A Modification of the Kinetic Equations Used for Describing the Thermoluminescence Phenomenon

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In the present work we discussed the validity of the traditional first-, second- and general-order kinetic equations used to describe the thermoluminescence (TL) phenomenon. These equations were written in their present forms to explain the TL glow peaks at constant value of the heating rate. However, a widespread mistake was found when one uses these equations to explain the TL at different heating rates. This mistake is a result of unreal definition of the rate of change of trapped carriers as a function of temperature. A modification of the above-mentioned equations has been considered during this work to define correctly the TL phenomenon. As a result of this modification some characteristics of the TL glow peaks, namely, the TL intensity and the total integral will be changed. Following to the present work, an emendation of Chen–Winer method used to determine the activation energy of TL glow peak was considered during this work. Also, a modification of the equation used to determine the relative value of initial concentration of the trapped carriers was considered during this work.

INTRODUCTION

Thermoluminescence (TL) is the emission of light from an insulator or semiconductor as it is heated and which is the result of previous exposure to ionizing radiation at low temperature. A typical TL measurement, made by increasing the sample temperature linearly, results in a curve of light intensity versus sample temperature. It is usually called a glow curve and contains one or more glow peaks. Randall–Wilkins, Garlick–Gibson and many others gave analytical descriptions of the glow peak for the three cases usually referred to as first-, second- and general-order TL kinetics, respectively [1–4]. The shape, position and intensity of the glow peaks are related to a various trapping parameters of the trapping states responsible for the TL emission. These parameters include the order of kinetics (b), the activation energy E (eV)

and the frequency factor S (s^{-1}) or the pre-exponential factor S (s^{-1}). Also, as it is seen in the present work, the shape, position and intensity of the glow peaks are related to the heating rates β (K s^{-1}) used for obtaining these glow peaks.

The relationship between the heating rate β and the temperature corresponding to the maximum intensity (T_m) has generated a number of methods for calculating the activation energy E (eV) by noting the change in T_m for a certain change of β [5–7], and/or by noting the changes in the peak height (I_m) and T_m for certain change of β, [8]. However, during the studies on these relations, one may found critical discrepancies between the characteristics of the TL glow peaks generated by using the first, second or general-order kinetics and the theoretical or experimental results obtained by many authors when they used different heating rates [1,9–26]. As we can see in the next section, the differential equations used to describe the rate of change of the trapped carriers with increasing the temperatures, which describe the intensity of TL glow peak in the case of first-, second- and general-order kinetics were not

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correctly defined in the past. Therefore, in the present work we try to modify these equations to be able to describe correctly the TL phenomenon and to distinguish between the TL and phosphorescence phenomena. As a result of this modification, we study here the glow peak characteristics. These characteristics include the peak height (I_m), peak area, the temperature corresponding to the maximum intensity (T_m) and the full width at the half maximum of the glow peak (FWHM).

THEORETICAL BACKGROUND OF THERMOLUMINESCENCE KINETICS

First-Order Kinetics

The theory of TL started with the introduction by Randall and Wilkins [1] of the first-order kinetic equation

$$I(t) = -\frac{dn}{dt} = n\,S\,\exp\left(-E/kT\right), \quad (1)$$

which shows the evaluation of $I(t)$, the intensity of emission with the time, where n (cm^{-3}) the electron concentration trapped at time $t(s)$ and k (eV/K) the Boltzmann's constant. Physically, Randall and Wilkins equation implicitly assumes that retrapping of the thermally released electrons from traps and their subsequent recombination with holes trapped in recombination centers is practically impossible. If the sample is heated up so that the temperature rises at a linear heating rate $\beta = dT/dt$ (K s^{-1}), then

$$dn/n = -(S/\beta)\exp\left(-E/kT\right)dT. \quad (2)$$

On integration of Eq. (2), one obtains

$$n(T) = n_0\,\exp\left[-(S/\beta)\int_{T_0}^{T}\exp\left(-E/kT'\right)dT'\right], \quad (3)$$

where n_0 (cm^{-3}) is the concentration of traps populated at the starting heating temperature T_0 (K). The temperature dependence of the emitted TL is given by [1]:

$$I(T) = n_0\,S\,\exp\left(-E/kT\right)$$
$$\times \exp\left[-\frac{S}{\beta}\int_{T_0}^{T}\exp\left(-\frac{E}{kT'}\right)dT'\right], \quad (4)$$

where n_0 (cm^{-3}) is the concentration of traps populated at the starting heating temperature T_0 (K). The position of the maximum can be determined by obtaining the derivative of Eq. (4) with respect to the temperature and equating it to 0, which yields:

$$\beta E/kT_m^2 = S\,\exp\left(-E/kT_m\right). \quad (5)$$

Second-Order Kinetics

Garlick and Gibson [2] considered the case of strong retrapping, which brought about the second-order kinetics:

$$I(t) = -\frac{dn}{dt} = \frac{n^2}{N}S\,\exp\left(-E/kT\right), \quad (6)$$

where N (cm^{-3}) is the trap concentration. The equation describing the TL intensity for second-order glow peak is given by [2]:

$$I(T) = \frac{n_0^2 S\,\exp\left(-E/kT\right)}{N\left[1 + \left[\frac{n_0 S}{N\beta}\right]\int_{T_0}^{T}\exp\left(-\frac{E}{kT'}\right)dT'\right]^2}. \quad (7)$$

The condition of the maximum of second-order glow peak is obtained by equating the derivative of Eq. (7) to 0, thus:

$$1 + (n_0/N)(S/\beta)\varphi = \frac{2n_0 S k T_m^2}{N\beta}\exp\left(-E/kT_m\right), \quad (8)$$

where

$$\varphi = \int_{T_0}^{T_m}\exp\left(-E/kT'\right)dT'. \quad (9)$$

General-Order Kinetics

Halperin and Braner [27] adopted a set of three simultaneous differential equations to describe the TL emission. This set of equations deals with the traffic of charge carriers during the heating of the sample, when one trapping state and one kind of recombination center are involved. These equations are:

$$I(t) = -\frac{dm}{dt} = A_m\,m\,n_c, \quad (10)$$

$$-\frac{dn}{dt} = n\,S\,\exp\left(-E/kT\right) - A_n n_c(N - n), \quad (11)$$

$$\frac{dm}{dt} = \frac{dn}{dt} + \frac{dn_c}{dt}, \quad (12)$$

where m (cm^{-3}) is the concentration of holes in centers, n (cm^{-3}) that of electrons in traps, N (cm^{-3}) the trap concentration, n_c (cm^{-3}) the concentration of free electrons in the conduction band, and A_m, A_n (cm^3 s^{-1}) the probabilities of recombination and retrapping, respectively.

Although numerical solutions of the set of three simultaneous differential Eqs. (10)–(12) have been carried out for given set of eight parameters, namely, S, E, A_m, A_n, N, m_0, n_0, and n_{co} (initial values of m, n, n_c), [10,17,28], the availability of such solutions has not been of much help in the more inverse problem, of analyzing a given glow curve to derive the values of the parameters from it.

To deal with this problem, early investigators assumed only first- and second-order kinetics, but later May and Partridge [29,30] and others have proposed an empirical equation to describe the TL glow peak when conditions for neither first-order nor second-order are satisfied. This equation is known as the general-order kinetics and its final form has been suggested as [4]:

$$I(t) = -\frac{dn}{dt} = \frac{n^b}{N^{b-1}} S \exp(-E/kT), \qquad (13)$$

where (b) is the order of kinetics. Usually (b) is assumed to be between 1 and 2, but some times can exceed this range [3]. It is ought to mention here that, the first- and second-order kinetics Eqs. (1) and (6), can be derived with certain simplifying assumptions from the rate equations describing the charge carrier traffic in a one-trap and one-centre model, the general-kinetics Eq. (13) stems from analogy of the mathematical structure of first- and second-order kinetics. The solution of Eq. (13) for $b \neq 1$ is given by [4]:

$$I(T) = \frac{n_0 S'' \exp(-E/kT)}{\left[1 + \left[\frac{(b-1)S''}{\beta}\right] \int_{T_0}^{T} \exp(-E/kT')dT'\right]^{b/(b-1)}}, \qquad (14)$$

where

$$S'' = S(n_0/N)^{b-1}. \qquad (15)$$

Similarly, the position of the maximum of general-order glow peak is obtained from Eq. (14) as [31]:

$$1 + (b-1)(n_0/N)^{b-1}(S/\beta)\varphi = S(bkT_m^2/\beta E)(n_0/N)^{b-1}$$
$$\times \exp(-E/kT_m). \qquad (16)$$

THE BASIS OF THE PRESENT WORK

It is worthwhile noting that our discussion is abbreviated here for the first-order kinetics. However the general concept followed in the case of first-order is also valid in the case of second- and general-order kinetics. According to Randall and Wilkins [1], the intensity of the glow I is proportional to the rate of supply of electrons to the luminescence centers,

$$I(t) = -\frac{dn}{dt} = n S \exp(-E/kT). \qquad (17)$$

As we have mentioned above, TL is defined as the emission of light from an insulator or semiconductor as it is heated after exposure the material to ionizing radiation at low temperature. Then increasing the sample temperatures usually makes the typical TL measurement. From

the definition of TL, Eq. (17) doesn't include any term, which shows a change of the TL emission with changing the temperature T. This means that, Eq. (17) gives the time-dependent intensity of emitted light from insulating material at constant temperature. Therefore, Eq. (17) in its present form is adequate only to describe the phosphorescence decay. This also means that Eq. (17) in its form is not adequate to describe the TL intensity. Not only that but also, Randall and Wilkins in their next paper [32], supposed that Eq. (17) might be used to describe the intensity of phosphorescence due to traps of depth E. This means that phosphorescence decay may be considered as TL at a heating function of $T(t) = $ constant. Actually, Randall and Wilkins [32], integrated Eq. (17) to obtain the intensity of phosphorescence decay at constant temperature due to a single trap level as follows:

$$I(t) = I_0 \exp\left[-St \exp\left(-\frac{E}{kT}\right)\right], \qquad (18)$$

where, I_0 is the initial intensity of phosphorescence at time $t = 0$, for a given temperature T. Now, it is difficult to understand that Eq. (17) can be used, basically, to describe the intensity of two anti-meaning phenomena. One of them the TL (Eq. (14)), where the intensity is temperature-dependent parameter and the second one is the phosphorescence (Eq. (18)), where the intensity is temperature-independent parameter. It should be noted here that once a certain heating function $T = T(t)$ is chosen in the experiment to obtain TL glow curve, $I(t)$ on the left-hand side of Eq. (17) cannot be replaced by $I(T)$. The evidence of that Eq. (17) doesn't include heating function to describe, basically, the TL intensity. Therefore, I believe that Eq. (17) in its present form is not adequate to describe the TL intensity where $T(t) \neq$ constant and must be modified. This also means that Eq. (4) is not correctly adequate to describe the TL intensity where $T = T(t)$ and should be modified. To be used to describe correctly the TL intensity Eq. (17) should be rewritten by replacing $I(T)$ instead of $I(t)$. This can be done by considering heating function β (K s^{-1}) where $\beta = dT/dt$. Then Eq. (17) may be modified as:

$$I(t) = -\frac{dn}{dT}\frac{dT}{dt} = n S \exp(-E/kT). \qquad (19)$$

Then,

$$I(t) = -\frac{dn}{dT}\beta = n S \exp(-E/kT). \qquad (20)$$

Finally,

$$I(T) = -\frac{dn}{dT} = \frac{n S}{\beta} \exp(-E/kT). \qquad (21)$$

Similarly, Eqs. (6) and (13) may be modified as:

$$I(T) = -\frac{dn}{dT} = \frac{n^2}{N\beta} S \exp(-E/kT). \qquad (22)$$

$$I(T) = -\frac{dn}{dT} = \frac{n^b}{\beta N^{b-1}} S \exp(-E/kT). \qquad (23)$$

As a result of these modifications, Eq. (4), which describes the TL intensity for first-order kinetics is rewritten as:

$$I(T) = \frac{n_0 S}{\beta} \exp(-E/kT)$$
$$\times \exp\left[-\frac{S}{\beta} \int_{T_0}^{T} \exp\left(-\frac{E}{kT'}\right) dT'\right]. \qquad (24)$$

Also, the equation describing the TL intensity for second-order glow peak, Eq. (7) is rewritten as:

$$I(T) = \frac{N_0^2 S \exp(-E/kT)}{N\beta \left[1 + \left[\frac{n_0 S}{N\beta}\right] \int_{T_0}^{T} \exp\left(-\frac{E}{kT'}\right) dT'\right]^2}. \qquad (25)$$

The empirical Eq. (14), which describes the general-order kinetics, may be rewritten as:

$$I(T) = \frac{n_0 S'' \exp(-E/kT)}{\beta \left[1 + \left[\frac{(b-1)S''}{\beta}\right] \int_{T_0}^{T} \exp(-E/kT') dT'\right]^{b/(b-1)}}. \qquad (26)$$

It is worthwhile noting that, in spite of the above-mentioned modifications, Eqs. (5), (8) and (16) which describe the condition of the maximum intensities of first-, second- and general-order glow peak still hold true even after these modifications.

RESULTS AND DISCUSSION

The main differences between the original first-, second- and general-order kinetic equations, namely Eqs. (4), (7) and (14) and the modified first-, second- and general-order kinetic equations, namely Eqs. (24)–(26), respectively, are restricting in the shape characteristics of the glow peak when one uses different heating rates.

Figures 1 and 2 show glow peaks are generated at different heating rates in the case of general-order of kinetics using Eqs. (14) and (26), respectively. From these figures, it is clear that the temperature (T_m) corresponding to the maximum intensity of the glow peaks shifts to the higher side of temperatures with increasing the heating rate. The shifts are identical in both cases. This is not surprising since in spite of the above-mentioned modification, Eq. (16), which describe the condition of the max-
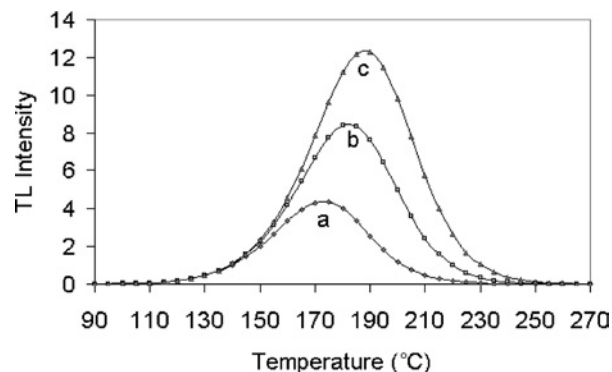

Fig. 1. General-order glow peaks computed according to Eq. (14) with $b = 1.5$, $n_0 = N$, $S = 5 \times 10^{12}$ s^{-1} and $E = 1.2$ eV. The heating rates β are given as: (a) 2, (b) 4 and (c) 6 ($^\circ$C s^{-1}).

imum of general-order glow peak, still hold true before and after modification. This is in contrast with previous works [11,14,22], which show peaks shift to the lower degree of temperatures. Again, the shift to the higher degrees of temperature with increasing the heating rates is expected if one takes into account the condition assumed in Eq. (16).

Also, Fig. 1 shows an enhancement increase of the height (I_m) of the glow peaks with increasing of the heating rates. In contrast to Fig. 1, Fig. 2 shows an apparent decrease of the height (I_m) of the glow peaks as the heating rates (β) increase.

Figure 1 shows that the area under the glow peaks increase as the heating rates (β) increase. However, Fig. 2 shows stability of the area under the glow peaks independent of the heating rates (β). Figures 3 and 4 show the evaluation of the peak area and peak height as a function of the heating rate according to Eqs. (14) and (26),
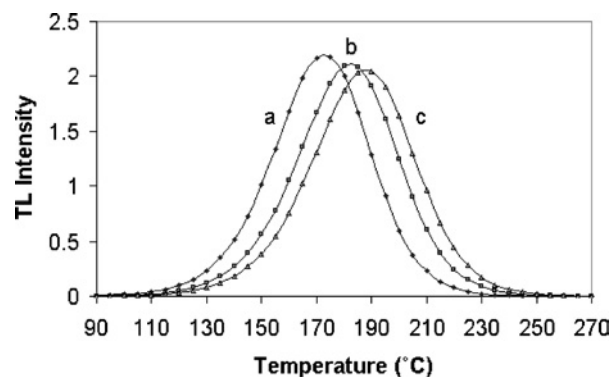

Fig. 2. Modified general-order glow peaks computed according to Eq. (26) with $b = 1.5$, $n_0 = N$, $S = 5 \times 10^{12}$ s^{-1} and $E = 1.2$ eV. The heating rates β are given as: (a) 2, (b) 4 and (c) 6 ($^\circ$C s^{-1}).
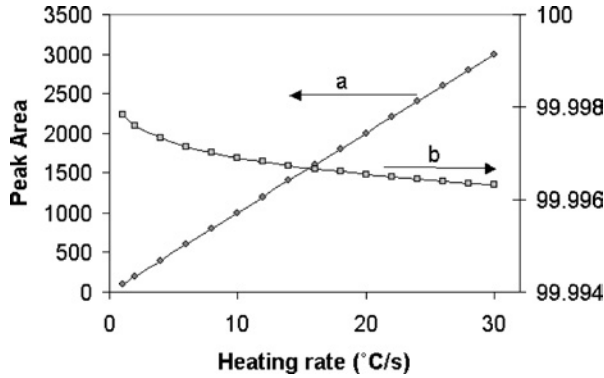
Fig. 3. The peak area as a function of the heating rate. Curve (a) according to Eq. (14) and curve (b) according to Eq. (26), respectively.
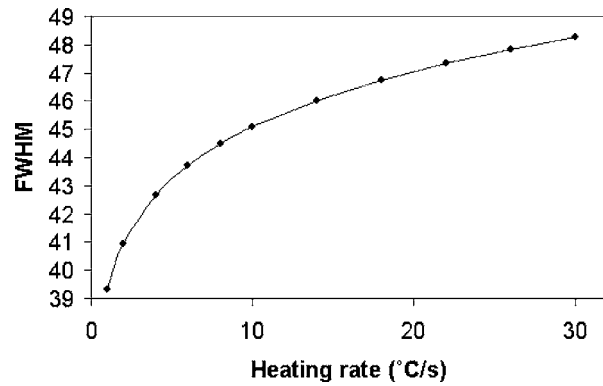

Fig. 5. The FWHM as a function of the heating rate according to Eqs. (14) and (26), respectively.

respectively. There is no physical reason to let the area increase with increasing the heating rate.

From Figs. 1 and 2, it is clear that the full width at the half maximum (FWHM) of the glow peaks increase with increasing the heating rates. It is worthwhile noting that the increase of FWHM is identical in the case of original and modified general-order equations. Figure 5 shows the values of FWHM as a function of the heating rate are depicted according to Eqs. (14) and (26), respectively.

As shown in Fig. 2, it can be easily observed that the qualitative behavior of the glow curves is in agreement with what it is expected from the modified kinetic models of TL; such as, the increase of the T_m and FWHM as well as the decrease of the peak height as the heating rate increases. According to the modified kinetic models of TL, since the test dose given is always the same, the glow curve integral must be stable and independent of the heating rate. On the other hand, the peak height must decrease and the increase of the FWHM as the heating rate increases due to the stability of the glow curve integral.
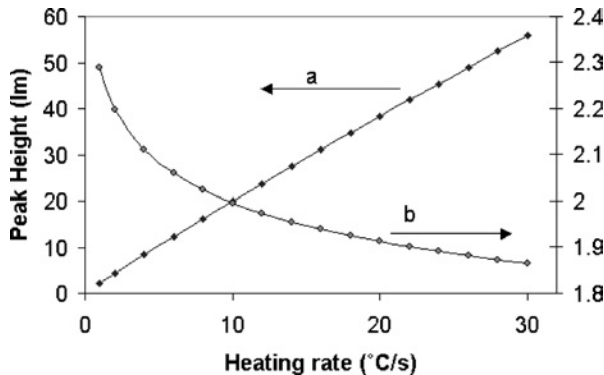

Fig. 4. The peak height as a function of the heating rate. Curve (a) according to Eq. (14) and curve (b) according to Eq. (26), respectively.

Generally, the results appearing here are in agreement with previous works [13,21–23,25,26], which show decreases of the peak height intensities (I_m) with increasing the heating rate and in contrast with previous work [1,9–12,14–20,24], which show increases of the peak height intensities (I_m) with increasing the heating rate. Also, as shown in Figs. 2–5, besides of the trap parameters b, E (eV) and S (s^{-1}), the shape, position and intensity of the glow peaks are related also to the heating rates β (K s^{-1}) used for obtaining these glow peaks.

The next point, which must be discussed here, is related actually to the methods that use the heating rate or/and that use the heating rate peak height intensity to obtain the activation energy E (eV) of TL glow peak. It is worthwhile noting that, the relationship between the heating rate β and the temperature corresponding to the maximum intensity T_m has generated a number of methods for calculating the activation energy E (eV) by noting the change in T_m for a certain change of (β). The most important methods in this field are those due to Booth–Bohun method [5,6] and Hoogenstraaten [7], which are used intensively for obtaining the activation energy E (eV) of TL glow peaks. These methods can most easily be introduced by studying the equation for the maximum intensity of first-order of kinetics, Eq. (5). Booth [5] and Bohun [6] working independently have used different heating rates to evaluate E basing their works on the variation of T_m with the heating rate β. Therefore, Solving Eq. (5) for two different heating rates (β_1 and β_2) gives

$$E = [(kT_{m_1}T_{m_2})/(T_{m_1} - T_{m_2})]$$
$$\times \ln[(\beta_1/\beta_2)(T_{m_2}/T_{m_1})^2], \qquad (27)$$

where, T_{m_1} and T_{m_2} are the temperature values corresponding to the maximum intensity for β_1 and β_2, respectively. Also, Hoogenstraaten [7], has used Eq. (5) and several

heating rates β to obtain a linear relation between $\ln(T_m/\beta)$ and $1/T_m$ as follows:

$$\ln\left(T_m^2/\beta\right) = (E/k)(1/T_m) + \ln(E/kS). \quad (28)$$

Thus the plotting between $\ln(T_m^2/\beta)$ versus $(1/T_m)$ giving rise of a straight line of slope (E/k), from which E is evaluated. As we heave mentioned before, Eq. (5) was not changed before or after the modification. As a result, Booth–Bohun and Hoogenstraaten methods still hold true even after this modification, but the meaning of the results in the two cases is completely different. In the case of modified first-order obtained by Eq. (8), the results of Booth–Bohun and Hoogenstraaten methods determine the activation energy E (eV). However, in the case of the original first-order obtained by Eq. (2), the results of Booth–Bohun and Hoogenstraaten methods determine a mislead activation energy E (eV). This means that the experimental results obtained by many authors [1,9,11,16,18–20,24], which show increase in the intensities and/or areas with increasing the heating rates must be reinvestigated to understand the reasons for these conflicts in the results.

On the other hand, Chen and Winer [8] have suggested another method to obtain the activation energy in the case of general-order glow peak by using different heating rate. It can be shown by inserting Eq. (16) into Eq. (14) that the maximum intensity of TL glow peak (I_m) is related to (T_m) at different values of (β) by:

$$\ln\left(I_m^{b-1}\left(T_m^2/\beta\right)^b\right) = \ln\left[(N^{b-1}/S)(E/bk)^b\right] + (E/k)(1/T_m), \quad (29)$$

which suggests that a plot of $\ln\left[I_m^{b-1}(T_m^2/\beta)^b\right]$ versus $1/T_m$ will give a straight line with slope (E/k), [8]. However, Eq. (29) should be amended according to the above-mentioned modification. Starting now from Eq. (23), then:

$$I(T) = -\frac{dn}{dT} = \left(\frac{S}{\beta N^{b-1}}\right)n^b \exp(-E/kT). \quad (30)$$

Taking the derivative of $I(T)$ with regard to T, one obtains:

$$\frac{dI}{dT} = \left(\frac{S}{\beta N^{b-1}}\right)\left\{(bn^{b-1})\frac{dn}{dT} + n^b\left(\frac{E}{kT^2}\right)\right\}\exp\left(-\frac{E}{kT}\right). \quad (31)$$

Then, we have for $T = T_m$:

$$\left.\frac{dI}{dT}\right|_{T=T_m} = \left(\frac{S}{\beta N^{b-1}}\right)\left\{\left[(bn^{b-1})\left.\frac{dn}{dT}\right|_{T=T_m}\right. \right. \\ \left. \left. + n^b\left(\frac{E}{kT_m^2}\right)\right]\exp\left(-\frac{E}{kT}\right)\right\} = 0 \quad (32)$$

Re-arranging gives:

$$(b\,n^{b-1})\left.\frac{dn}{dT}\right|_{T=T_m} + n^b\left(\frac{E}{kT_m^2}\right) = 0, \quad (33)$$

Now from Eq. (30) we can deduce:

$$-\left.\frac{dn}{dT}\right|_{T=T_m} = I_m, \quad (34)$$

and

$$n^b(T_m) = I_m(T_m)\left(\frac{\beta N^{b-1}}{S}\right)\exp(E/kT_m). \quad (35)$$

Inserting these two equalities into Eq. (33) yields:

$$I_m^{b-1}\left(T_m^2\right)^b/\beta = (N^{b-1}/S)(E/bk)^b\exp(E/kT_m). \quad (36)$$

Taking the logarithm of Eq. (36) gives:

$$\ln\left(I_m^{b-1}\left(T_m^2\right)^b/\beta\right) = \ln\left[(N^{b-1}/S)(E/bk)^b\right] + (E/k)(1/T_m). \quad (37)$$

Following Chen and Winer [8], the plot of $\ln[I_m^{b-1}(T_m^2)^b/\beta]$ versus $1/T_m$ will give a straight line with a slope of (E/k).

The last modification to be mentioned here is related to a method suggested to obtain the initial concentration of trapped electrons n_0. Previously, a simple analytical method has been developed to obtain the relative value of the initial concentration of trapped electrons n_0 (cm^{-3}) in case of general-order glow peak, by inserting Eq. (16) into Eq. (14). In this case an expression for the relative value of n_0 can be found [31]:

$$n_0 = \frac{I_m\exp(E/kT_m)}{S''}\left(\frac{bkT_m^2 S''}{\beta E\,\exp(E/kT_m)}\right)^{b/(b-1)} \quad (38)$$

In Eq. (38), the parameters b, E, S'' and β are known and the values of I_m, T_m can be obtained from the shape of the glow peak. Then the relative value of n_0 (cm^{-3}) can be obtained. This equation must be modified, by inserting Eq. (16) into Eq. (26) to form:

$$n_0 = \frac{\beta I_m\exp(E/kT_m)}{S''}\left(\frac{bkT_m^2 S''}{\beta E\,\exp(E/kT_m)}\right)^{b/(b-1)} \quad (39)$$

CONCLUSION

The heating rate as an important parameter related to the change of temperature during the TL run has not been considered correctly in the previous equations used to describe the TL kinetics. This paper presents modifications of the well-known first-, second- and general-order kinetic equations when various heating rates are used. According to the present modification, with increasing the heating rates one should observe stability of the area, decrease in the height, increase in the FWHM and shift of Tm to the higher degrees of temperatures. As a result of this modification, Chen–Winer method, which is used to determine the activation energy of TL glow peak, has been modified considered during this work. Also, the equation, which describes the initial concentration of the trapped electrons, has been considered during this work.

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