

Glow Curve Analysis and Calculation of Thermoluminescence Parameters

Z.Vejnović, M. Pavlović, P. Hadžić, M. Davidović

1 **Abstract:** A new method for analysis of the glow curve with one maximum is described. This
2 method is based on fitting the experimental glow curve with a new type of asymmetric Gauss-
3 Lorentz (GL) function. This function was adapted to be very precise in describing glow curves.
4 A completely new algorithm for calculating the kinetic parameters of the process for the OTOR
5 (One Trap-One Recombination center) model or the model of ideal phosphor was developed.
6 The kinetics order concept was used for calculation of the kinetic parameters of the process.
7 The kinetics order parameter was defined by means of real physical parameters. A new function
8 was proposed to describe the dependence of the factor symmetry of order kinetics. It has
9 enabled a very accurate calculation of the activation energy parameter. The resulting relative
10 error was less than 0.5%.

11 **Keywords:** Thermoluminescence, Kinetics order, Glow curve, Kinetics model, Activation
12 energy

13 1 Introduction

14 For an accurate and reliable thermoluminescent analysis, it is important to know the phys-
15 ical models describing TL relaxation [1,2]. In this case, it is possible to connect them
16 with practical experiments and carry out the procedure for their qualitative and quantitative
17 evaluation. The experimental glow curve, with the use of certified and valuable physi-
18 cal models can provide accurate calculation of the physical parameters of the TL process
19 [3,4,5,6,7,8,9,10]. If the accuracy of the obtained parameter values is confirmed by other
20 methods of analysis and measurement, this is a reliable indicator that a given model can
21 be used. The calculated values of the kinetic parameters of the real model can be used for
22 reconstruction of the TL process, its analysis, and for its better understanding.

23 The general order kinetics model is commonly used for TL analysis [5, 11–14]. It is a
24 mathematical model, which is designed to describe the shapes of glow curves. Therefore,

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Z.Vejnović, M.Pavlović, P.Hadžić, M.Davidović are with the Institut Gosha, Belgrade, Serbia.

25 the use of this model for calculating the real physical parameters is limited or excluded. It
26 can be shown in many cases, in practice, that some physical parameters can not be calcu-
27 lated, and the calculated values of other parameters are not accurate enough. The use of
28 realistic physical models, due to difficulties with the calculation of the process parameters,
29 has almost completely been eliminated. This significantly limits the understanding of the
30 process and slows down progress in understanding complex phenomena. An important step
31 in further progress in the research of the existing model is to develop an algorithm for the
32 calculation of real physical parameters.

33 A number of different models exist in the theory of TL processes [1–3, 6, 7, 14, 16–19].
34 One of the important tasks is a good estimate which of the existing models is applicable.
35 This ensures that the values of the kinetic parameters of the TL process are accurately cal-
36 culated. Another important task is to develop an algorithm for accurately calculating the
37 parameters. This means that for each model there is a corresponding calculation algorithm.
38 Specific tasks, dealing with the development of the calculation algorithms are: determi-
39 nation of model restrictions, which parameters have the greatest influence on the process,
40 which parameters can be independently calculated, and so on. Answers to these questions
41 enables improvement of existing models and development of new advanced models.

42 The main objective of this paper is calculation of the real physical parameters of the
43 glow curve based on an experimentally obtained TL curve followed by reconstruction of
44 the physical process of TL relaxation. Therefore, criteria enabling estimation of the model
45 describing the kinetics of the process must be developed first. Practice has shown that visual
46 evaluation of the kinetics model is not possible as it requires more accurate and reliable
47 criterion. It is very important to accurately calculate the parameters of the process [20].

48 In this paper, calculation of the physical parameters of the process was applied only to
49 the model of ideal phosphor. This model is based on theoretical differential equations that
50 describe the kinetics of the process in ideal phosphors and are given in the following books
51 and papers [3, 17, 21, 22]. The term ideal phosphor involves phosphors that can be modeled
52 with one type of traps and one type of light emission center.

53 The algorithm for calculation of the parameters of TL kinetics, for the model of ideal
54 phosphors, was based on the concept of kinetics order [1, 2, 12–14]. In order to apply this
55 concept it is necessary to define kinetics order in a new way. Definition of kinetics order
56 applied in the general order kinetics is completely inappropriate for the model of ideal
57 phosphors. The equation for calculating the activation energy is used to define a kinetics
58 order parameter.

59 Starting from this equation glow curves were analyzed for typical values of kinetic
60 parameters. Based on these results, the relationship between the order of kinetics l and
61 symmetry factor μ_s was determined. This relationship enabled the use of the main principles
62 of an existing algorithm, from the general order kinetics model, to calculate the activation
63 energy E [5, 7, 13, 20].

64 2 Asymmetrical Gauss Lorentz function

65 It has previously been shown that the TL relaxation rate or phosphorescent decay follows
66 an exponential law in the case of first-order kinetics and an approximately hyperbolic law
67 in the case of second-order kinetics.

68 From there, with respect to the linear heating rate, one can assume that the first order
69 kinetics curve can be approximated with the Gauss function and the second order kinetics
70 can be approximated with the modified Gaussian or GL function [20, 24].

71 In general, it can be assumed that the glow curve, whose kinetic order value is between
72 one and two, can be described with a curve that connects the Gauss and Lorentz functions.

73 One form of the function, connecting the Gaussian and Lorentz curves, is given as:

$$74 \quad I = I_m \left\{ 1 + (2^Z - 1) \frac{\left\{ \exp \left(\frac{[\ln(1+b) - \ln(1-b)](T - T_m)}{\omega [1 - \ln(1-b)] [1 - \ln(1+b)] + [\ln(1+b) - \ln(1-b)](T - T_m)} \right) - 1 \right\}^2}{b^2} \right\}^{-\frac{1}{Z}} \quad (1)$$

75 where I_m is the maximum value of the function, T_m is the position of the maximum, ω is
76 the half-width, b characterizes the degree of asymmetry, and Z is a parameter determining
77 the shape. For $Z=1$ eq.(1) describes the Lorentz function and for $Z \rightarrow 0$ the Gauss function.
78 For $Z < 1/12$ eq.(1) differs negligibly from the Gauss function [20].

79 It can be shown that equation (1) can be successfully used for fitting synthetic glow
80 curves and real glow curves with one maximum. Complex analysis of a real glow curve
81 assumes its comparison with theoretical curves.

82 3 Evaluation the applicability of models

83 Experimental glow curves with one maximum are the consequence of the fact that phosphor
84 contains only one type of active traps. The simplest physically realistic process, which is
85 described by the glow curve with one maximum, occurs in the case of phosphor consisting
86 of one type of traps and one type of luminescent centers. This model is known as the OTOR
87 model (One Trap-One Recombination center) or the model of ideal phosphor [2, 17]. A
88 more complex model, consisting of one type of active traps, one type of isolated or inactive
89 traps and one type of recombination centers, is called the non-interactive kinetics model or
90 the NMTS model (Non-interactive MultiTrap System) [1, 2]. The model of ideal phosphor
91 determines the limit of applicability of the NMTS model, as it is applied in the case of a
92 negligible concentration of inactive traps. The kinetics model, which is known as the model
93 of mixed order or the MO model (Mixed Order) [2, 15, 17, 23], is only one special form of
94 non-interactive kinetics.

95 A general differential equation that describes the kinetics of these models is given as
96 [1,2]:

$$97 \quad I = -\frac{dp_l}{dt} = \frac{s \exp\left(-\frac{E}{kT}\right) n(n+m)}{(n+m) + r(N-n)} \quad (2)$$

98 where at a time t (s), I ($\text{cm}^{-3} \text{s}^{-1}$) is the TL intensity, p_l (cm^{-3}) is the concentration of
99 luminescent centers, n (cm^{-3}) is the electron trap concentration, N (cm^{-3}) is the concen-
100 tration of active traps, m (cm^{-3}) is the initial electron inactive trap concentration, s (s^{-1}) is
101 the frequency factor, E (eV) is the energy depth of a single active trap, T (K) is the mate-
102 rial temperature, and k (eVK^{-1}) is the Boltzmann constant, r is γ_t/γ_l , where γ_t (cm^3s^{-1})
103 and γ_l (cm^3s^{-1}) are the recombination probabilities between the conduction band electrons
104 and the traps or the luminescent centers, respectively. To obtain this equation, an approx-
105 imation describing the quasi-stationary equilibrium of electrons in the conduction band is
106 used. This approximation assumes that the rate of change of the concentration of electrons
107 in the conduction zone is much smaller than the speed of change of the electron in the traps.
108 Inactive traps are, in fact, deep traps, which cannot be filled and emptied during thermal
109 relaxation. Equation (2) describes the NMTS model, for $r=1$ it describes the MO model,
110 and for $m=0$ it describes the OTOR model or the model of ideal phosphor. Equation (2) can
111 be written as follows:

$$112 \quad I = -\frac{dn}{dt} = \frac{s \exp\left(-\frac{E}{kT}\right) n}{1 + r\frac{N}{n}\frac{n}{n+m} - r\frac{n}{n+m}} = \frac{Sn}{1 + r\frac{\alpha}{f} - r\alpha} = \frac{Sn}{1 + r\alpha\left(\frac{1}{f} - 1\right)} \quad (3)$$

113 where $S=s\exp(-E/kT)$, $f=n/N$ is the filling factor, and $\alpha = n/(n+m)$ is the factor that di-
114 rectly determines the value of kinetic order in the model of mixed-order kinetics. It can be
115 concluded that factor α determines the shape of the glow curve, in the same manner as the
116 retrapping factor r . The $r\alpha/f_0$ ratio determines whether the parameter kinetics order will
117 have a value between one and two. The values of this ratio may be the same for a large
118 number of different values of parameters r , α_0 and f_0 , where $f_0 = n_0/N$, $\alpha_0 = n_0/(n_0+m)$
119 is the initial electron concentration in the active traps. Therefore, it is necessary to choose
120 the value of the f_0 factor. This factor can have any value in the range from 0 to 1. For
121 simplicity it can be assumed that $f_0=1$. Based on this it was possible, to make the diagram
122 describing how the value of the shape factor z depends on the symmetry factor μ_s , for glow
123 curves obtained by numerical solution of differential equations (3), for defined parameter
124 values s , E , r , f_0 , α_0 (Fig.1.) [25]. Parameters s and E determine the position of the diagram
125 in Fig.1, using the values of parameter $\Delta = 2kT_m/E$, where T_m is the temperature of the
126 glow curve maximum. When the value of parameter Δ decreases, the position of the curves
127 in the diagram are shifted to the left (curve 3 in the diagram), and in the opposite case to
128 the right.

129 Good evaluation of the model, where TL relaxation takes place, requires calculation of
130 the value of Δ , for a glow curve that is experimentally obtained. This value, with sufficient

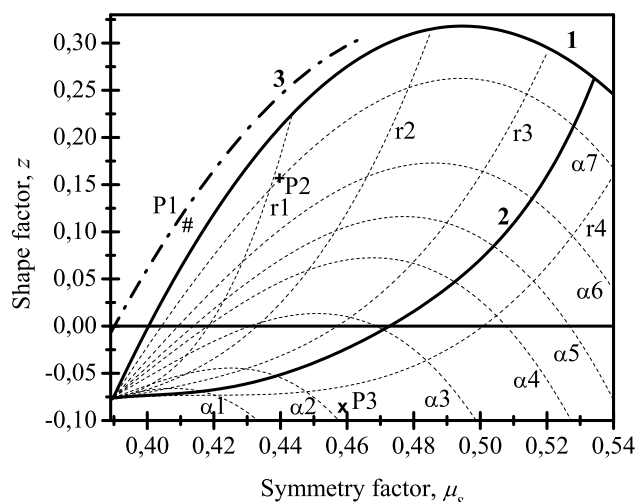


Fig. 1. Criteria for determining which kinetic model can describe a certain glow curve, for the value of $\Delta = 0.07957$, for different values of parameters r and α : 1-model of ideal phosphor $\alpha = 1$, $\alpha_1 = 0.3$, $\alpha_2 = 0.5$, $\alpha_3 = 0.7$, $\alpha_4 = 0.8$, $\alpha_5 = 0.85$, $\alpha_6 = 0.9$, $\alpha_7 = 0.96$, 2-mixed order kinetics model $r=1$, $r_1=0.1$, $r_2=0.3$, $r_3=0.7$, $r_4=1.5$. The criteria, for the model of ideal phosphor, for the value of $\Delta = 0.037545$ is shown with a dot-dash line (3). Values, which were determined for some dosimeters, are shown as follows: P1(#)-TLD700H(7 LiF:Mg,Cu,P), P2(+)-TLD400(CaF:Mn) i P3(x)-TLD500(Al_2O_3 :C)

131 accuracy, can be determined relatively easily using existing iterative methods. Such an
 132 iterative method is described in this paper. The procedure used in the calculation of the
 133 model parameters in general-order kinetics can also be used.

134 Analysis of a large number of theoretical curves for the NMTS model, enabled conclu-
 135 sion that the factors describing the shape (Z) and asymmetry (b) of the curves, change in
 136 different ways, depending on the values of the parameters that define the kinetics of the pro-
 137 cess. This allows determination whether the kinetic process can be described using a model
 138 of an ideal phosphor or mixed-order kinetics model or models of noninteractive kinetics
 139 using a simple determination of their values.

140 Values of the shape factor and symmetry factor were determined and are shown in Fig.1
 141 for some typical TL dosimeters [10, 16, 26–29].

142 4 Advanced asymmetric Gauss-Lorentz function

143 In the analysis of TL glow curves with GL functions, it has been noted that in some cases
 144 they cannot be described with sufficient precision. It is shown that values of the shape and
 145 the asymmetry factor, for any particular TL glow curve are not constant [2,14,22]. Thus,

146 at the beginning of the glow curve, TL relaxation may be followed by first order kinetics,
 147 while the second order kinetics can be applied at its end. The opposite may apply too. In
 148 both cases the factor asymmetry and the shape factor have different values at the start and
 149 at the end of the process. In areas where the change in the value of factors is significant or
 150 where the changes oscillate, imprecision in describing the curves is higher.

151 Function (1) has five parameters, and if the normalized curve is used, which is applica-
 152 ble to curves with only one peak then it has four parameters. The geometrical parameters
 153 ω and T_m can be taken from the glow curve directly and they are important characteris-
 154 tics. Precise calculation of these parameters is especially important for calculating values
 155 of physical parameters of the kinetic process. Therefore, it was assumed that their value
 156 depended on the time at which the process was observed. Then it can be assumed that the
 157 factors of asymmetry and shape factors have changeable values versus time t , or versus
 158 heating temperatures T . Analysis of synthetic TL glow curves showed less accurate fitting
 159 with the GL function, in areas where significant changes in the shape factor occurred. It
 160 can be concluded that the shape factor is a more complex function than the asymmetry fac-
 161 tor. Therefore, it is appropriate that the shape factor Z is approximated by the following
 162 function:

$$163 \quad Z = z_2 (T - T_m)^2 + z_1 (T - T_m) + z, \quad (4)$$

164 and the factor of asymmetry with a linear function:

$$165 \quad b = b_1 (T - T_m) + b_0 \quad (5)$$

166 After replacing the expressions for the shape factor (4) and the symmetry factor (5) in
 167 equation (1), the new, improved GL function is obtained. This function has three new
 168 parameters and computer simulated curves showed better fitting of the TL glow curves.

169 **5 Kinetics of the model of ideal phosphors**

170 After substitution of $\alpha = 1$ in (3), the equation of the model of ideal phosphors is obtained:

$$171 \quad I = -\frac{dn}{dt} = \frac{ns \exp\left(-\frac{E}{kT}\right)}{1 + r\left(\frac{n}{N} - 1\right)} = \frac{Sn}{1 + r\left(\frac{1}{f} - 1\right)} \quad (6)$$

172 When $r=0$ then equation (6) becomes the equation of first order kinetics models, and
 173 when $n \ll N$ or $r=1$ it becomes the equation of the second order kinetics model. At the
 174 beginning of the TL relaxation time $t=0$, the expression in the denominator of equation (6)
 175 will be:

$$176 \quad 1 - r + \frac{r}{f_0} \quad (7)$$

177 where $f_0 = n_0/N$ is the filling factor of traps at time $t=0$. The filling factor has a value from
 178 0 to 1. It may be noted, that when the value of r is greater than one then the value of the

179 kinetics order is greater than two, due to high electron retrapping. When $r < f_0$, there are two
 180 cases. For $r \ll f_0$ TL relaxation kinetics follows the first-order kinetics model, due to very
 181 low retrapping. The kinetics, which can be defined as kinetics of the l -th order, corresponds
 182 to the case when $r \approx f_0$, and when the value of r is less than f , in less than two orders of
 183 magnitude. It should be noted, that even with very low occupancy of traps kinetics of the
 184 l -th order may occur, if the value of the retrapping factor is also proportionately very small.

185 In general, equation (6) cannot be solved, making it difficult to analyze the characteris-
 186 tics of the model and analysis of experimental curves. Therefore, the exponential type GL
 187 function and concept of kinetics order have been used, for analysis of the characteristics
 188 of TL kinetics. To make the concept of kinetics order could be used in realistic physical
 189 models, it is necessary previously defined parameter of kinetics order using real physical
 190 parameters.

191 6 Kinetics order concept

192 The concept of kinetics order is based on a model of general-order kinetics, which approxi-
 193 mately describes the real experimental glow curves in the range between the first and second
 194 order kinetics [11–14]. Practice has shown that this is quite an inaccurate approximation,
 195 and does not enable calculation of all parameters. Values of the activation energy of traps
 196 can be calculated approximately, while other parameter values parameters cannot even be
 197 roughly estimated. However, the concept of order kinetics has demonstrated that a theo-
 198 retical curve can be obtained, which is nearly a perfect match with experimental curves,
 199 and parameters of the theoretical curve are not derived from the process of TL relaxation.
 200 These parameters are the result of mathematical assumptions. Therefore, reconstruction of
 201 the kinetics of the TL process is not possible based on these values. This prevents success-
 202 ful research and understanding of the kinetics of TL relaxation. It has been shown, that one
 203 of the results of applying the concept of order kinetics is that the values of activation energy
 204 of traps can be calculated using the relation [5, 13]:

$$205 \quad E = \frac{l k T_m^2}{\delta_{eff}} \quad (8)$$

206 where l is the kinetics order, δ_{eff} is the part of the integral of glow curve from the maxi-
 207 mum to its end. It should be noted that this solution is obtained by analytically solving a
 208 differential equation, which describes the general order kinetics model [5]. Since the glow
 209 curve of the general order model may perfectly fit any experimental TL curve, it can be
 210 concluded that equation (8) can also be applied to any experimental curve. This can be
 211 proved, because the same equation is used to calculate the activation energy for the cases of
 212 models of first order [3,18,30], second order [3,19,30] and mixed order [6,7,31].

213 When equation (8) is used, in order to calculate the values of experimental parameters

214 of the glow curve, the values which are obtained, sometimes, considerably deviate from
 215 accurate values. In equation (8) k is a constant, and parameters T_m and δ_{eff} are geometrical
 216 parameters, which can be very accurately obtained from experimental curves. Obviously,
 217 deviation from the correct value is due to the inability to determine the exact value of the
 218 kinetics order l . It was concluded, based on comparisons of differential equations of the
 219 model of ideal phosphor and general order kinetics model that this is due to changes in
 220 the value of parameter l , for the duration of the relaxation process. Thus, parameter l
 221 is related to the kinetics of the process and cannot be calculated. However, there are no
 222 processes of the l -th order, but processes of the first and second order kinetics. The equation
 223 for calculating the activation energy, for the mixed-order kinetic model [7], indicates that
 224 parameter l has a constant value. It refers to a previously determined glow curve, and
 225 depends on initial conditions. This means that for the same phosphors different values of
 226 order kinetics may be obtained if the initial conditions are different. Therefore, the value
 227 of kinetics order, which is constant, can be calculated for each experimental curve. On the
 228 basis of this conclusion, it is possible to introduce an expression that defines the kinetics
 229 order for the model of ideal phosphor:

$$230 \quad l = \frac{E\delta_{eff}}{kT_m^2} \quad (9)$$

231 The value of kinetics order, which is defined in such a way, should be introduced in the
 232 calculation algorithm, in order to obtain the correct values of physical parameters. In all
 233 the above models, the same principle is used to develop the calculation procedures. This
 234 ensures accurate calculation of the parameters of TL kinetics.

235 When the concept of kinetics order is applied to a model of ideal phosphor, it is neces-
 236 sary to examine the effect of the r/f_0 ratio on the shape of the glow curve, or on the value
 237 of parameter l . After testing a large number of glow curves, it was determined that the
 238 function r/f_0 versus l , depends only on the initial occupancy of traps f_0 . Figure 2 shows
 239 this dependence, in the range of parameter values of practical interest for the analysis of
 240 experimental glow curves with one maximum. The obtained diagram is generally valid, for
 241 the model of ideal phosphors. Therefore, this diagram can be used to calculate the kinetic
 242 parameters of TL.

243 **7 Calculating the parameters of the model of ideal phosphors**

244 When the theoretical equation (6) is used for reconstruction of the kinetics of the TL pro-
 245 cess, it is necessary to know the values of the parameters that define the process. For the
 246 normalized glow curve it is necessary to know the values of parameters s , r , f_0 and E .

247 The glow curve must be presented in an analytical form for quick and accurate analysis.
 248 Analysis of the glow curve, which uses the improved GL function of the exponential type,

249 allows accurate calculation of geometrical parameters T_m , δ_{eff} and μ_s [20,32]. Using equa-
 250 tion (8) and the diagram in Fig.2 it is possible to determine the values of activation energy
 251 E and the r/f_0 ratio. First, the value of kinetics order l has to be calculated.

252 Kinetics order is determined from the diagram that describes how the symmetry factor
 253 μ_s is dependent on the kinetics order, for different values of $\Delta = 2kT_m/E$. The method is
 254 based on an iterative procedure and on the proven assumption that the value of Δ is located
 255 within a limited range of values, from 0.03 to 0.15. For this entire range, a functional
 256 dependence of $\mu_s(l)$ exists allowing calculation of the value of l .

257 If it is assumed that the r/f_0 ratio is in the range of 0 to 3, then glow curves obtained by
 258 numerical solution of the differential equation (6) can be analyzed for four groups of initial
 259 values. Differential equations were solved numerically using the Runge Kutta IV order.
 260 Four groups of parameter values were chosen to cover all values of Δ , which are of interest
 261 (from 0.03 to 0.15): the first group $E_1=3\text{eV}$, $s_1=10^{22}\text{s}^{-1}$ ($\Delta_1=0.03755$), the second group
 262 $E_2=1\text{eV}$, $s_2=10^{12}\text{s}^{-1}$ ($\Delta_2 = 0.06605$), the third group $E_3=0.2\text{eV}$, $s_3=10^8\text{s}^{-1}$ ($\Delta_3 = 0.09865$)
 263 and the fourth group $E_4=0.1\text{eV}$, $s_4=10^5\text{s}^{-1}$ ($\Delta_4 = 0.14815$). The value of the filling factor
 264 of traps was taken to be $f_0=1$, to ensure that changes in Δ values are as little as possible,
 265 for glow curves of the same group. Obtained simulated curves were analyzed using the
 266 improved GL function of the exponential type. The value of T_m can be obtained directly
 267 by fitting, while parameters values δ_{eff} and ω_{eff} , required application of numerical cal-
 268 culation methods. Parameter ω_{eff} is the integral of the normalized glow curve. Then, the
 269 following values were calculated: l from equation (9) and μ_s from the $\delta_{eff}/\omega_{eff}$ relation-
 270 ship. The resulting function $\mu_s(l)$ can be represented with the following empirical function
 271 with sufficient precision:

$$272 \quad \mu_s = \mu_{s0} + A_1 l^2 + A_2 \ln l \quad (10)$$

273 Coefficients, μ_{s0} , A_1 and A_2 are linear functions of Δ . It enables quick and accurate
 274 calculation of the values of kinetics order. The results are shown in SnplaceTable SnI. and
 275 Fig.3.

Table I. The values of coefficients for calculating the symmetry factor according to equation (10)

Δ	μ_{s0}	A_1	A_2
0.03755	0.402790	-0.021942	0.291690
0.06605	0.410165	-0.019808	0.286996
0.09865	0.418489	-0.017411	0.280375
0.14815	0.431729	-0.015532	0.275972

276 An iterative procedure must be established to calculate the value of kinetics order, using
 277 the diagram in Fig.3. First, a value of Δ is assumed (eg about 0.1). Then, the value of
 278 kinetics order l is calculated, with the use of the corresponding curve in Fig. 3. The value

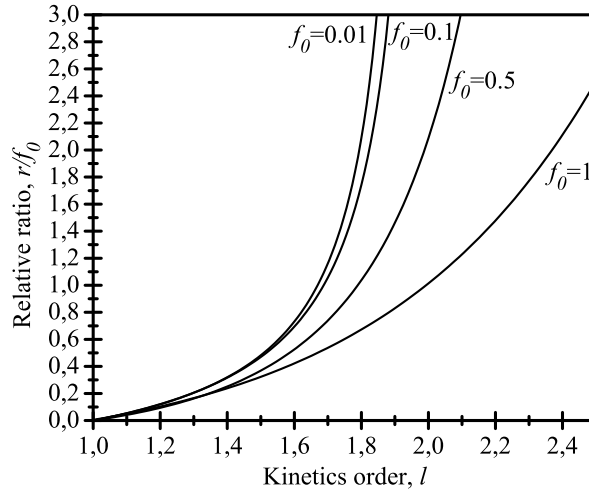


Fig. 2. The diagram shows how the ratio between the retrapping factor and filling factor depends on the kinetics order, for different filling factor values

279 of the activation energy E is calculated using equation (8). This value is substituted in the
 280 equation $2kT_m/E$ to calculate Δ , and its new value is calculated. The resulting new value
 281 of Δ is more accurate than the previous one. The new value is used in the next step. This
 282 procedure may be repeated until the value of kinetics order reaches the desired accuracy.
 283 Usually, three or four iterations are necessary, in order to achieve satisfactory accuracy.

284 For a complete analysis and determination of all parameters necessary for the recon-
 285 struction of the glow curve, the value of the frequency factor must also be calculated. The
 286 value of the frequency factor, for the model of first order kinetics is calculated from the
 287 equation:

$$288 \quad s = \frac{RE}{kT_m^2} \exp\left(\frac{E}{kT_m}\right) \quad (11)$$

289 where $R(K/s)$ is the heating rate of phosphor. However, it has been proved that it is not pos-
 290 sible to calculate values of the frequency factor for models of the second-order [33], mixed
 291 order [7] and general order [5, 13]. The derived theoretical equations enable calculation of
 292 only a pre-exponential factor, in which the frequency factor is one of the factors.

293 The frequency factor determines the probability of emission of electrons from electron
 294 traps, for high values of the heating temperature. Then the value of the exponential factor
 295 $\exp(-E/kT_m)$ is approximately equal to one. When, there is a high concentration of empty
 296 traps in the phosphor material and when the value of the retrapping factor is not negligible
 297 then a significant number of electrons are retrapped. This can be seen on the glow curve,

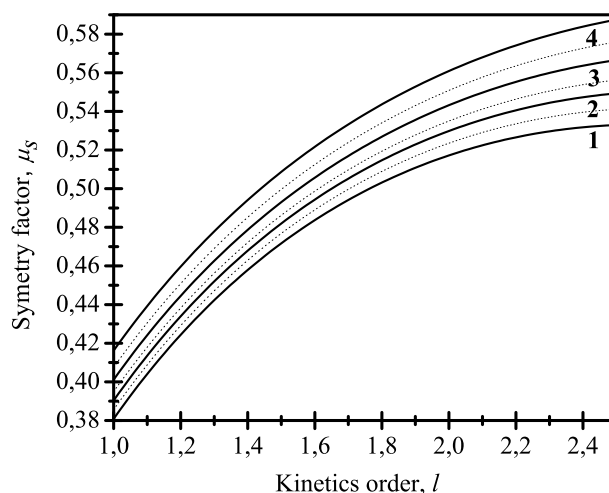


Fig. 3. Dependence of the symmetry factor of kinetics order, for the model of ideal phosphors, for the following values: 1- $\Delta=0.03755$, 2- $\Delta=0.06605$, 3- $\Delta=0.09865$ and 4- $\Delta=0.14815$. The dotted line shows the curves obtained by linear interpolation, which are the values of Δ equal to half the sum of Δ of two neighboring curves.

298 as the TL peak shifts to higher values as a result of reducing the velocity of luminescent
 299 recombination. Since the filling factor of traps is not known, it is not possible to make a
 300 correction of its influence on the rate of TL relaxation.

301 In the initial stage of relaxation, the relative impact of retrapping is very small, because
 302 the total amount of electrons, which are retrapped in phosphor, is small. This means that, in
 303 the initial phase, the glow curve does not depend on the retrapping factor and the filling fac-
 304 tor. Therefore, the temperature of the maximum will be shifted towards lower temperature
 305 values, because significant redistribution of the electrons from the trap will be performed
 306 only after the maximum. In this case, the correction, that needs to be done, depends on the
 307 kinetics order l and factor Δ . For the range of kinetics order values between 1 and 2, the
 308 following expression is obtained [34]:

$$309 \quad s = \frac{R}{\delta_{eff} [2 - l + \mu_s f_0 (l - 1)]} \exp\left(\frac{E}{kT_m}\right) \quad (12)$$

310 For the value of $f_0=1$, in the phosphor material, there are no empty traps and then the
 311 above formula may be used to calculate values of the frequency factor. In other cases, the
 312 formula can be used to calculate the pre-exponential factor.

313 Models of the first and second order kinetics are special cases of the kinetics model of
 314 ideal phosphors. For the first-order kinetics model, after substitution of $l=1$, equation (12)
 315 becomes the same as equation (11). For the second-order kinetic model, after substitution

316 of $l=2$, the corrective factor of equation (12) becomes $1+0.9\Delta$. In the theory of this model,
 317 this factor should be $1+\Delta$. The difference between corrective factors is negligible, because,
 318 in both cases, they are obtained by approximation, and the values of Δ are very small.

319 8 Results

320 The algorithm for calculating the kinetics parameters of TL, for the model of ideal phosphor,
 321 was tested on a number of computer-simulated glow curves. Values of parameters
 322 characterizing phosphors (r, E, s) and the parameter f_0 , which determines the initial value of
 323 relaxation were selected for the model of ideal phosphors shown by differential equation
 324 (6). For simplicity of calculation, it was taken that in all cases the heating rate of phosphors
 325 is $R=1\text{K/s}$. The parameter values were chosen so that the values of order kinetics were
 326 between 1 and 2, and corresponded to the values that characterize phosphors in dosimetry.
 327 These values, for some characteristic cases, are shown in Table II. The r/f_0 ratio is separately
 328 shown in the Table, due to its special relevance in the calculation of values of kinetics order.
 329 Simulated glow curves were obtained for the parameter values given in Table II, and using
 330 the numerical method of Runge Kutta IV order to solve differential equation (6). Glow
 331 curves were normalized, and then adjusted with improved GL functions of the exponential
 332 type. Results of fitting are shown in Table III.

Table II. The values of parameters, for computer simulation of glow curves, for the model of ideal phosphor

No	$E(eV)$	$s(s^{-1})$	f_0	r	r/f_0
1	2.4	10^{25}	1	0.075	0.075
2	0.4	10^{13}	0.1	0.01	0.1
3	1.5	10^9	0.01	0.0005	0.05
4	0.1	10^9	0.1	0.003	0.03
5	1.6	10^5	0.1	0.001	0.01
6	0.4	10^5	0.5	0.025	0.05
7	1.5	10^9	0.01	0.008	0.8
8	0.4	10^5	0.5	0.45	0.9
9	0.1	10^9	1	0.55	0.55
10	1.6	10^5	1	0.9	0.9

333 In Table III, χ^2 or chi-square is a quantity that determines the quality of fitting.

334 Displaying the glow curve in analytical form allows precise determination of parameters,
 335 which is especially important for the T_m value, since it is in an exponential function.
 336 Also, the value δ_{eff} was calculated more easily and more accurately. This value is very
 337 difficult to calculate precisely, because glow curves asymptotically approach the zero value
 338 at the end of the relaxation process. Parameter values δ_{eff} , ω_{eff} and μ_s were numerically

Table III. Results of fitting glow curves, from Table II., with the GL function

No	T_m	ω	z	z_1	z_2	b_0	b_1	χ^2
1	466.67	19.00	0.0558	0.00923	8.23×10^{-5}	-0.2855	3.34×10^{-3}	2.22×10^{-6}
2	147.55	11.32	0.1130	0.01368	1.58×10^{-4}	-0.2408	4.11×10^{-3}	3.07×10^{-6}
3	721.79	69.38	0.0093	0.00232	6.17×10^{-6}	-0.2729	8.70×10^{-4}	2.27×10^{-6}
4	53.653	5.657	-0.0543	0.02887	1.16×10^{-3}	-0.2901	0.01181	2.25×10^{-6}
5	1174.0	168.8	0.0086	3.60×10^{-4}	7.77×10^{-7}	-0.2808	-1.44×10^{-4}	8.08×10^{-7}
6	317.76	49.06	0.0317	3.10×10^{-3}	1.26×10^{-5}	-0.2305	8.85×10^{-4}	1.68×10^{-6}
7	734.64	87.74	0.3906	9.73×10^{-4}	-1.62×10^{-6}	-0.0303	-3.37×10^{-4}	5.52×10^{-7}
8	322.68	65.00	0.4236	1.42×10^{-3}	-2.68×10^{-6}	0.0598	-1.01×10^{-3}	5.52×10^{-8}
9	53.382	7.166	0.4031	0.01068	-2.70×10^{-4}	0.0069	-5.47×10^{-3}	2.00×10^{-7}
10	1166.7	228.3	0.3445	2.16×10^{-4}	-3.23×10^{-7}	0.1088	-1.30×10^{-4}	1.85×10^{-7}

339 calculated and are shown in Table IV. Romberg's method was used for the calculation of
 340 δ_{eff} and ω_{eff} integrals.

341 Based on the parameter values of δ_{eff} , μ_s and T_m given in Tables III and IV, and us-
 342 ing the previously described procedure for calculation, it is possible to calculate values of
 343 parameters of glow curves E , l , r , Δ and s . Table V shows the calculated values of these
 344 parameters E_c , l_c , r_c , Δ_c and $s^{(0)}$. The relative errors of calculation of parameters E and r
 345 can be determined, since their exact values are known. For parameter s , it is possible only
 346 when $f_0=1$, as then values of the pre-exponential and frequency factor are identical. The
 347 exact values of parameters l and Δ are not known, so it is not possible to calculate their
 348 relative errors. The accuracy, with which it is possible to calculate individual parameters
 349 can be estimated by analyzing values of the relative error. This analysis also provides an
 350 assessment of the validity of the method used for the calculation of parameters [34].

Table IV. Calculated values of glow curve parameters

No	ω_{eff}	δ_{eff}	μ_s
1	21.911	9.1504	0.41762
2	12.940	5.5918	0.43213
3	78.701	33.135	0.42102
4	6.3718	2.6382	0.41404
5	182.63	75.432	0.41304
6	54.967	24.011	0.43683
7	99.790	50.150	0.50255
8	73.288	39.206	0.53495
9	8.1341	4.1886	0.51494
10	259.08	141.77	0.54718

351 Since in practice it is not possible to calculate the values of parameters r and s , if the
 352 value of f_0 is not known, then these values are calculated for an assumed value of $f_0=1$. It
 353 has no effect on the shape of the curve, because it depends on the r/f_0 ratio. For a calculated

Table V. Calculated values of thermoluminescence parameters

No	l_c	Δ_c	$E_c(eV)$	$dE/E(\%)$	$s^{(0)}(s^{-1})$	$ds/s(\%)$	r_c	$dr/r(\%)$
1	1.174	0.0334	2.408	-0.32	1.23×10^{25}	22.528	0.08697	-15.96
2	1.190	0.0637	0.399	0.16	8.83×10^{12}	-	0.01133	-13.28
3	1.110	0.0827	1.504	-0.26	1.02×10^9	-	0.00059	-18.38
4	1.064	0.0924	0.100	-0.04	9.84×10^8	-	0.00334	-11.49
5	1.015	0.1267	1.597	0.17	9.63×10^4	-	0.00072	27.74
6	1.105	0.1368	0.400	-0.08	9.89×10^4	-	0.02190	12.38
7	1.621	0.0842	1.503	-0.20	5.91×10^8	-	0.00801	-0.112
8	1.748	0.1390	0.400	-0.03	6.97×10^4	-	0.45418	-0.929
9	1.707	0.0919	0.100	-0.07	1.02×10^9	2.1384	0.54636	0.662
10	1.929	0.1260	1.596	0.25	9.49×10^4	-5.1196	0.88050	2.167

354 value of the kinetics order, one can select an r/f_0 ratio, in order to obtain the curve shape
 355 which is the same as the shape of the experimental curve. The value of $f_0=1$ is chosen,
 356 because, in this case, it is possible to calculate the value of the frequency factor. This is just
 357 one, of many possible solutions, for the given experimental curve, when the value of f_0 is
 358 not known, and the value of kinetics order is between 1 and 2. In this way, the obtained
 359 solution, for parameter s , has a minimum possible value, while for parameter r , a maximum
 360 possible value. Physically this means that the actual value of parameter s , is greater than
 361 the calculated value and the value of parameter r is lower.

362 Based on the calculated values of kinetic parameters of the process, it is possible to
 363 reconstruct the real physical process using differential equation models and calculated pa-
 364 rameters. This allows examination of the kinetics of real TL phosphors. First, the glow
 365 curve corresponding to the model of ideal phosphor should be identified. This means that
 366 the curve should meet the requirements described in this paper. The conditions are that
 367 the glow curve has only one maximum and it needs to meet the criterion determined by
 368 the geometric parameters (shape factor and asymmetry factor), for a given value of pa-
 369 rameter Δ . Among phosphors, which are used in dosimetry, TLD700H(⁷LiF:Mg,Cu,P),
 370 TLD400(CaF:Mn) and TLD500(Al₂O₃:C) are the closest to fulfilling the conditions. The
 371 tests proved that the dominant peak for TLD700H phosphor, approximately meets all re-
 372 quirements. The results are shown in Fig.1.

373 For typical glow curve of the dosimeter TLD700H [34] the same procedure of calcula-
 374 tion is applied as well as the simulated glow curves. When the dominant peak is adjusted
 375 with GL function, the following values were obtained: $T_m=482.75$, $\omega = 19.58$, $z = 0.1259$,
 376 $z_1 = 2.79 \times 10^{-3}$, $z_2 = -4.99 \times 10^{-5}$, $b = -0.2722$, $b_1 = 8.56 \times 10^{-5}$, $\chi^2 = 1.32 \times 10^{-5}$. Then,
 377 values $\omega_{eff} = 21.871$, $\delta_{eff} = 9.0078$, $\mu_s = 0.41187$ were calculated. Heating rate of
 378 phosphor was $R=1K/s$. Finally, values of TL parameter were calculated $l_p = 1.145(1.15)$
 379 $\Delta_p = 0.0326(0.03245)$, $E_p = 2.553eV(2.564)$, $s_p^{(0)} = 5.53 \times 10^{25}$, $s^{-1}(7.11 \times 10^{25} s)$,

380 $r_p^{(0)} = 0.07324(0.06776)$.

381 The obtained parameter values were substituted in the differential equation (3). The
 382 differential equation was solved numerically with these parameters to obtain the glow curve.
 383 The results are shown in Fig. 4. The figure shows that the glow curve completely coincides
 384 with the experimental curve. It can be concluded that the dominant peak of the glow curve,
 385 for TLD700H phosphor, may be described using kinetics of the model of ideal phosphor.

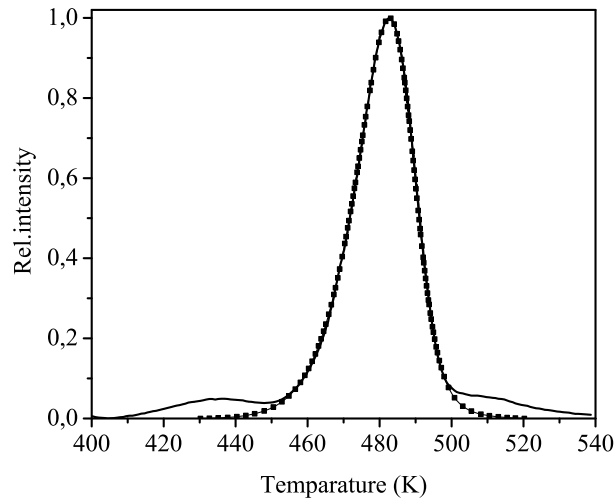


Fig. 4. Experimental TL glow curve for a TLD700H dosimeter (${}^7\text{LiF: Mg, Cu, P}$) (solid line) and glow curve obtained by calculation using the model of ideal phosphor (solid square) (taken from paper [34])

386 9 Discussion

387 Application of GL functions to analyze TL curves with a maximum allows detection and
 388 quantification of changes in asymmetry and shape of the glow curve. Compared to the cur-
 389 rent glow curve analysis, based only on its asymmetry, GL functions allow definition of
 390 their type, ie, how much they deviate from the pure form of Gaussian and Lorentz func-
 391 tions. First order kinetics is an exponential decay, and then the TL curve is an asymmetric
 392 Gaussian. Second order kinetics is a decay, which may be approximately described by the
 393 hyperbolic function. Since the glow curve of the second-order kinetics model, in the very
 394 beginning, has an exponential form, and in the second part, a hyperbolic form, then it can
 395 be described with a function, that is a combination of Gaussian and Lorentz functions. GL
 396 functions analysis confirms this fact fully.

397 Analysis of the model, that describes non-interactive kinetics, refers to glow curves

398 whose kinetic processes can be completed with first order kinetics or second order kinetics.
399 Kinetics, at the end of the TL process, in case of the model of ideal phosphor ($m=0$), is
400 of the second order and in the case of a mixed-order model ($r=1$) is of the first order. This
401 means, that for the model of ideal phosphor, kinetics of the first order is an unstable process,
402 while kinetics of the second order is stable. For the model of ideal phosphor, changes in the
403 vicinity of kinetics of first order are faster and larger than in the vicinity of kinetics of the
404 second order. Thereby, the shape and asymmetry of glow curves changed. For mixed-order
405 kinetics, all this is opposite. This ensures that in the entire field of non-interactive kinetics,
406 kinetic processes in models, of ideal phosphor, mixed order, and all the rest, belonging to
407 non-interactive kinetics, can be mutually different.

408 When the model of TL relaxation is identified, based on an established criterion, it is
409 necessary to calculate parameters of the TL process. This paper proposes a procedure for
410 calculating the kinetic parameters for the simplest model. It is the model of ideal phosphor.
411 A method for calculating parameters was based on the concept of kinetics order. The kinet-
412 ics order parameter was defined by an equation, for which there is evidence that it will be
413 valid for models that are part of the non-interactive kinetics model. These are models that
414 have analytical solutions for corresponding differential equations (models of first, second,
415 general and mixed order kinetics). The general order kinetics model can be treated as an
416 approximation of the model of ideal phosphor. It has been assumed that the kinetics order
417 parameter, is not characteristic of the TL process, but the glow curve is. In this way, vari-
418 ation of the value of kinetics order parameter during TL relaxation is avoided. The same
419 procedures are used for the development of techniques for calculating the parameters of
420 the model of ideal phosphor as in the case of calculating parameters for the general order
421 kinetics model. The main task was to ensure that the obtained parameter values satisfied
422 the differential equation of the model of ideal phosphor. In this way, the resulting solutions
423 describe the physical process, and provide an assessment of the model.

424 The developed calculation method enables obtaining of the value of activation energy,
425 with the relative error of calculation less than 0.5%. Values of the retrapping factor and
426 frequency factor cannot be determined based on experimentally obtained glow curves. This
427 would not be possible, unless the value of the filling factor is previously known even if
428 there is an analytical solution of the differential equation, Therefore, a given glow curve
429 has a great number of solutions. Any solution can be chosen, but it is convenient to choose
430 a solution that allows the simplest analysis of the kinetic process, i.e. when the value of the
431 filling factor is $f_0=1$. This solution was chosen because it is valid for the limiting case and
432 defines the limits of validity of the model. Changes in the value of Δ , for a range of values
433 of l from 1 to 2, are very small and can be ignored. This is important, in order to obtain
434 high calculation accuracy. Then it can be relatively easy to apply an empirical formula for
435 calculating the frequency factor.

436 Criteria for assessing the kinetics model and algorithm for calculating the TL kinetic
437 parameters, which are presented in this paper may be very useful in analyzing and un-

438 derstanding the kinetics of the TL process. This is demonstrated by analysis of the three
439 types of widely available and applied dosimeters. These dosimeters have commercial codes
440 TLD400, TLD500 and TLD700H [10,16,26,27]. Glow curves of these dosimeters have
441 dominant and isolated peaks. These peaks were analyzed and they originated only from
442 one kind of traps. The results are very interesting and cannot be obtained by visual as-
443 sessment. All three dosimeters have glow curves, which are visibly asymmetric. It has
444 been estimated that these curves can be described with the general order kinetics model
445 and kinetics order values between 1 and 2. If the values of the asymmetry factors are ana-
446 lyzed only, significant differences between the three curves cannot be identified. However,
447 analysis of the values of the shape factor revealed important differences. The TLD700H
448 dosimeter glow curve takes the form very similar to the curve of the model of ideal phos-
449 phor. This is quite unexpected, since in addition to the dominant peak there is a plurality of
450 small peaks. These small peaks originated from other types of active traps and their impact
451 on the dominant peak is negligible. The reason for this is that the small peaks are substan-
452 tially smaller than the dominant peak, and the value of the retrapping factor for traps, from
453 which the dominant peak arises, is very small. The glow curve of TLD500 corresponded to
454 the model of non-interactive kinetics with a relatively high concentration of inactive traps.
455 It may be noted that the value of the retrapping factor was greater than one, which means
456 that the probability of retrapping was greater than the probability of luminescent recombi-
457 nation. However, because of the relatively high concentration of non-active traps, the glow
458 curve takes a form, which can be described with the values of order kinetics between the 1
459 and 2. The TLD400 dosimeter has a low value of the retrapping factor, but not negligible.
460 This value becomes more important when the active traps are more depleted. Also, the
461 relatively low concentration of inactive traps has a significant influence on the shape of the
462 curve when traps are less populated. It can be concluded that TL relaxation of the TLD400
463 dosimeter cannot be described with the model of ideal phosphor, or the mixed order kinetics
464 model.

465 The analysis showed that the algorithm for calculating glow curve parameters, for the
466 model of ideal phosphors, can be applied only to the dominant peak of the glow curve of the
467 TLD700H dosimeter. In practice, kinetics parameters, in this case of phosphor, are often
468 calculated using the first-order kinetics model or the general order kinetics model. Some
469 authors, using deconvolution, proved that the shape of the dominant peak corresponds to the
470 model of first order kinetics. As small peaks overlap with the dominant peak, it is possible,
471 in the deconvolution process that such changes in shapes of small peaks will make the shape
472 of the dominant peak correspond to the curve for the first order kinetics model. However,
473 most authors reported that deconvolutions with a general order kinetic model obtained better
474 compatibility with the experimental curve. Since the model of general-order kinetics is an
475 approximation of the model of ideal phosphor, better accuracy and compatibility with the
476 experimental curve is expected when the model of ideal phosphor is applied. The following
477 values were obtained for kinetic models of the first order and general order, for the dominant

478 peak of the TLD700H dosimeter:

479 1. first order kinetics: $l=1, E=2.229\text{eV}, \Delta=0.0373, s=3.56 \times 10^{26}\text{s}^{-1}, r=0,$

480 2. general order kinetics: $l=1.18, E=2.628\text{eV}, \Delta=0.0317, s^{(l)}=3.54 \times 10^{26}\text{s}^{-1}$

481 This analysis shows why, when using the model of general order kinetics, accurate
482 results, or very approximate, or incorrect with a relatively large error, can be obtained. The
483 general order kinetics model is designed to simply and as accurately as possible describe
484 the asymmetry of the curves. If the experimental glow curve coincides, in shape, with the
485 glow curve formed based on this model, then the values of the calculated parameters will be
486 accurate. If these shapes differ slightly, then, in the fitting process, the calculated parameter
487 values shift slightly, in order to correct for variations in the value of the shape factor. If
488 the difference between the shape factor of the theoretical glow curve and its actual value is
489 greater, deviation from the exact value of TL parameters will be higher.

490 10 Conclusions

491 In this paper a completely new method for calculating the kinetic parameters of the process
492 relating to TL relaxation in ideal phosphors was developed. This process is close to real
493 situations as it relates to the model with realistic physical parameters.

494 A method for calculating the parameters of TL relaxation for the model of ideal phos-
495 phors is based on the concept of kinetics order. The kinetics order is defined by an equation
496 applied to models of the first, second, general and mixed order kinetics. The new definition
497 includes also a new meaning for the parameter of kinetics order. The kinetics order, in this
498 case, cannot be understood as a parameter that defines the TL process, but as a parameter
499 that determines the glow curve.

500 Using the GL asymmetric function, it was revealed that the dominant peak of a TLD700H
501 ($^7\text{LiF:Mg,Cu,P}$) dosimeter, could be described by the model of ideal phosphors. The values
502 of physical parameters, which determine the kinetics of TL were evaluated based on the
503 method for calculating parameters of the model of ideal phosphor. Values of the re trapping
504 factor, frequency factor and activation energy were calculated for the assumed value of the
505 filling of traps of $f_0=1$. The resulting glow curve coincided with the experimental curve.

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