

GLOW CURVE ANALYSIS BY GAUSS-LORENTZ FUNCTION

by

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A new method for fitting glow curves, described in a mixed order kinetics model, with Gauss-Lorentz function is shown. Theoretical expressions of the mixed order kinetics model are shown in a new way, so that the values of kinetic parameters can be obtained through the geometric parameters. When the model is described in this way it is possible to calculate precisely the kinetic parameters such as activation energy, pre-exponential factor and the factor $\alpha = n_0/(h + n_0)$. At the same time, obtained values of geometric parameters of the experimental curve, which is described with the Gauss-Lorentz function, can be used to estimate the kinetic model, in which thermoluminescence relaxation occurs. This gives a possibility of a new application of Gauss-Lorentz function to be used as a criterion for assessing model of relaxation, when it is not known in advance. The accuracy of fitting is studied, for the specific cases of computer simulated thermoluminescent curves with one peak.

Key words: thermoluminescence, mixed order kinetics, general order kinetics, Gauss-Lorentz function

INTRODUCTION

The general order and mixed order kinetic models [1, 2] have a great theoretical importance, because those can be accurately described by mathematical expressions, which help a better understanding of the physical phenomena and the influence of various parameters on the thermoluminescent (TL) process. While the general order model is the mathematical model which approximately describes any real TL process, the mixed order kinetics model describes the real physical process which is less possible. However, they are important as tools which enable us to find the ways in which TL processes can be analyzed. Their advantage is that the results can be accurately compared due to the exact mathematical expressions that describe them. This paper refers to the procedure for obtaining the analytical parameters for calculating the TL model of the mixed order kinetics and examines the possibility of evaluation of the kinetic model for the TL relaxation, from the shape of experimental curves.

Practice has shown that the TL curves with a maximum, which can be described with the different kinetics models, are very similar and it is difficult to determine the correct TL kinetics model from the shape of the obtained experimental curves. Also, the simultaneous usage of several different fitting func-

tions and choosing the closest one can provide only limited results, because it often happens that experimental curve can be successfully adjusted with different functions. The results of these adjustments are TL parameters and values that significantly deviate from the correct values, because the results strongly depend on the applied model. On the other hand, TL relaxation curves obtained from different models certainly differ. The idea of this paper is to find such a fitting function, where TL values of parameters will not be acquired immediately, but the parameter values that will give an estimate of the kinetic model and the TL values of the parameters will then be calculated on the basis of the calculation method, applicable to this model.

This fitting function is described with geometrical parameters and contains all the parameters required to describe its shape. The obtained values of geometric parameters have to enable accurate calculation of the TL model parameters, so that this adjustment process should be verified.

MIXED ORDER KINETICS MODEL

Mixed order kinetics model is shown with the following equation [2]

$$I \frac{dn}{dT} = \frac{s}{R} n(n+h) \exp \left(-\frac{E}{kT} \right) \quad (1)$$

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where at a time t [s], I [$\text{cm}^{-3}\text{s}^{-1}$] is the TL intensity, n [cm^{-3}] – the electron concentration in traps, h [cm^{-3}] – the initial electron concentration in inactive traps, s' [cm^3s^{-1}] – the pre-exponential factor, T (K) – the material temperature, R [Ks^{-1}] – the heating rate, E [eV] – the energy position of a single active trap, and k [eVK^{-1}] – the Boltzmann constant. As shown, for the second order kinetics [3], the calculation of the term h that is multiplied by s' is impossible. Therefore, it is more convenient to introduce, instead of pre-exponential factor s' , a new pre-exponential factor $s^{(h)} = s'h$ [3]. TL intensity could be shown by the equation [4]

$$I = \frac{s^{(h)} h \alpha \exp \frac{s^{(h)} T}{R} \exp \frac{E}{kT} dT}{\exp \frac{s^{(h)} T}{R} \exp \frac{E}{kT} dT} \alpha \quad (2)$$

When calculating the parameters by using a normalized form of glow curves i , where I_m is glow curve maximum, and n_m is the concentration of free carriers at the maximum, then the following relation is valid [5]

$$\delta_{\text{eff}} = \frac{\int_{T_m}^{\infty} i dt}{I_m} \frac{R n_m}{I_m} \quad (3)$$

One of the important parameters of TL curves, which can be obtained directly from the experimental TL curve, is the symmetry factor μ_s , which is defined by the relationship [6]

$$\mu_s = \frac{n_m}{n_0} \quad (4)$$

From the above relations, the expressions for calculating the activation energy E of the active traps can be obtained

$$E = \frac{1}{\alpha} \frac{\alpha(1 - 2\mu_s) k T_m^2}{\alpha(1 - \mu_s) \delta_{\text{eff}}} \quad (5)$$

and the symmetry factor

$$\mu_s = \frac{1}{\alpha} \frac{\alpha}{\exp \frac{1}{\alpha(1 - 2\mu_s)} (1 - \Delta_m) \alpha} \quad (6)$$

where $\Delta_m = 2 k T_m / E$ and pre-exponential factor $s^{(h)}$

$$s^{(h)} = R \frac{2}{\delta_{\text{eff}}} \frac{E}{k T_m^2} \exp \frac{E}{k T_m} \quad (7)$$

Using the equations listed above, for each specific experimental TL curve, which is described with the mixed order kinetics model, the parameters E , $s^{(h)}$, and α can be calculated. Calculation method, based on the iterative procedure is shown in detail in ref. [4].

If the eq. (5) for calculating the activation energy is written as

$$E = \frac{l k T_m^2}{\delta_{\text{eff}}} \quad (8)$$

then, one can define a parameter that describes the order of kinetics l in the mixed order kinetics model [4, 7]

$$l = \frac{1}{\alpha} \frac{\alpha(1 - 2\mu_s)}{\alpha(1 - \mu_s)} \quad (9)$$

When the parameter α obtained from this equation, is replaced in the eq. (6) it can be obtained

$$\alpha = \frac{1}{1 - \mu_s} \frac{2}{l} \frac{l}{1} \quad (10)$$

and the equation for calculating the symmetry factor is

$$\mu_s = \frac{1}{2} \frac{1}{l} \exp \frac{2}{l} \frac{l}{1} (1 - \Delta_m) \frac{l}{2} \frac{1}{l} \quad (11)$$

Derived equation is much more suitable for analytical calculations because the equation takes the form in which the value of the symmetry factor is explicitly shown. To accurately calculate the symmetry factor more members of the development can be used

$$1 - \Delta_m \left[\frac{3}{2} \Delta_m^2 - 3 \Delta_m^3 + \frac{15}{2} \Delta_m^4 - \frac{45}{2} \Delta_m^5 + \dots \right] \quad (12)$$

and the following equation is obtained

$$\mu_s = \frac{1}{2} \frac{1}{l} \exp \frac{2}{l} \frac{l}{1} \left[1 - \Delta_m \left(\frac{3}{2} \Delta_m^2 - 3 \Delta_m^3 + \frac{15}{2} \Delta_m^4 - \frac{45}{2} \Delta_m^5 + \dots \right) \right] \frac{l}{2} \frac{1}{l} \quad (13)$$

Based on this equation, it is possible to draw a curve symmetry factor as a function of kinetics order, for each specific parameter value Δ_m (fig. 1).

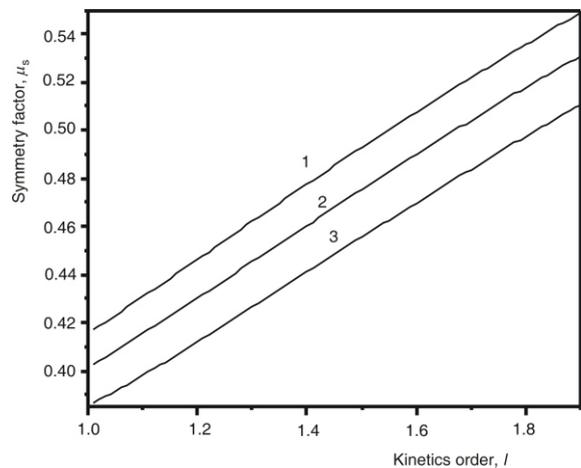


Figure 1. Dependence of the symmetry factor on the order of kinetics for different values of Δ_m : (1) 0.15, (2) 0.1, and (3) 0.05

The important range of values, for practical cases is $0.15 < \Delta_m < 0.05$ [5]. Analyzing the curves for different values of the μ_s it can be concluded that in the range of l values, from 1 to 2, the function can be replaced by a linear function with a high precision

$$\mu_s = A l + B \quad (14)$$

where A and B are the factors obtained by adjusting the linear function (14) with the exact values obtained using eq. (13). To achieve higher accuracy of calculations when calculating the symmetry factor more members of the order can be taken, but it is enough to take the first three members of the development.

If the distances between individual curves are linearized, the equation describes the μ_s dependence on l and Δ_m is obtained. For an approximation factor, it is more convenient to use a first-order polynomial, substituting

$$\mu_s = (0.18\Delta_m^2 + 0.105\Delta_m + 0.136) l - 0.277\Delta_m + 0.23 \quad (15)$$

Solving the eqs. (14) and (8) to calculate the order of kinetics l , a quadratic equation is obtained

$$0.136l^2 - 0.105\frac{2\delta_{\text{eff}}}{T_m} - 0.23 - \mu_s = 0 \quad (16)$$

$$0.276\frac{2\delta_{\text{eff}}}{T_m} - 0.18 - \frac{2\delta_{\text{eff}}}{T_m} = 0$$

Parameter values δ_{eff} , T_m , and $\mu_s = \delta_{\text{eff}}/\omega_{\text{eff}}$ are directly obtained from the experimental curve. Parameters E , $s^{(h)}$, and α are calculated analytically from eqs. (7), (8), and (10). The advantage of this equation is that the value of kinetics order calculated immediately and does not require an iterative process.

ASYMMETRIC GAUSS-LORENTZ FUNCTION

It was shown earlier that the relaxation rate or phosphorescence in TL process flows by the exponential law, in the case of first-order kinetics model and by the hyperbolic law, in the case of second order kinetics model [8]. Therefore, at the linear change in temperature of the sample, it can be assumed that the TL glow curves can be described by some forms of Gauss-Lorentz function [9] in the range between the first and the second order kinetics. Since TL curves generally have an asymmetric shape, it is necessary to use asymmetric forms of Gauss-Lorentz functions. Based on current forms Gaussian, Lorentz, Gauss-Lorentz, and asymmetric function it is proposed to use the following form of Gauss-Lorentz asymmetric function [10]

$$I = I_m \left(2^z - 1 \right) \frac{\ln^2 \left(1 + \frac{2(T - T_m) \text{sh } b}{\omega} \right)^{\frac{1}{z}}}{b^2} \quad (17)$$

where ω is the curve width, at half maximum intensity, z – the parameter which determines shape of the curve, b – the parameter which characterizes the degree of asymmetry of the curve, and I_m – the value of maximum intensity of TL curve. When $z = 1$ eq. (17) describes an asymmetric Lorentz curve, for $z = 0$ asymmetric Gaussian curve. In practice, the curves obtained by using eq. (17) were quite a bit different from the Gaussian curve, for the range of values $z < 0.08$ [9, 10]. At the same time, it was shown that this function, describes quite well the glow curves that were obtained by general order kinetics model [10]. It can, also, be assumed that the curves obtained by the mixed order kinetics model could be described with the same function as they are very similar to the curves of the general order kinetics model.

For the calculation parameters of the model of the mixed order kinetics it is necessary to determine the parameters δ_{eff} , T_m , and $\mu_s = \delta_{\text{eff}}/\omega_{\text{eff}}$. When by definition is [5]

$$\omega_{\text{eff}} = \int_{-\infty}^{\infty} I dt = \frac{Rn_m}{I_m} \quad (18)$$

It is necessary to calculate the integrals of functions (18) and (3) in the ranges from $-\infty$ to $+\infty$ and from T_m to $+\infty$. It was shown, that the values of these integrals could be calculated with sufficient precision. If $Z = 1/z$ is substituted in eq. (17) before calculating the integral, the three groups of solutions were obtained.

For the positive value of the parameter Z , function integrals (3) and (18) are calculated using the equations

$$\delta_{\text{eff}} = \frac{\omega}{4 \text{sh } b \Gamma(Z)} \int_0^{\infty} \frac{b^j}{\sqrt{2^{1/Z} - 1}} \frac{(j-1)!!}{2^{j/2}} \sqrt{\pi} \Gamma(Z) \frac{j-1}{2} \quad (19)$$

$$\omega_{\text{eff}} = \frac{\omega}{2 \text{sh } b \Gamma(Z)} \int_0^{\infty} \frac{b^{2j-1}}{\sqrt{2^{1/Z} - 1}} \frac{1}{2j!} \frac{(2j-1)!!}{2^j} \sqrt{\pi} \Gamma(Z) \frac{j-1}{2} \quad (20)$$

for the negative value

$$\delta_{\text{eff}} = \frac{\omega}{4 \text{sh } b} \Gamma(|Z|) \int_0^{\infty} \frac{b^j}{\sqrt{1 - 2^{1/Z}}} \frac{(j-1)!!}{2^{j/2}} \sqrt{\pi} \frac{1}{\Gamma(|Z|) \frac{j-3}{2}} \quad (21)$$

$$\omega_{\text{eff}} = \frac{\omega}{2shb} \Gamma(|Z| - 1) \frac{j}{j-1} \frac{b}{\sqrt{1 - 2^{1/Z}}} \frac{1}{2j!} \frac{(2j-1)!!}{2^j} \sqrt{\pi} \frac{1}{\Gamma(|Z| - j + \frac{3}{2})} \quad (22)$$

and for the value $z = 0$, or $Z = \infty$

$$\delta_{\text{eff}} = \frac{\omega}{4shb} \frac{j}{j-1} \frac{1}{j!} \sqrt{\frac{\ln 2}{b^2}} \frac{j}{\Gamma(j + \frac{1}{2})} \quad (23)$$

$$\omega_{\text{eff}} = \frac{\omega}{2shb} \frac{j}{j-1} \frac{1}{j!} \sqrt{\frac{\ln 2}{b^2}} \frac{2j-1}{\Gamma(j - \frac{1}{2})} \quad (24)$$

where $\Gamma(Z)$ is the value of the gamma function of the parameter Z .

In tab. 1 the values $T_m(K)$, $\omega(K)$, b , and z of fitting the synthetic curves to Gauss-Lorentz function and the calculated values of the parameters E_c , $s_c^{(h)}$, and α_c , are shown. The synthetic curves were established for the following constant parameters: $E = 1 \text{ keV}$, $s = 10^{10}/\text{s}$, $R = 1 \text{ K/s}$, $N = 10^{10}/\text{cm}^3$, and for changeable parameters: trap fillings f and factor α .

DISCUSSION

According to the developed and accepted theory of phosphorescent and TL relaxation, the glow curves are the result of competition between the capture centers in the phosphor material [11]. The complexity of the problem, depends on the number of different types of the capture centers and their physical characteristics. When solving the simpler problems, one usually assumes the presence of one type of recombination centers and the one or two types of traps or capture centers. Based on these characteristics of phosphor material, the glow curve having one maximum is ob-

tained and it matches only a specific kinetics model. Visually, it is impossible to estimate what type of kinetics model should be applied to the analysis of some glow curves. The general feature of these kinetics models is that the luminescent process can dominate and then the TL process can be described by the first order kinetics model, or if re-trapping processes are dominant, then the TL process can be described by the model of second-order kinetics. When the value of kinetics order is between one and two, then none of the above processes are dominant and on the basis of obtaining glow curve, it is not possible to estimate what type of kinetics model is applicable and what is the value of its order of kinetics.

In the case of kinetic models, defined for phosphor materials, with one type of luminescent centers and one type of traps, and if there is a re-trapping process in the material, then the probability of re-trapping depends on re-trapping coefficient and the concentration of empty traps. When the concentration of empty traps is small at initial time, then TL process takes place approximately as the first-order kinetics model. However, as the progress of the process leads to increase the concentration of empty traps and re-trapping process increases, which means that the process of TL relaxation is closer to the second-order kinetics model. This process goes relatively quickly, because, as the probability of re-trapping increases, so the probability of luminescence decreases due to decrease of free carriers concentration in the traps. Therefore, this shape of glow curve is approaching fast the shape of the curve described by the Lorentz function. When a sufficient number of traps are depleted, so that re-trapping becomes dominant, than the TL relaxation will be carried out by the model of second-order kinetics, continuously. This type of kinetics is described by a differential equation which cannot be solved analytically, but it is often approximated, by the general order kinetics model, which gives equation that is solvable analytically.

Table 1. The calculated values of the parameters of synthetic curves of mixed order kinetics model

α	f	$T_m[\text{K}]$	$\omega[\text{K}]$	b	z	CHI^2	$E_c[\text{keV}]$	$s_c^{(h)}[\text{s}^{-1}]$	α_c
0.20	1.00	424.444	38.650	-0.280	0.0072	$1.18 \cdot 10^{-7}$	1.00	$3.52 \cdot 10^{10}$	0.19
0.60	1.00	441.211	48.811	-0.137	-0.0020	$1.62 \cdot 10^{-5}$	1.01	$7.73 \cdot 10^9$	0.61
0.99	1.00	447.261	56.881	-0.086	0.2665	$4.63 \cdot 10^{-6}$	0.99	$3.80 \cdot 10^7$	0.99
0.20	0.75	428.600	39.401	-0.277	0.0013	$9.40 \cdot 10^{-8}$	1.00	$2.94 \cdot 10^{10}$	0.21
0.60	0.75	445.747	49.794	-0.137	-0.0025	$1.35 \cdot 10^{-5}$	1.01	$5.76 \cdot 10^9$	0.61
0.99	0.75	451.902	58.037	-0.087	0.2658	$7.33 \cdot 10^{-7}$	0.99	$7.33 \cdot 10^7$	0.99
0.20	0.50	434.666	40.495	-0.276	0.0012	$1.01 \cdot 10^{-7}$	1.00	$1.94 \cdot 10^{10}$	0.20
0.60	0.50	452.280	51.225	-0.135	-0.0026	$1.33 \cdot 10^{-5}$	1.01	$3.86 \cdot 10^9$	0.61
0.99	0.50	458.600	59.587	0.089	0.2772	$4.04 \cdot 10^{-6}$	0.99	$3.97 \cdot 10^7$	0.99
0.20	0.25	445.400	42.478	-0.274	0.0071	$9.04 \cdot 10^{-8}$	1.00	$9.67 \cdot 10^9$	0.20
0.60	0.25	463.918	53.792	-0.134	-0.0079	$131 \cdot 10^{-5}$	1.01	$1.97 \cdot 10^9$	0.61
0.99	0.25	470.500	62.577	-0.091	0.2655	$3.99 \cdot 10^{-6}$	0.99	$2.40 \cdot 10^7$	0.99

The TL relaxation for general case of phosphor materials, with one type of recombination centers and two types of traps, of which the one type of centers active, and the other inactive, it is known as a non-interactive kinetics.

Kinetics order for a simple phosphor material that contains only one type of traps and one type of luminescent centers depends on the relative ratio between the re-trapping factor and filling factor. The second-order kinetics is achieved when traps are filled slightly, or the re-trapping factor is high. It could be assumed, that the value of re-trapping factor is small, for some phosphors. Because at some moment, at the end of the process of relaxation, traps filling are much smaller than the free carriers re-trapping, which have a constant value for a given material, the relaxation process becomes the second-order kinetics and ends with it. In theory, the kinetics of a process can begin and end with the second-order kinetics, even in the case of high traps filling, when the value of re-trapping factor is equal to one. The reason for this is, that recombination is always identical to decrease of re-trapping events and increase of recombination is always equal to the reduction of the re-trapping events. The result of this is, that the sum of recombination and re-trapping events is always constant and the whole relaxation process flows in the same way from the start to the end. It can be concluded, that the second-order kinetics process is stable and the relaxation process is approaching slowly to the above kinetics. The first-order kinetics process takes place in a completely different way. Relatively small changes in the concentration of the active traps, will cause relatively large changes in the curve shape, in the reduction recombination and increase in re-trapping.

When phosphor contains inactive traps, it significantly affects the relaxation process itself. Because of electrical neutrality of materials, the inactive traps increase the concentration of recombination centers. If the process starts from the first-order kinetics, then emptying the traps will not significantly impact the process performance, because the probability of recombination will be always much higher than the probability of re-trapping, due to the much higher concentration of recombination centers. The situation is quite different when the process begins to perform with the second-order kinetics. In this case, the re-trapping processes are dominant till the moment when the relative ratio of re-trapping coefficients and coefficients of the luminescent recombination prevail over the ratio of concentrations of recombination centers and active traps. Due to the presence of a constant concentration of recombination centers in the material, at one point, the kinetics of the process will change relatively quickly into the first order kinetics. In the materials, near to the first-order kinetics, there are no significant changes in the process and it is stable. Based on the given analysis, it can be concluded that the processes, used to describe changes of the kinetics in the general order and

mixed order kinetics model are opposite. This can be seen from the analysis of parameter z values, which describes the shape of the Gauss-Lorentz curve (fig. 2). Therefore, it can be concluded that the shape factor z , can be used to estimate which of the two kinetics models (mixed or general order kinetics) should be applied in the analysis some of unknown glow curves. In this way, the Gauss-Lorentz function can also be used as a criterion to evaluate the kinetics model that can be applied in the analysis of a glow curve.

CONCLUSIONS

In this paper it is shown that the glow curve, which is described by the mixed order kinetics model, can be successfully adjusted with the Gauss-Lorentz function. At the same time, it is possible to derive a calculation method, based on the mixed order kinetics model, using Gauss-Lorentz function. This method enables the analytical calculation of the TL process parameters. The geometric parameters, obtained by fitting the glow curve with Gauss Lorentz function, are used for the calculation of TL parameters in this method. Calculation method applies the concept of using parameter of kinetics order, which depends on the symmetry factor. In previous studies, the parameter of kinetics order for mixed-order kinetics was defined and, in this paper, it is shown that the correlation between the kinetic parameters of order and symmetry factor is linear. It allows faster and more accurate calculation of the value of kinetics order for some TL glow curve. Then, the TL process parameters are calculated from the equations, previously derived using the calculated value of kinetics order. This calculation method uses the integral values δ_{eff} and ω_{eff} , which al-

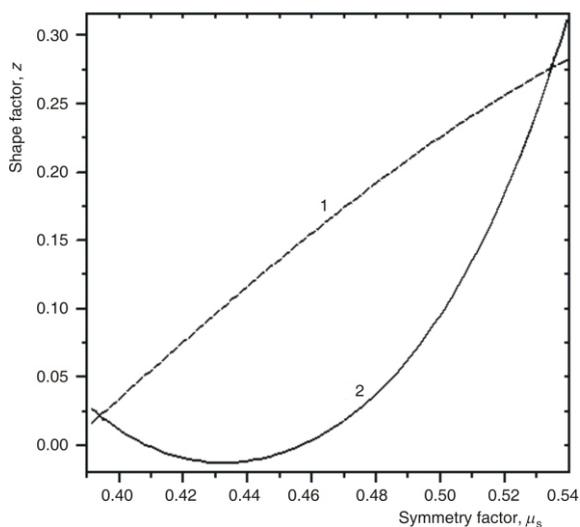


Figure 2. Dependence shape factor z in terms of symmetry factor μ_s : (1) for general kinetics model and (2) mixed kinetics model

low more precise calculation of parameter values, because the integral values are less sensitive to the random fluctuations of the intensity of the glow curve in some points. Also, fitting the TL curve with the Gauss Lorentz function enables accurate calculation of the integral values of the parameters. This is important for measuring radiation dose and also for the computation of the TL process parameters [12].

Analysis of glow curves, obtained from the equations as described by the general and the mixed kinetics order model, show that the shape of the curve function is not described by symmetry factor only, but by the factor that indicates, whether the shape of the curve is closer to the form given by the Gaussian function or to the form that is closer to the Lorentz function. There was a significant difference of the obtained shape factor values in the symmetry factor function, for general and mixed order kinetics model. This fact makes it possible to use the obtained values of shape factor to assess by which kinetics model the relaxation is performing.

Therefore it can be concluded, that the Gauss-Lorentz function can be used to analyze the glow curves by fitting the experimental glow curve with it, to obtain an accurate curve and to determine its geometric parameters and then, for accurate determination of the values of kinetics order and integral parameters, it is necessary to calculate the kinetic parameters of TL and as the criterion for assessing model by which the TL relaxation occurs [13].

AUTHORS CONTRIBUTIONS

All authors were equally involved in models analysis and then in the analysis and discussion of results. The manuscript was written and the figures were prepared by Z. M. Vejnović.

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АНАЛИЗА КРИВЕ ИСИЈАВАЊА ПОМОЋУ ГАУС-ЛОРЕНЦОВЕ ФУНКЦИЈЕ

Приказан је нов метод помоћу кога се криве исијавања, које се описују моделом кинетике мешовитог реда, подешавају са Гаус-Лоренцовом функцијом. Теоријски изрази модела кинетике мешовитог реда су приказани на потпуно нов начин тако да се вредности параметара кинетике могу добити преко геометријских параметара. Овакав начин приказа омогућава да се прецизно израчунају кинетички параметри модела кинетике као што су активациона енергија, преекспоненцијални фактор и фактор $\alpha = n_0/(h + n_0)$. Истовремено се може, на основу добијених вредности геометријских параметара експерименталне криве, која је описана Гаус-Лоренцовом функцијом, проценити модел кинетике по коме се обавља термолуминесцентна релаксација. То омогућава и нову примену Гаус-Лоренцове функције која се може користити и као критеријум за процену типа кинетике по којој се одвија релаксација, када то није унапред познато. Испитивана је тачност подешавања за карактеристичне случајеве рачунарски симулираних ТЛ кривих са једним максимумом.

Кључне речи: термолуминесценција, кинетика мешовитог реда, кинетика отишлог реда,
Гаус-Лоренцова функција
