

CALCULATION OF PARAMETERS FOR THE MODEL OF AN IDEAL PHOSPHOR

by

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Equations for the calculation of kinetic parameters of thermoluminescent processes are theoretically derived for a model of an ideal phosphor. The values used in the calculation are obtained from glow curves and the function that describes the normalized glow curve generated. On the basis of this function, the equations for activation energy, frequency factor, and retrapping factor, were derived. All expressions are valid for a general case, when the filling factor of traps is $f_0 = 1$.

The concept of kinetics order was used for the calculation of parameters and the parameter of kinetics order was defined by means of real physical parameters. Results obtained by the analysis of synthetic curves and experimental glow curves of phosphor materials provide a deeper understanding of thermoluminescent kinetics.

Key words: thermoluminescence, kinetics order, glow curve, kinetics model, activation energy

INTRODUCTION

The energy band theory is the basis of the modern theory of solids. It offers an understanding and explanation of the important properties of thermoluminescent (TL) semiconductors and insulators. The important features of the behavior of TL materials can be correlated by means of a mechanism based on the energy of the band theory of solid semiconductors and insulators. For example, in the first phase after excitation, which is essentially an ionization of a luminescent center, the dissociated electron is very likely to be trapped and held localized in a discrete energy level within the forbidden region. Later on, the trapped electron recombines with the luminescent center and emits the characteristic luminescence only after it has been freed from this trapped state by thermal excitation.

The simplest physically possible process, which is described by a simple curve with one maximum, occurs in the case of phosphor that consists of one type of traps and one type of luminescent centers. This model is known as the OTOR model (One Trap-One Recombination center) [1] or the model of ideal phosphor [2].

The net rate of change in conduction band electron concentration n_c [cm^{-3}] is the thermal generation rate $sn \exp(E/kT)$ minus the recombination rates $\gamma_t n_c n$ and $\gamma_1 n_c p_1$

$$\frac{dn_c}{dt} = sn \exp \frac{E}{kT} - \gamma_t n_c (N - n) - \gamma_1 n_c p_1 \quad (1)$$

where in time t [s], n [cm^{-3}] is the electron trap concentration, N [cm^{-3}] – the concentration of traps, p_1 [cm^{-3}] – the concentration of empty luminescent centers, s [s^{-1}] – the frequency factor; E [eV] – the energy depth of a single active trap, T [K] – the material temperature, k [eV K^{-1}] is the Boltzmann constant, where γ_t [$\text{cm}^3 \text{s}^{-1}$] and γ_1 [$\text{cm}^3 \text{s}^{-1}$] are the recombination probabilities between the conduction band electrons and the traps or the luminescent centers, respectively.

Assuming a very short life time of conduction band electrons, we can write relations: $|dn_c/dt| \ll |dn/dt|$ and $|dn_c/dt| \ll |dp_1/dt|$. Then, due to the charge neutrality requirement, the concentration of trapped electrons n is equal to the concentration of empty luminescent centers p_1 . The dependence of the radiative recombination rate or TL intensity I_{TL} [$\text{cm}^{-3} \text{s}^{-1}$] on the conduction band electron concentration n_c and concentration of empty luminescent centers p_1 is described by the formulas

$$I_{\text{TL}} = \frac{dp_1}{dt} = \frac{dn}{dt} = \frac{\gamma_1 n_c p_1}{\gamma_1 n + \gamma_t (N - n)} n^2 \quad (2)$$

If we take that $r = \gamma_t/\gamma_1$ is the retrapping factor, the differential equation describing the model of ideal phosphor is given as [2, 3]

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$$I_{TL} \frac{dn}{dt} = \frac{s \exp \frac{E}{kT} n^2}{n r_t (N - n)} \quad (3)$$

An approximation, describing the quasi-stationary equilibrium of electrons in the conduction band is used to obtain the equation of an ideal phosphor model. The approximation assumes that the rate of change of electron concentration in the conduction zone is much smaller than the speed of change of the electron in the traps (the consequences being that $dn_c/dt = 0$ and $p_1 = n$). These assumptions seem reasonable in many cases.

The solution of differential eq. (3) is a glow-curve function of three parameters (s , r_t and E) and initial conditions (heating rate of the sample and the initial trap filling).

The differential eq. (3) cannot be solved analytically. This problem has created difficulties in the analysis of the TL process and calculation of TL parameters. A reasonable solution was to use the general order kinetics model [4-6]. The application of the general order kinetics model is quite satisfactory in radiation dosimetry, but it cannot be used to characterize the material since it does not provide accurate data for the actual parameters of the physical process.

The value calculation of trap activation energy by the general order kinetics method is subject to error if the best fitted value of the kinetics order is found to be different from 1 and 2 [1]. The method based on the kinetics order has serious deficiencies: first, a fixed parameter to represent an evolving process and, secondly, instead of an experimental value, an assumed value which cannot correspond to reality [7].

A number of methods for the calculation of particular kinetic parameters of phosphors exist in literature. However, no calculation model or method is based on the differential eq. (3) which at the same time contains analytical solutions for all parameters.

The calculation model, known as the Initial Rise Method, uses the general characteristic of the TL process. Namely, that starting part of the glow curve is a part of exponential growths. This method uses only the starting part of the glow curve described by the differential eq. (3) to calculate activation energy values. This is the so-called analytical-graphical calculation method. In fact, eq. (3) in this method is used only to prove and affirm the general characteristic of the exponential phase of the TL process. Calculations of parameters based on other mentioned methods lack the kinetic analysis of processes with phosphors as a basis and are not comparable with the solution obtained directly from the differential eq. (3) for an ideal phosphor. The best and most accurate values for kinetic parameters of ideal phosphors could, consequently, be obtained only by solving the differential eq. (3). In this report, we demonstrate the method for the solution of the equation which gives the most accurate calculation of kinetic parameters of activation energy, frequency

parameters, and retrapping factors. To the best of our knowledge, our method offers an accurate calculation of parameters not only by describing the glow curve, but the TL process itself as well. At the same time, our method offers exact calculations of kinetic parameters obtained from the experimental glow curve while following the changes in parameters with the changes in starting conditions.

KINETICS OF AN IDEAL PHOSPHOR MODEL

TL emission is a result of the heating of phosphor. In case of a constant heating rate R [K/s], temperatures T [K] are given by $T = Rt + T_0$, where t [s] is time and T_0 (K) is the initial value of TL material temperature. Then, the emission intensity of thermoluminescence is

$$I = \frac{1}{R} \frac{dn}{dt} = \frac{1}{R} \frac{s \exp \frac{E}{kT} n^2}{n r_t (N - n)} \quad (4)$$

The integral of the function of the TL glow curve for the temperature interval from zero to infinity equals n_0 by definition. Parameter n_0 is the initial electron trap concentration. The initial value of temperature can be zero, because the value of the part of the integral in the range of 0 to T_0 always equals zero.

First, a new parameter w_{eff} [cm^{-3}] is defined. Parameter w_{eff} is the integral of the normalized TL curve i_N from initial until the end of TL emission [4]. Parameter w_{eff} [cm^{-3}] can be defined as

$$w_{\text{eff}} = \frac{n_0}{I_m} \int_0^{\infty} i_N dT \quad (5)$$

where I_m is the maximum of TL intensity in the expression $I = I_m i_N$. In this case, I_m is a dimensionless parameter. From eq. (5), it is evident that I_m equals to one when $n_0 = w_{\text{eff}}$. The maximum value for the experimental TL function I_m is difficult to obtain; for practical calculations of kinetic parameters, normalized curves are used. Substitution $N = w_{\text{eff}}$ in eq. (4) gives the function

$$i = \frac{dn}{dT} = \frac{1}{R} \frac{s \exp \frac{E}{kT} n^2}{n r_t (w_{\text{eff}} - n)} \quad (6)$$

From this function, its normalized form i_N could be obtained by taking $n_0 = w_{\text{eff}}$. The integral of function (6) for heating temperature interval from zero to infinity equals w_{eff} , and, assuming that other parameters rest unchanged, the value of the function maximum equals 1. The function (6) has its maximum $I_m = 1$ only if $f_0 = 1$, for other values of f_0 , the function maximum is less than 1 (meaning that the concentration of electrons in traps could not exceed the trap concentration). If the value of the trap filling at start is $f_0 < 1$, the integral in eq. (5) could be calculated and the starting concentration of electrons in traps is $n_0 = w$. Relation $N = w_{\text{eff}}$ also holds. On the basis of the eq. (5), it follows

$$w_{\text{eff}} = \frac{w}{I_{\text{mf}}} \int_0^{\infty} i_N dT \quad (7)$$

where I_{mf} is the maximum of TL intensity if $f_0 < 1$. From this equation it is obtained that $I_{\text{mf}} = f_0$, since $w/w_{\text{eff}} = n_0/N = f_0$.

Also, characteristic and important parameters in equations for kinetic parameters calculations are b_{eff} [cm^{-3}], b [cm^{-3}], and μ_s . Parameter b_{eff} is the integral of the normalized TL curve (6) from T_m until the end of TL emission for $f_0 = 1$ and is equal to the concentration of electrons in traps at the moment $T = T_m$ [4]

$$b_{\text{eff}} = \int_{T_m}^{\infty} i_N dT \quad (8)$$

For $f_0 < 1$, the same parameter is referred as b , and the relation $b/b_{\text{eff}} = f_0$ holds in that situation. Symmetry factor μ_s is obtained by dividing parameters b_{eff} and w_{eff} . It should be noted that the value for symmetry factor μ_s is always the same for the same phosphor entity, regarding different values of f_0 , so $b_{\text{eff}}/w_{\text{eff}} = b/w$. The normalization of the TL glow curve does not change this relationship [2, 4]

$$\mu_s = \frac{b}{w} = \frac{b_{\text{eff}}}{f_0 w_{\text{eff}}} \quad (9)$$

Differential eq. (6) can be solved by separation of variables

$$\frac{n}{n^2} r_t (w_{\text{eff}} n) dn = \frac{1}{R} s \exp \left(-\frac{E}{kT} \right) dT \quad (10)$$

When the kinetic process is monitored from the initial heating temperature $T_0 = 0$ ($n_0 = w = f_0 w_{\text{eff}}$) to the temperature of the maximum glow curve $T = T_m$ (the maximum concentration of electrons in traps is $n_m = b$), definite integrals are obtained

$$\int_{f_0 w_{\text{eff}}}^b \frac{n}{n^2} r_t (w_{\text{eff}} n) dn = \frac{1}{R} s \int_0^{T_m} \exp \left(-\frac{E}{kT} \right) dT \quad (11)$$

This equation can be rearranged and tabular integrals obtained by the left-hand expression

$$(1 - r_t) \int_{f_0 w_{\text{eff}}}^b \frac{dn}{n} = w_{\text{eff}} r_t \int_{f_0 w_{\text{eff}}}^b \frac{dn}{n^2} = \frac{1}{R} s \int_0^{T_m} \exp \left(-\frac{E}{kT} \right) dT \quad (12)$$

followed by

$$(1 - r_t) \ln \frac{f_0 w_{\text{eff}}}{b} = w_{\text{eff}} r_t \left[\frac{1}{b} - \frac{1}{f_0 w_{\text{eff}}} \right] = \frac{1}{R} s \int_0^{T_m} \exp \left(-\frac{E}{kT} \right) dT \quad (13)$$

The integral on the right side of the expression (13) is known as the integral of the Boltzmann factor. For an integral of this type there is no exact analytical solution, but a number of approximations could be successfully used: Frank-Kamenetskii approximation [8]

$$\int_0^{T_m} \exp \left(-\frac{E}{kT} \right) dT = \frac{kT_m}{E} \exp \left(-\frac{E}{kT_m} \right) \quad (14)$$

or, a more accurate Gorbachev approximation [8, 9]

$$\int_0^{T_m} \exp \left(-\frac{E}{kT} \right) dT = \frac{T_m}{E} \exp \left(-\frac{E}{kT_m} \right) \quad (15)$$

Based on eq. (13), and by using the approximation (15), the equation for calculating the value of the frequency factor is obtained

$$s = R \frac{E}{kT_m^2} \frac{2}{T_m} \exp \left(-\frac{E}{kT_m} \right) (1 - r_t) \ln \frac{1}{\mu_s} = \frac{r_t}{f_0} \frac{1}{\mu_s} \quad (16)$$

By replacing the term $\Delta = 2kT_m/E$ in the above equation we obtain

$$s = \frac{RE}{kT_m^2} \exp \left(-\frac{E}{kT_m} \right) (1 - r_t) \ln \frac{1}{\mu_s} = \frac{r_t}{f_0} \frac{1}{\mu_s} (1 - \Delta) \quad (17)$$

When the resulting expression (17) is substituted into (6), the equation describing the glow curve of the ideal phosphor model is finally obtained

$$i = \frac{dn}{dT} = \frac{E}{kT_m^2} \exp \left(-\frac{E}{kT} \right) \frac{E}{kT} (1 - \Delta) (1 - r_t) \ln \frac{1}{\mu_s} = \frac{r_t}{f_0} \frac{1}{\mu_s} \frac{1}{n} \frac{n^2}{r_t (w_{\text{eff}} n)} \quad (18)$$

EQUATIONS FOR THE CALCULATION OF TL PARAMETERS

The first derivative of the model function of ideal phosphors (6) is one of the equations that connect the kinetics parameters [10]. The position of the glow curve maximum is at the point where the first derivative equals to zero. By differentiating eq. (6) we obtain

$$\frac{di}{dT} = \frac{1}{R} \frac{d}{dT} \frac{s \exp \left(-\frac{E}{kT} \right) n^2}{n r_t (w_{\text{eff}} n)} \quad (19)$$

The first derivative of the function (6) is obtained by the differentiation of the complex function

$$\frac{di}{dT} = \frac{1}{R} \frac{1}{[n r_t (w_{\text{eff}} n)]^2} \frac{d}{dT} s \exp \left(-\frac{E}{kT} \right) n^2 [n r_t (w_{\text{eff}} n)] = \frac{d}{dT} [n r_t (w_{\text{eff}} n)] s \exp \left(-\frac{E}{kT} \right) n^2 \quad (20)$$

After performing the differentiation, the following expression is obtained

$$\frac{di}{dT} \frac{1}{R} \frac{1}{[n - r_t (w_{\text{eff}} - n)]^2} - \frac{E}{kT} - 2ni \frac{E}{kT^2} n^2 \text{sexp} \frac{E}{kT} n^2 (i - r_t i) \quad (21)$$

The requirement that relation (21) equals to zero means that the numerator of this expression equals to zero. When the function (6) reaches its maximum, then $T = T_m$, $i = f_{0i} = f_0 [\text{cm}^{-3}\text{K}^{-1}]$ and $n_m = b$

$$[b - r_t (w_{\text{eff}} - b)] - 2bf_{0i} \frac{E}{kT_m^2} b^2 - b^2 [f_{0i} - r_t f_{0i}] = 0 \quad (22)$$

It should be noted that the variable i is expressed in $\text{cm}^{-3}\text{K}^{-1}$ units, so that f_{0i} is not dimensionless. After rearranging the expression (22), it is obtained

$$1 - r_t \frac{w_{\text{eff}}}{b} - r_t - 2 \frac{Eb}{f_{0i} kT_m^2} - 1 - r_t \quad (23)$$

Now, a new parameter δ_{eff} [K] is introduced. This parameter is called: part of the effective value of halfwidth from the right side of the glow curve and was defined by the following relation [4, 5]

$$\delta_{\text{eff}} = \int_{T_m}^{\infty} i_{\text{Nd}} dT \quad (24)$$

In eq. (24) i_{Nd} is the same function as i_{N} but dimensionless. More details relating to parameter δ_{eff} can be found in the following papers [4, 5]. For $f_0 < 1$, the same parameter called part of effective value of halfwidth from the right side of the glow curve is referred as δ . By comparing eqs. (8) and (24), it could be seen that parameters δ_{eff} and b_{eff} , then δ and b , are numerically identical but dimensionally different. It is evident for a factor $Eb/(f_{0i} kT_m^2)$ from eq. (23), that the following identity applies

$$\frac{1}{f_{0i}} \frac{E}{kT_m^2} b = \frac{1}{f_0} \frac{E}{kT_m^2} \delta = \frac{E\delta}{f_0 kT_m^2} \quad (25)$$

Based on eq. (25), eq. (23) could be rearranged. The value of activation energy may now be calculated from the rearranged expression (23)

$$E = \frac{f_0 kT_m^2}{\delta} \frac{f_0 \mu_s - r_t (2 - \mu_s f_0)}{f_0 \mu_s - r_t (1 - \mu_s f_0)} \quad (26)$$

As previously discussed, $\delta_{\text{eff}} = \delta/f_0$. Now, a new parameter is introduced, defined as

$$l = \frac{E\delta_{\text{eff}}}{kT_m^2} \quad (27)$$

We call this parameter kinetics order l , as it is defined with the same relation which was earlier obtained for first-order kinetics, second-order kinetics, mixed-order kinetics and general-order kinetics models [4-6, 11-13]. It should be noted that parameter l , defining the kinetic order, is not defined through the model of general-order kinetics differential equation and is not connected to this model.

From eq. (26), the kinetics order parameter can be written as

$$l = \frac{f_0 \mu_s - r_t (2 - \mu_s f_0)}{f_0 \mu_s - r_t (1 - \mu_s f_0)} \quad (28)$$

TL intensity can be calculated at the point of maximum $T = T_m$, using eq. (18). It is obtained

$$f_{0i} = \frac{E}{kT_m^2} (1 - \Delta) (1 - r_t) \ln \frac{1}{\mu_s} - \frac{r_t}{f_0} \frac{1}{\mu_s} - 1 - \frac{b^2}{r_t (w_{\text{eff}} - b)} \quad (29)$$

As in the case of eq. (22), it should again be noted that the value for i has its dimensions $\text{cm}^{-3}\text{K}^{-1}$, giving dimensions to f_{0i} at the point of TL intensity.

Applying the definition of l (27) and identity (25), eq. (29) is written as follows

$$r_t = \frac{\mu_s f_0 [1 - \ln(\mu_s) l (1 - \Delta)]}{l (1 - \Delta) [\mu_s f_0 \ln(\mu_s) - 1 - \mu_s] (1 - \mu_s f_0)} \quad (30)$$

When parameter r_t from eq. (30) is substituted in eq. (28), the equation for the calculating kinetics order is obtained

$$l^2 (1 - \Delta) [1 - \mu_s \ln(\mu_s)] - l (1 - \Delta) [1 - \mu_s - 2 \ln(\mu_s)] - 1 = 0 \quad (31)$$

The attained equations show that the calculation of the kinetics order parameter does not depend on the filling factor of traps. It is, therefore, possible to calculate the value of activation energy from the data obtained from the normalized glow curve. However, the

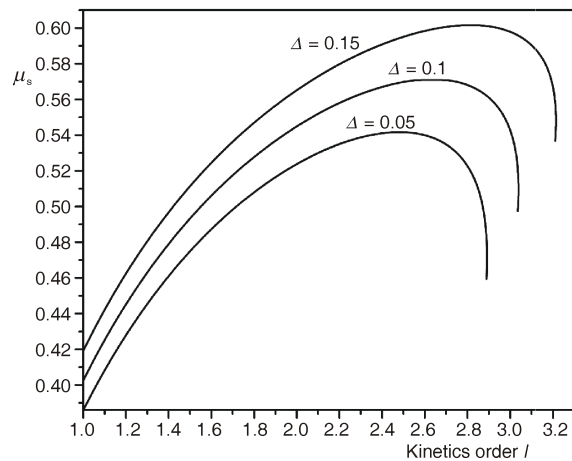


Figure 1. Dependence of the symmetry factor μ_s on the kinetics order l , for different values of Δ

exact values of the frequency factor and retrapping factor can, in general, be calculated only in the case when the value of the filling factor of traps is known. Equation (31) allows the calculation of the kinetics order parameter to depend on the symmetry factor μ_s and factor Δ (fig. 1).

Further replacing Δ , from eq. (27)

$$\Delta = \frac{2kT_m}{E} \frac{2\delta_{\text{eff}}}{T_m l} \quad (32)$$

in eq. (31), a quadratic equation for the analytical calculation of the value of kinetics order is obtained

$$l^2 [1 - \mu_s \ln(\mu_s)] - l \frac{2\delta_{\text{eff}}}{T_m} [1 - \mu_s \ln(\mu_s)] + \ln(\mu_s) - \frac{2\delta_{\text{eff}}}{T_m} [\mu_s - 1 - 2 \ln(\mu_s)] = 0 \quad (33)$$

Equation (29) can be written as

$$1 - \frac{r_t}{f_0} \frac{1}{\mu_s} - r_t - l(1 - \Delta) (1 - r_t) \ln \frac{1}{\mu_s} - \frac{r_t}{f_0} \frac{1}{\mu_s} = 1 \quad (34)$$

From (28) it is calculated

$$r_t = \frac{f_0 \mu_s (l - 1)}{2 - l - \mu_s f_0 (l - 1)} \quad (35)$$

and substituted in (34)

$$l(1 - \Delta) (1 - r_t) \ln \frac{1}{\mu_s} - \frac{r_t}{f_0} \frac{1}{\mu_s} = 1 \quad (36)$$

Using eqs. (36) and (27), the equation for the frequency factor (17) is given in the following form

$$s = \frac{R}{\delta_{\text{eff}} [2 - l - \mu_s f_0 (l - 1)]} \exp \frac{E}{kT_m} \quad (37)$$

In cases where $f_0 \ll 1$, when this value is three orders of magnitude smaller than one, *e. g.* $f_0 < 0.001$, it turns out that $(2 - l) \gg \mu_s f_0 (l - 1)$ and the following equation could be used for frequency factor calculation

$$s = \frac{R}{\delta_{\text{eff}} (2 - l)} \exp \frac{E}{kT_m} \quad (38)$$

At the same time, eq. (35) could be rearranged as

$$\frac{r_t}{f_0} = \frac{\mu_s (l - 1)}{2 - l} \quad (39)$$

In general, eq. (37) is one and includes solutions for both first and second-order kinetics. For $l = 1$, using eq. (27), eq. (37) is transformed in eq. (40) [14]

$$s = \frac{R}{\delta_{\text{eff}}} \exp \frac{E}{kT_m} - \frac{RE}{kT_m^2} \exp \frac{E}{kT_m} \quad (40)$$

representing the equation for calculating the frequency factor in a first-order kinetics model.

For $l = 2$, analogously, and with approximation used in second-order kinetics calculations, that symmetry factor is regarded as $\mu_s (1 + \Delta)/2$, so that one obtains

$$s^{(2)} = sf_0 \frac{R}{\mu_s \delta_{\text{eff}}} \exp \frac{E}{kT_m} - \frac{1}{2\mu_s} \frac{RE}{kT_m^2} \exp \frac{E}{kT_m} - \frac{RE}{kT_m^2} \frac{1}{(1 - \Delta)} \exp \frac{E}{kT_m} \quad (41)$$

Equation (41) is a well-known instrument for calculating the pre-exponential factor $s^{(2)}$ for second-order kinetics calculations [15].

Equation (28) describes all of the three well-known conditions governing the kinetics process for an ideal phosphor:

- The situation when the retrapping probability is insignificant: retrapping factor $r_t = 0$, and one obtains $l = 1$.
- The situation when the trap filling is small and there is retrapping: $f_0 = 0$, $r_t > 0$, and $l = 2$.
- The situation when the retrapping probability equals the probability of recombinant luminescence; retrapping value $r_t = 1$, and $l = 2$.

Equation (28) also describes the cases related to the high values of the retrapping factor, *i. e.* when $r_t (1 - f_0 \mu_s) \gg f_0 \mu_s$. In that case, the retrapping factor tends to infinity, $r_t \rightarrow \infty$. As symmetry factor values are about 0.5, f_0 values are in a 0 to 1 interval. It could thus be concluded that the maximal value for l is obtained for maximal f_0 , *i. e.* when $f_0 = 1$. At the same time, this is the final limit of the ideal phosphor model kinetics

$$I_{\text{max}} = \frac{2}{1} \frac{\mu_s f_0}{\mu_s f_0} \quad (42)$$

Equation (42) determines the maximal value of the preselected kinetics order Δ . From fig. 1, it can be seen that for $\Delta = 0.1$, the value $I_{\text{max}} = 3.04$ is obtained.

It turns out that, for high values of r_t , the value of the kinetics order parameter is independent of trap filling. Equation (31) (fig. 1), shows that kinetics order values have their maximal value for every single value of Δ . In practice, this means that in the vicinity of this maximal value even a high increase in the retrapping factor value has no influence on the kinetics order. Kinetics order values could not be higher than 3.2 for symmetry factor μ_s in the range $0.38 < \mu_s < 0.6$ ($\Delta = 0.15$).

The value of the retrapping factor, as seen from eq. (35), is very high and tends to infinity when $(2 - l) \mu_s f_0 (l - 1)$. In fact, in this situation, it is impossible to calculate parameter r_t from the shape of the glow curve.

In cases of high values of the retrapping factor in eq. (16), one can obtain

$$R = \frac{E}{kT_m^2} \frac{2}{T_m} \exp \frac{E}{kT_m} - \frac{1}{\mu_s} \left(1 - f_0 \ln \frac{1}{\mu_s} \right) s \frac{f_0}{r_t} \quad (43)$$

i. e., the pre-exponential factor s' is obtained as a quotient of frequency factor s and r_t/f_0 .

CALCULATION METHOD

For the calculation of kinetic parameters l , E , s , and r_t , the heating rate should be known, while other parameters are obtained from the normalized experimental glow curve. The kinetic order is calculated from eq. (33). For calculation of kinetic order l based on eq. (33), it is necessary to normalize the experimental glow curve by putting $I_m = 1$, and then determine the value T_m of the maximum temperature and integrals w_{eff} and $b_{\text{eff}}(\delta_{\text{eff}})$. The w_{eff} value is obtained from eq. (5) by the integration of the normalized experimental glow curve from starting temperature T_0 to the temperature at which all traps are empty. The integral b_{eff} and δ_{eff} is obtained from (8) and (24), respectively, by integration of the normalized experimental glow curve from maximum temperature T_m to the temperature at which all traps are empty. It is clear that both of these values are identical.

The value of symmetry factor μ_s is calculated from (9) as relation $b_{\text{eff}}/w_{\text{eff}}$. Parameter w_{eff} is obtained by the numerical integration of the normalized glow curve. There are a number of methods of numerical integration; in this report, the Romberg method was used. By solving quadratic eq. (33), two values for kinetic order l are obtained.

Of the cases presented in fig. 1, sometimes two real solutions are obtained. When so, one should calculate the parameters for both situations and take the solution which corresponds to the experimental glow curve. In most cases, only numerical results between 1 and 2 are taken into account.

The obtained kinetic order value l is further used for calculating activation energy E , according to eq. (27).

The values for retrapping factor r_t and frequency factor s could not be exactly calculated if the trap filling factor f_0 is not known. If f_0 is known, then r_t is calculated from eqs. (30) or (35), and s is calculated from eq. (37). Both eqs., (30) and (35), give the same results.

In practice, with thermoluminescent materials or dosimeters, the values of the trap filling factor are often very small. For $f_0 < 0.001$, the influence of the trap filling factor on the frequency factor could be ignored and the frequency factor could be calculated from eq. (38). The ratio r_t/f_0 obtained from eq. (39) is under the same restrictions.

RESULTS

The equations for calculating the kinetic parameters of a TL model for an ideal phosphor were tested on a number of computer-simulated glow curves. The values of parameters that characterize phosphors (r , E , s) and parameter f_0 , which determines the initial value of relaxation, were selected for the model of the ideal phosphor, shown by the differential eq. (6). For simplicity of calculation, it was taken that, in all cases, the heating rate of phosphors is $R = 1$ K/s. Parameter values were chosen so that the values of the kinetics order were between 1 and 2 and corresponded to the values that characterize phosphor in dosimetry. For certain characteristic cases, these values are shown in tab. 1. Simulated glow curves were obtained for parameter values given in tab. 1 and, using the numerical method of Runge Kutta IV order, for solving the differential eq. (6). Parameter values b_{eff} , w_{eff} , and μ_s were numerically calculated and are also shown in tab. 1. Romberg's method was used for the calculation of b_{eff} and w_{eff} integrals. The values of maximum temperatures were read from the glow curve and are shown in tab. 1.

Based on parameter values of $b_{\text{eff}}(\delta_{\text{eff}})$, μ_s and T_m from tab. 1, and by using the proposed procedure for calculation, it is possible to calculate the values of parameters of glow curves E , l , r , Δ and s . Table 2 shows the calculated values of these parameters, E_c , l_c , r_c , Δ_c , and s_c . The relative errors of calculation of parameters E , s , and r can be determined since their exact values are known. The exact values of parameters l and Δ are not known, so it is not possible to calculate their relative errors. The accuracy of individual parameters calculation can be estimated by analyzing the values of the relative error. The said analyses also provide an as-

Table 1. Parameter values for computer simulations of glow curves for the model of an ideal phosphor

No.	E [eV]	s [s^{-1}]	f_0	r	w_{eff} [cm^{-3}]	b_{eff} [cm^{-3}]	μ_s	T_m [K]
1	1.00	10^{12}	1	0.08	34.525	14.684	0.4253	383.871
2	0.10	10^5	1	0.18	17.516	8.4388	0.4818	86.1556
3	0.14	$3 \cdot 10^6$	1	0.3	17.094	8.4697	0.4955	96.7878
4	0.20	10^8	1	0.433	17.922	9.0800	0.5066	114.479
5	0.40	10^{10}	1	0.6	24.932	12.817	0.5141	184.696
6	0.20	10^8	0.5	0.2	17.503	8.6551	0.4945	115.441
7	0.20	10^8	0.1	0.04	17.403	8.5124	0.4891	116.112
8	0.20	10^8	0.01	0.004	17.391	8.4903	0.4882	116.254
9	0.20	10^8	0.001	0.0004	17.391	8.4888	0.4881	116.268

Table 2. Calculated values of glow curve parameters

No.	l_c	Δ_c	E_c [eV]	dE/E [%]	s_c [s^{-1}]	ds/s [%]	r_c	dr/r [%]
1	1.156	0.0662	1.000	$3.93 \cdot 10^{-4}$	$1.01 \cdot 10^{12}$	-0.75	0.07310	8.63
2	1.308	0.1498	0.099	0.86	$8.88 \cdot 10^4$	11.15	0.17655	1.91
3	1.462	0.1197	0.139	0.47	$2.78 \cdot 10^5$	7.58	0.29844	0.52
4	1.605	0.0989	0.200	0.21	$9.59 \cdot 10^7$	4.05	0.43656	-0.82
5	1.737	0.0800	0.398	0.42	$8.99 \cdot 10^9$	10.06	0.58994	1.68
6	1.507	0.0995	0.200	$5.99 \cdot 10^{-3}$	$1.01 \cdot 10^8$	-0.63	0.20291	-1.45
7	1.467	0.1001	0.200	-0.10	$1.03 \cdot 10^8$	-3.43	0.04108	-2.70
8	1.460	0.1000	0.200	-0.13	$1.04 \cdot 10^8$	-3.47	0.00444	-3.54
9	1.459	0.1001	0.200	-0.13	$1.05 \cdot 10^8$	-4.51	$4.15 \cdot 10^{-4}$	-4.15

assessment of the validity of the method used for the calculation of parameters.

For a typical glow curve of dosimeter TLD700H ($^7\text{LiF}:\text{Mg, Cu, P}$) [16-19], the same procedure of calculation is applied, as well as the simulated glow curves. Values $w_{\text{eff}} = 21.871 \text{ cm}^{-3}$, $b_{\text{eff}} = 9.0078 \text{ cm}^{-3}$ ($\delta_{\text{eff}} = 9.0078 \text{ K}$), $\mu_s = 0.41187$, were calculated from the glow curve. The heating rate of phosphor was $R = 1 \text{ K/s}$. Finally, the values of the TL parameter were calculated $l_c = 1.15$, $E_c = 2.564 \text{ eV}$ and, assuming that $f_0 = 1$, the following values were obtained: $s_c^{(0)} = 7.05 \cdot 10^{25} \text{ s}^{-1}$ and $r_c^{(0)} = 0.06776$.

The requirement $f_0 = 1$ is never fulfilled in dosimetric application. In cases where $f_0 \ll 1$, when this value is three magnitude orders smaller than one, e. g. $f_0 < 0.001$, one can use the eq. (38) to calculate the frequency factor. In that case, the value of the retrapping factor depends directly on the filling factor of traps and can be calculated from eq. (39).

When eqs. (38) and (39) are used, the following values of parameters $s_c = 7.56 \cdot 10^{25} \text{ s}^{-1}$ and $r_c/f_0 = 0.07268$, are obtained. Based on these results and the above assumptions, it can be concluded that $r_c = 0.07268 f_0$, or $r_c < 0.07268 \cdot 10^{-3}$. This means that all

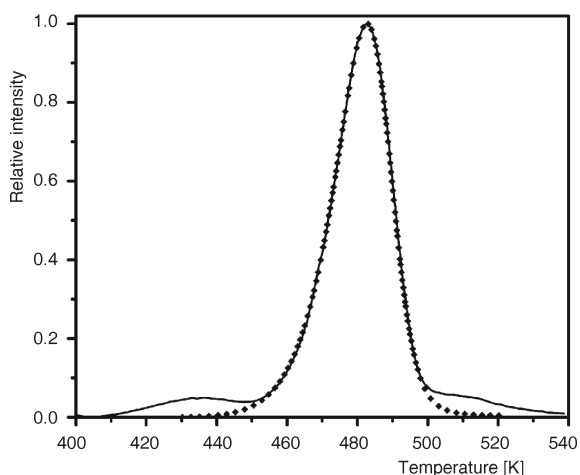


Figure 2. Experimental TL glow curve for a TLD700H dosimeter ($^7\text{LiF}:\text{Mg, Cu, P}$) (solid line) and glow curve obtained by calculation with the use of a model of an ideal phosphor (solid square)

glow curves with the above calculated values of parameters are almost identical.

Based on the calculated values of kinetic parameters of the process, it is possible to reconstruct the real physical process by using differential equation models and calculated parameters. The obtained parameter values $E = 2.564 \text{ eV}$, $f_0 = 0.001$, $s = 7.56 \cdot 10^{25} \text{ s}^{-1}$, and $r = 0.07268 \cdot 10^{-3}$ were substituted in the differential eq. (6). The differential equation was solved numerically with these parameters in order to obtain the glow curve. The results are shown in fig. 2. The figure shows that the calculated glow curve completely coincides with the experimental curve. It can thus be concluded that the dominant peak of the glow curve for TLD700H phosphor may be described by using the kinetics of the ideal phosphor model.

For a practical illustration of the TL curve kinetics parameter for calculating experimentally obtained values, the glow curve of TLD700H dosimeter was selected.

The measurement error for TL dosimeters depends on a number of factors. Besides the inherent instability of traps which boost the laboratory measurement error to about 2%, statistical uncertainty should also be calculated; it is usually less than 5% [20, 21]. So, the total expected measurement error for the TLD700H dosimeter is considered to be less than 10% [22].

The glow curve of the TLD700H dosimeter is convenient for demonstration of the calculation method because it has a single isolated dominant maximum resembling the ideal glow curve. Consequently, experimental and calculated glow curves revealed practical identity in shape.

Interestingly, the value of the frequency factor was unexpectedly high, something also noted in calculations based on other models.

It should be indicated that our proposed model for calculation has nothing in common with calculations based on the general kinetics order model of calculation. Namely, among equations based on general-order kinetics, the retrapping factor r_t does not exist at all, so there is no possibility for r_t to be calculated [6]. It has also been shown earlier that, based on the general-order kinetics model, the value of the fre-

quency factor s could not be calculated. Only the value of the pre-exponential factor $s^{(l)} = sf_0^{l-1}$ [4, 23] could be calculated. Accordingly, only values obtained in calculation of trap depth or activation energy E could be compared. This value is calculated by use of empirical interpolation-extrapolation Chen's equations [5, 23]; the exactness of calculation is, after all, reflected in the name of the method.

As an illustration, the values of activation energy are calculated in frames of interpolation-extrapolation equations for the emission curve at points 4 and 8 from tab. 1, and the following results, respectively, obtained: $E = 0.228$ eV (relative error 14%) and $E = 0.236$ eV (relative error 18%). As for pre-exponential factor values, the results are $s^{(l)} = 2.155 \cdot 10^9 \text{ s}^{-1}$ (relative error 20.55%) and $s^{(l)} = 3.31 \cdot 10^9 \text{ s}^{-1}$ (relative error 32.10%). For the first calculation, the value for f_0 was taken to be $f_0 = 1$, meaning that the pre-exponential factor value is the same as $s^{(l)} = s1^{l-1} = s$. However, in the second case, the value for f_0 was $f_0 = 0.01$ and, in final calculation, the relative error was greater for an order of magnitude. A simple comparison with our results reveals the qualitative and quantitative superiority of our method of parameter calculation.

DISCUSSION

The method for parameter calculation described in this report is based on the concept of kinetics order. In order to obtain the value of the kinetics order which would be correct for the ideal phosphor model, it is necessary to define this parameter in a new manner. This is done through the equation for calculating activation energy. This form of equations is known from models that have previously been investigated in detail (models of first, second, general and mixed-order kinetics) [1-6, 12-15, 23-25]. It has been assumed that the kinetics order parameter is not characteristic of the TL process, while the glow curve is. In this manner, the variation of the value of kinetics order parameter during TL relaxation is avoided. The parameter of kinetics order in the model of ideal phosphor has nothing in common with the model of general-order kinetics.

This concept allows simple equations for the calculation of parameters and a better understanding of the TL process.

Equations for the calculation of parameters E (32), r_t (30 and 35), and s (37) are obtained directly from differential eq. (6) which describes the ideal phosphor model. For known starting values of f_0 and heating rate R , all parameters of the differential equation, or TL kinetics determining the glow curve, could be calculated. The calculation method described above includes all parameters present in the differential equation and represent a complete algorithm. All equations for the calculation are derived from the basic differential equation and should, therefore, be re-

garded as precise. The requirement for calculation correctness is that the specimen has characteristics close to the ideal phosphor *i. e.*, that contains one kind of trap and one kind of center of luminescence.

Because of its accuracy, the described model could not be used as an universal model or a non-selective one for all phosphors. However, for the same reasons, the described model could be used for recognizing ideal phosphors.

For the ideal phosphor model, only the activation energy value E could be determined directly from the glow curve. To determine the values for parameters r_t and s , the trap filling factor f_0 should be known. However, this is not a significant obstacle as, for f_0 values starting from $f_0 < 0.001$, it is possible to calculate the frequency factor s (in reality, condition $f_0 < 0.001$ is generally fulfilled). If $f_0 < 0.001$, it is possible to obtain the equation for r_t/f_0 quotient calculation, eq. (39). This quotient is always the same for the determined kinetic order l and for all values $f_0 < 0.001$. This condition enables the calculation of all parameters of the differential eq. (6) which determine the TL curve. With the calculated relation r_t/f_0 , the value f_0 that satisfies $f_0 \ll 0.001$ could be arbitrarily chosen. For this particular value of f_0 , r_t is calculated, and both f_0 and r_t are inserted into eq. (6), resulting in a glow curve with the same shape for all values of r_t/f_0 and $f_0 < 0.001$. The relation r_t/f_0 is significant for determining the glow curve shape.

Detailed analysis reveals that the kinetics order value could be higher than 2, but not significantly. For values of factor Δ between 0.05 and 0.15, the kinetics order value is less than 3.2, meaning high trap filling ($f_0 > 0.01$).

Quadratic eq. (33) has two solutions. The possible solutions for parameter l compared to μ_s are presented in fig. 1. From fig. 1, it can be seen that the two solutions of (33) are real values near the function maximum. For values obtained for parameters T_m , μ_s , and δ_{eff} there are two different viable real values for l . To determine the proper value, obtained TL curves are compared to the experimental curve. The calculation curves for $T_m = 114.53$ K, $\mu_s = 0.528$, $b_{\text{eff}} = 10.27 \text{ cm}^{-3}$ ($\delta_{\text{eff}} = 10.27$ K), and $f_0 = 1$ are presented in fig. 3. The calculated curves are very similar. However, the solution on the right side of the curve maximum (fig. 1) gives higher values of halfwidth (FWHM- fullwidth at half maximum).

The calculation algorithm could be simplified if the trap filling is very small ($f_0 < 0.001$), so the l value could not be higher than 2. With this assumption, all solutions are on the left from the maximum of the function $\mu_s(l)$ in fig. 1. This assumption is almost always fulfilled in practice.

Results show that the parameters of simulated glow curves for the model of ideal phosphors can be calculated with great precision. The error, obtained by calculating the value of activation energy, is generally

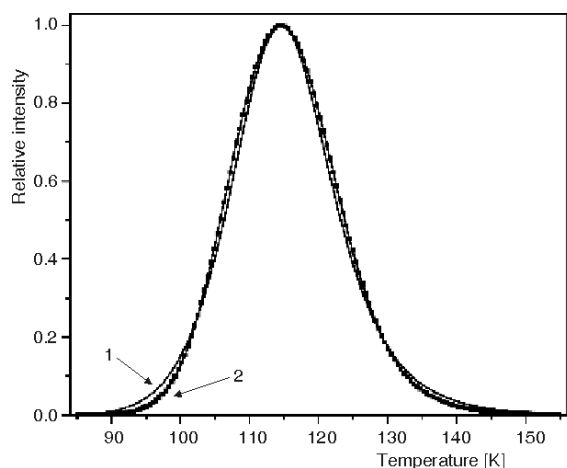


Figure 3. TL glow curves obtained from differential eq. (4) for following parameter values: $T_m = 114.53$ K, $\mu_s = 0.528$, $b_{\text{eff}} = 10.27 \text{ cm}^{-3}$ ($\delta_{\text{eff}} = 10.27$ K) and $f_0 = 1$. Obtained solutions are $E = 0.2$, $\Delta = 0.1$, $l = 1.82$ for curve 1, and $E = 0.314$, $\Delta = 0.063$, $l = 2.85$ for curve 2

below 1%. The resulting relative error of the frequency factor is the greatest because of the expression for its calculation that includes the exponential function. The largest relative errors are obtained for the values of factors $\Delta > 0.15$ and values $l \geq 2$, because increasing the value of parameter Δ causes a decrease in the accuracy of the approximation (15).

In practice, it is very difficult to find materials with an isolated TL peak which can be described by a model of an ideal phosphor. Closest to this model is the dominant peak that occurs during the processing of dosimeter TLD700H. Kinetic parameters are, in this case, often calculated by using the first-order kinetics model or the general-order kinetics model. Some authors, using the deconvolution method, proved that the shape of the dominant peak corresponds to the model of first-order kinetics. As small peaks overlap with the dominant peak in the deconvolution process, it is possible that such changes in shapes of small peaks will make the shape of the dominant peak to correspond to the first-order kinetics model. It has also been noted that deconvolutions with a general-order kinetics model attain better compatibility with the experimental curve. Since the model of general order kinetics is an approximation of the model of an ideal phosphor, better accuracy and compatibility with the experimental curve is expected when the model of the ideal phosphor is applied. Results obtained show that the model of an ideal phosphor faithfully describes the dominant peak, not only in relation to the asymmetry of the curve, but also to its shape.

CONCLUSIONS

A new method for calculating the relaxation of kinetic parameters in an ideal phosphor is developed.

The calculation results are close to real situations and correlated to the realistic physical parameters.

Equations for the calculation of kinetic parameters r_i , s , and E of TL processes are theoretically derived for the model of an ideal phosphor. The calculation method of TL relaxation parameters for the ideal phosphor is based on the kinetics order concept. The kinetics order is defined by an appropriate equation for models of the first, second, general and mixed-order kinetics. In this report, it has been proved that the same form of the equation can be used in the ideal phosphor model.

In our research, theoretically grounded on physical principles, the kinetics order is redefined as a parameter defining the glow curve instead of being a parameter that defines the TL process. The kinetics order parameter has a physical interpretation which is given by eq. (27) and corresponds to interpretations by the first and second order kinetics theory. Equation (27) is a general equation, theoretically defining the kinetics order parameter for an ideal phosphor model. The values of kinetics order parameters are calculated according to eq. (33). Their accuracy depends on the compatibility between experimental and theoretical glow curves.

Equation (27), which defines the kinetics order parameter of the ideal phosphor model, is identical in form with the equation which defines the kinetics order parameter for the general-order kinetics model. However, while being formally identical, some fundamental difference exists. The kinetics order parameter for the general-order kinetics model is calculated by the differential equation of the general-order kinetics model. This calculation method is often treated as a mathematical formalism because it cannot be successfully applied in a real physical model. At the same time, some significant parameters are not included in the general-order kinetics model. Nevertheless, the overall calculation is very precise within the limits of the general-order kinetics model. The main obstacle is that calculation does not correspond to any known real physical model describing TL kinetics. Therefore, the accuracy of calculation using this model is fundamentally inferior to the calculation based on the physical model, which we propose. We demonstrate it by comparing the results obtained by these two methods of calculation in parallel.

The results obtained prove that the calculation method for the model of the ideal phosphor may be used for calculating the targeted parameters with high accuracy. However, in order to obtain the most precise calculation results, accurate determination of T_m , b_{eff} , and w_{eff} parameters is necessary.

The values of physical parameters for a dominant peak of a TLD700H (7LiF: Mg, Cu, P) dosimeter are evaluated based on the method for parameters calculating the ideal phosphor model. The resulting glow curve coincided with the experimental curve. The obtained experimental glow curve corresponds to the theoretical model of ideal phosphor and the values of physical parameters describing the TL process can accurately be calculated.

AUTHORS' CONTRIBUTIONS

All authors have contributed to model analysis and the analyses and discussion of results. The manuscript was written and figures prepared by Z. M. Vejnović.

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ИЗРАЧУНАВАЊЕ ПАРАМЕТАРА ЗА МОДЕЛ ИДЕАЛНОГ ФОСФОРА

Теоријски су изведене једначине за израчунавање параметара кинетике термолуминисцентног процеса за модел идеалног фосфора. За прорачун су коришћене вредности параметара које су добијене из кривих исијавања и функција криве исијавања у нормализованом облику. На основу ове функције изведене су једначине за израчунавање активационе енергије, фреквентног фактора и фактора ретраповања. Сви изрази важе у општем случају када је вредност фактора попуњености трапова $f_0 = 1$.

Концепт реда кинетике користи се за прорачуне параметара, а параметар ред кинетике дефинише се помоћу реалних физичких параметара. Резултати добијени анализом синтетичких и експерименталних кривих исијавања за фосфорне материјале омогућавају дубље разумевање процеса термолуминисцентне кинетике.

Кључне речи: термолуминисценција, ред кинетике, крива исијавања, модел кинетике, активациона енергија