ curve, integrating over equation (31) from $-\infty$ to ∞ yields, again, $\int_{-\infty}^{\infty} y dx = 2n_0$,

which is identical to equation (35) for first order, and includes the important case of second-order kinetics.

The consequences concerning using the effect for dosimetry and dating and for the other phenomena within the unified presentation are obvious.

5. Numerical demonstration of the two presentations of LM-OSL

In this section, we show the different dependences of the maximum intensity of LM-OSL on the initial concentration n_0 in the two mentioned presentations in the non-first-order cases. For the demonstration, we limit the discussion to the second-order case, which is an extreme case in the range of orders of kinetics between 1 and 2. Let us re-write equation (14) for the LM-OSL as

$$L(t) = \frac{n_0^2 C t}{[1 + (n_0 C / 2) t^2]^2}, \quad (37)$$

where $C = \alpha I_0 / (N \theta)$. For the numerical demonstration, it is sufficient to choose a value for C and we have chosen $C = 10^{-11} \text{ cm}^3 \text{ s}^{-2}$. We have also taken a range of time from 1 to 100s, and varied n_0 from $1 \times 10^8 \text{ cm}^{-3}$ to $5 \times 10^8 \text{ cm}^{-3}$, as shown in curves (a)–(e) in figure 1. The maximum point t_{max} is seen to decrease with increasing n_0 , which could be predicted from equation (17). As for the maximum intensity, equation (18) along with (16) shows that in the second-order case, the dependence on n_0 should be as a power of 3/2. This is seen in figure 1. For example, the maximum of curve (e) is ~ 11.2 times that of (a) for a change of n_0 by a factor of 5, and indeed, $5^{3/2} \approx 11.2$. In view of equations (15) and (17), one can expect superlinearity, though with less than a 3/2 power for the dependence of L_{max} on n_0 for orders b between 1 and 2. Thus, one can expect a superlinear dependence of the conventional LM-OSL presentation on the excitation in ranges where n_0 is linearly dependent on the dose.

As for the new presentation of LM-OSL, multiplying equation (37) by t yields

$$y = t \cdot L(t) = \frac{n_0^2 C t^2}{[1 + (n_0 C / 2) t^2]^2}. \quad (38)$$

and using, as before, $t^2 = e^x$ ($x = 2 \ln t$), we get

$$y(x) = \frac{n_0^2 C e^x}{[1 + (n_0 C / 2) e^x]^2}. \quad (39)$$

For the given range $1 \leq t \leq 100$, we have $0 \leq x \leq 9.2$. Using the same value for C and the same five values of n_0 between 1×10^8 and $5 \times 10^8 \text{ cm}^{-3}$, we get figure 2. It is clearly seen that the shape of the curves is much more similar to TL curves, be it with linear (see e.g. Chen, (1969a)) or hyperbolic (see e.g. Kelly and Laubitz, (1967)) heating functions, than in the original LM-OSL presentation. Here too the maximum point x_m gets smaller with increasing n_0 , in the same way, which is very familiar for second-order TL, be it with a linear or a hyperbolic heating function. As for the maximum intensity, it is clearly seen that it is strictly linear with n_0 , and therefore, in many cases, with the applied dose. As mentioned above, this linear dependence in the new LM-OSL presentation has important practical consequences for dosimetry.

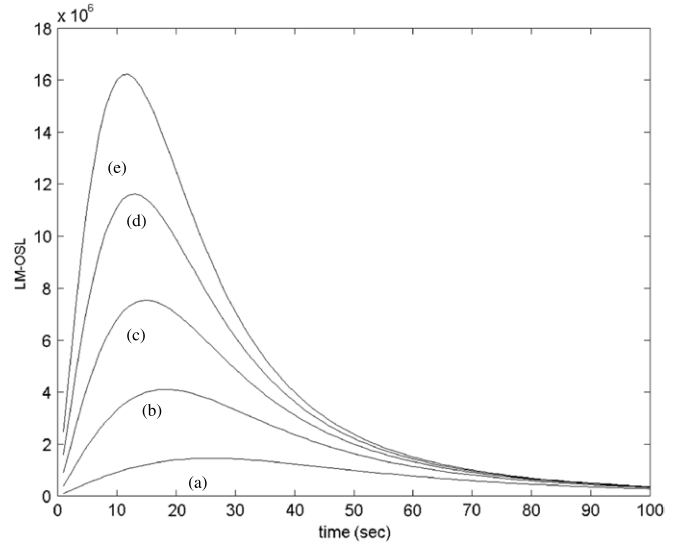


Figure 1. Simulated conventional LM-OSL curves. The parameters used are given in the text. The ratios of the values of n_0 in curves (a)–(e) are 1 : 2 : 3 : 4 : 5.

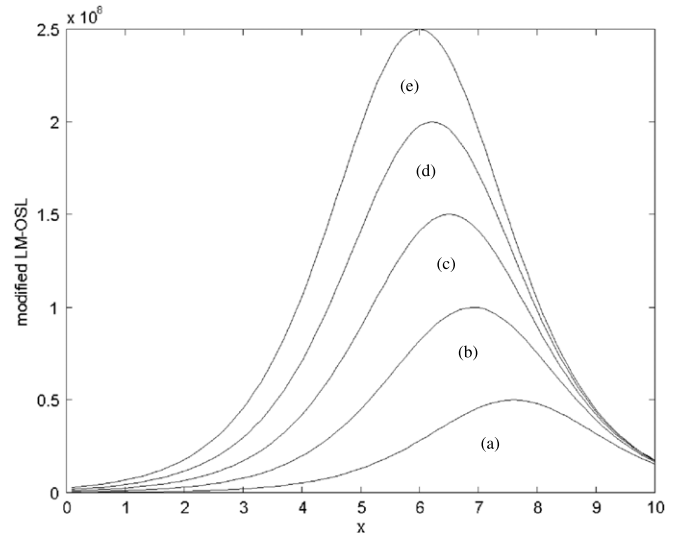


Figure 2. Simulated LM-OSL in the new presentation (see text). The parameters used are the same as in figure 1, and so are the values of n_0 .

6. Conclusions

In this work, it has been shown that a unified presentation can be given to different luminescence curves. These include TL measured under hyperbolic heating function, TL-like phosphorescence, TL-like OSL decay as well as LM-OSL and pseudo-LM-OSL. In all these phenomena, the results can be presented in the same way; examples are equation (20) for first-order kinetics and equation (27) for general-order kinetics with equation (28) being the special case for $b = 2$. The main importance of this presentation is that it transforms a featureless decay curve into a peak-shaped curve, the maximum of which is proportional to n_0 , the initial concentration of carriers which, under the appropriate conditions, carries the information of the previously administered dose. Of course, it is much easier

to determine accurately such a maximum value than the area under a featureless decay curve. Also, the area under the defined $y(x)$ curve is shown to be proportional to n_0 . In addition, the use of the mentioned figure from Chen and Kristianpoller (1986) can yield in all these phenomena the effective kinetic-order b due to the one-to-one correspondence between the symmetry factor μ_g and the order of kinetics.

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