EAST EUROPEAN JOURNAL OF PHYSICS. 2. 182-186 (2023)
DOI:10.26565/2312-4334-2023-2-18 ISSN 2312-4334

THE THERMOLUMINESCENCE PARAMETERS OF IRRADIATED K-FELDSPAR†

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Received October 31, 2022; revised March 10, 2023; accepted March 13, 2023

Isothermal decay of the TL glow curve has been studied at ambient temperature. Heating of feldspar at 600ºC leads to increased sensitivity of the samples upon irradiation for the whole range of glow curve. In general, we observe a sensitivity increase of about five times. Fading of the glow curve is observed at the low-temperature part of the glow curve while it has been kept in the dark at the ambient, constant temperature. After a certain period, approximately in 40 to 50 days the low-temperature region of the glow curve fades down while the high-temperature part remains unchanged. Peaks at the low-temperature region of the TL glow curve were isolated by the curve subtraction method. Activation energy and frequency factor parameters of the isolated peaks were calculated, taking first and second-order kinetics into account. The values of the calculated activation energy vary between 0.7 to 1.1 eV, and frequency factor values of the isolated peaks change within the order of 10^9 to $10^{13}s^{-1}$. The μ values clearly indicate that all isolated peaks are more likely to be second-order kinetics.
Keywords: *Feldspar; Isothermal decay; Activation energy; Frequency factor*
PACS: 78.60.Kn

INTRODUCTION

Feldspar fraction extracted from sediments has been increasingly investigated for the dating of archeological artifacts and the age of geological sediments. The main advantage of using feldspars instead of quartz is their higher luminescence brightness and saturation dose, which could extend the range of dating applications. At the same time, the luminescence emission of feldspar exhibits a large variety due to the substantial natural abundance of the mineralogical composition [1-3]; the presence of adverse luminescent phenomena like anomalous fading and sensitivity changes; irradiation and thermal history [4]. Investigations were mainly made on natural K-feldspars separated from geological sediments and pure feldspar minerals. They were devoted to clarifying the luminescence behavior of such materials, describing the optical bleaching mechanism [5], identifying traps and recombination centers, and assessing a reliable protocol for dating procedures [6-9].

The exact composition of TL luminescence curves in feldspars and their kinetics is still an open question, although numerous studies have been carried out. In particular, it is necessary to find out whether the broad TL luminescence curves consist of individual narrow TL peaks or are the result of a superposition of continuous energy levels [7].

To contribute to the knowledge of the luminescent mechanisms in feldspars, we present the results of a study of activation energy and frequency factor values on the natural feldspar mineral.

MATERIALS AND METHODS

We have taken the feldspar kindly presented by the Institute of Inorganic Chemistry and Catalysis ANAS as a sample. Samples were gently crashed and sieved, and the fraction of 100-160 µm of feldspar was used for TL measurement. Hydrochloric acid (1N) was used to remove carbonates and then rinsed with deionized water. Magnetic separation was applied to remove any magnetic inclusions. Any high-density components were separated using heavy liquid Sodium poly-tungsten. Irradiation was performed at ambient temperature with a ^{60}Co gamma source with a dose rate of 3.73 mGy/s. The dose rate of the ^{60}Co source was determined using a Magnette Miniscope MS400 EPR spectrometer using individually packed BioMax alanine dosimetry films with barcode markings (developed by Eastman Kodak Company). For the experiments, it was used both natural and thermally treated feldspar samples to compare the changes in radiation sensitivity due to thermal treatment. Thermally treated feldspar was heated at 600ºC for one hour before irradiation.

The Harshaw TLD 3500 Manual Reader is used to measure the characteristics of TL samples. TL measurements were performed using a linear heating rate of 20ºC/s from 50ºC to 400ºC. Three aliquots of 5 mg of each sample were used for each measurement. TL data points represent the average of three different aliquots of the sample. A thin and uniform layer of feldspar grains was laid on the planchet surface to get complete contact, ensuring a consistent TL signal from the sample.

Several experimental methods for evaluating the basic trapping parameters from a TL peak are based on the peak shape. The peak shape method enables the evaluation of "activation energy" and "frequency factor" using the temperature at the peak maximum (T_m) and the low (T_1) and high (T_2) half-width temperatures (Fig. 1) of the experimental glow curve.

The formulas for finding the activation energies by these methods usually contain one of the following factors: (a) $\tau=T_m-T_1$, the half-width at the low-temperature side of the peak, (b) $\delta=T_2-T_m$, the half-width towards the falloff of the glow peak, or (c) $\omega=T_2-T_1$, the total half-width. These formulas were developed and systemized by Chen R. [10]. Here are the summarized function and relevant coefficients (Table 1) for the first and second-order kinetics:

† *Cite as:* S. Mammadov, A. Ahadova, A. Abishov, and A. Ahadov, East Eur. J. Phys. **2**, 182 (2023), https://doi.org/10.26565/2312-4334-2023-2-18

$$E = C_{\propto}\left(\frac{kT_m^2}{\propto}\right) - b_{\propto}(2kT_m) \tag{1}$$

where k = 8.617×10^{-5} eV/K is the Boltzmann constant and α is δ, τ or ω respectively.

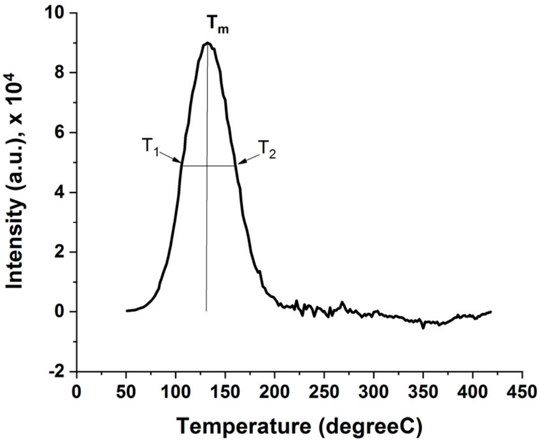

Figure 1. Isolated TL peak. Tm is a temperature at the peak maximum; T1 is a temperature at the point of the left side half-width; T2 is a temperature at the end of the right-side half-width

The values of C_α and b_α for each of the three methods are given in Table 1. Note that the order of kinetics of a peak is usually determined by its symmetry factor, defined as

$$\mu = {}^{\delta}/_{\omega}, \tag{2}$$

where a characteristic value of μ for the first order peaks is 0.42 and for the second order is 0.52 [11].

Table 1. Coefficients appearing in Eq.1 for the various methods of calculating activation energies [11]

	First order			Second order		
	τ	δ	ω	τ	δ	ω
$C_\propto$	1.51	0.976	2.52	1.81	1.71	3.54
$b_\propto$	1.58	0	1	2	0	1

Once the activation energy of a first-order peak is defined, the frequency factor can be determined by using the formula:

$$s = \frac{\beta E}{kT_m^2}\exp\left(E/kT_m\right), \tag{3}$$

where β (K/s) is the constant heating rate.
For the second-order kinetics, the formula needs to be written as:

$$s = \frac{\beta E}{kT_m^2\left(1+\frac{2kT_m}{E}\right)}\exp\left(E/kT_m\right) \tag{4}$$

RESULTS AND DISCUSSIONS

The continuum of energy states creates a broad TL glow curve shown in Figs. 2 and 3. This broad spectrum cannot be considered a single peak. It rather can be described as a linear combination of many narrower peaks, each corresponding to a separate but closely spaced energy level. These closely spaced energy levels can be identified by subtracting the TL luminescence curves. For example, Strickertsson et all [12] examined the TL signals from microclines and identified six overlapping peaks using the fractional glow technique to estimate the activation energy *E* as a function of the temperature *T*. Applying the T_{stop} –T_{max} method, they reported a continuous distribution of energies between E ~1.0 eV and E ~1.7 eV at the low-temperature region (below the 280°C). For higher T_{stop} values, the activation energy *E* was constant at a value of E ~1.75 eV [12].

In our case, we used the isothermal decay method. Several aliquots of the material are irradiated with a gamma dose of 34.12 Gy, and the TL glow curve is obtained. The process is then repeated several times using the remaining part of the aliquots after a certain period, i.e., after five days, 15 days, etc. This procedure produces a series of TL glow curves shown in Fig. 2 and 3, essentially corresponding to a gradual isothermal (i.e. at ambient temperature) cleaning of this sample's overall TL glow curve.

The comparison of Fig. 2 and Fig. 3 shows that heating of feldspar at 600ºC leads to increased sensitivity of the samples upon irradiation for the whole range of the glow curve. In general, we observe a sensitivity increase of about five times. Fading of the glow curve is observed at the low-temperature part of the glow curve while it has been kept in the dark at the ambient, constant temperature. After approximately 40 to 50 days, the low-temperature region of the glow curve fades down while the high-temperature part remains unchanged. It is a common phenomenon for irradiated feldspar and anomalous fading. Explanations of the anomalous fading effect have been based on various proposed models, such as the tunneling model, the localized transition model, and a model based on competition with radiationless transitions. Currently, the most accepted explanations of AF are based on quantum mechanical tunneling from the ground state or the excited state of the trap [13 and references therein].

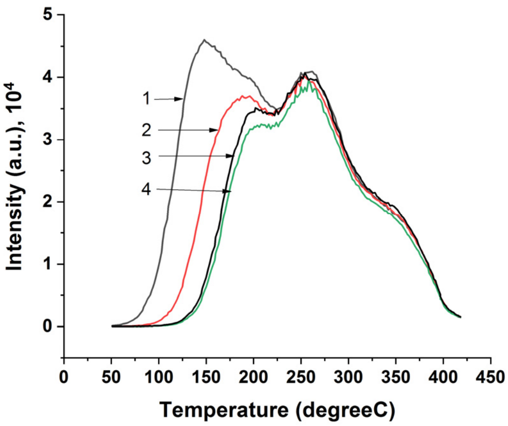

Figure 2. TL glow curve of unheated natural feldspar irradiated at 34.12 Gy (1); TL glow curve of the same sample after 15 days (2), 22 days (3), and 27 days (4)

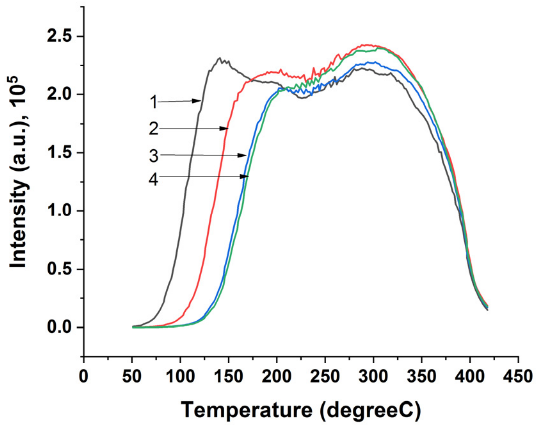

Figure 3. TL glow curve of natural feldspar heated at 600ºC for one hour and irradiated at 34.12 Gy (1); TL glow curve of the same sample after five days (2), 12 days (3), and 40 days (4). Curve (5) represents the isolated peak by subtracting curve (4) from curve (1)

Specifically, Figs. 4 and 5 show the results of subtracting the TL glow curve for the two samples as individual peaks.

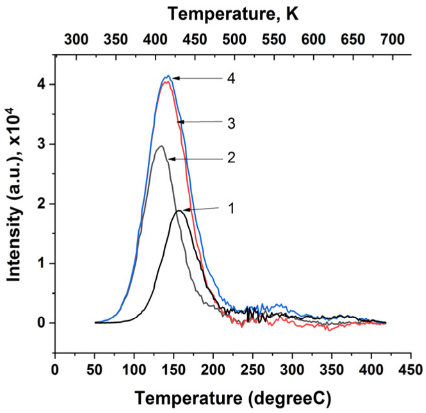

Figure 4. Isolated glow curves were obtained by subtracting method. Curve (1) represents the isolated peak by subtracting glow curve (4) presented in Fig. 2 from curve (2) presented in Fig. 2, i.e (1) = Fig 2 curve (2) – Fig.2 curve (4); and subsequently (2) = Fig 2 curve (1) – Fig. 2 curve (2); (3) = Fig 2 curve (1) – Fig. 2 curve (3); (4) = Fig 2 curve (1) – Fig. 2 curve (4)

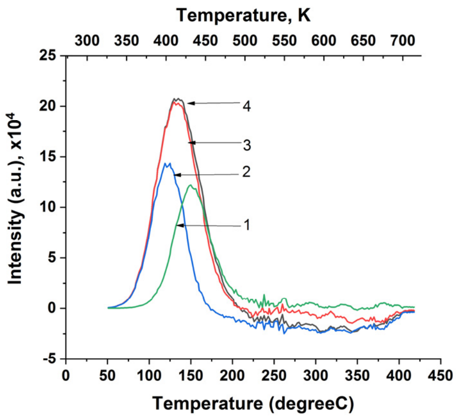

Figure 5. Isolated glow curves obtained by subtracting method: Curve (1) represents the isolated peak by subtracting glow curve (4) presented in Fig. 3 from curve(2) presented in Fig. 3, i.e (1) = Fig 3, curve (2) – Fig. 3, curve (4); and subsequently (2) = Fig 3, curve (1) – Fig.3, curve (2); (3) = Fig 3, curve (1) – Fig. 3, curve (3); (4) = Fig 3, curve (1) – Fig.3, curve (4)

Table 2 illustrates the major geometric parameters of the isolated peaks for the unheated and irradiated feldspar. The values of μ indicate that all isolated peaks belong to second-order kinetics.

Table 2. TL parameters of the isolated peaks for the unheated feldspar irradiated at 34.12 Gy (For the punctuation of the peaks, please, refer to Fig. 4)

Peak identity	T_m, K	T_1, K	T_2, K	ω	δ	τ	μ
1	429	405	457	52	28	24	0.54
2	408	381	433	52	25	27	0.48
3	414	385	444	59	30	29	0.51
4	414	385	447	62	33	29	0.53

However, one hardly observes curves with pure first-order or second-order kinetics. Experimental glow curves usually show values between 0.42 and 0.52 or around them (please refer to Tables 2 and 3). It is suggested [11] to interpolate the constants listed in Table 1 for the first and second-order kinetics to find activation energies. A much more suitable interpolation parameter seems to be μ which is found directly from the peak's geometrical shape (refer to Tables 2 and 3). We have to write general equations so that they would give the first-order case for μ=0.42 and the second-order

case μ=0.52. Thus, with the parameters of μ given in Table 1, the factors in equation 1 for the interpolated-extrapolated τ method would be
$C_\tau = 1.51 + 3.0\ (\mu-0.42)$; $b_\tau = 1.58 + 4.2\ (\mu-0.42)$
For the $\boldsymbol{\delta}$ and $\boldsymbol{\omega}$ method coefficients would be
$C_\delta = 0.976 + 7.3\ (\mu-0.42)$; $b_\delta = 0$ and
$C_\omega = 2.52 + 10.2\ (\mu-0.42)$; $b_\omega = 1$ respectively.

Table 3. Activation energy values of the isolated peaks for the unheated feldspar irradiated at 34.12 Gy (For the punctuation of the peaks, please, refer to Fig. 4)

Peak identity	E_τ (eV)	E_δ (eV)	E_ω (eV)
1	1.08	1.04	1.06
2	0.77	0.81	0.80
3	0.77	0.80	0.79
4	0.79	0.80	0.80

The frequency factor also easily might be calculated using the formula (3) for the first-order kinetics or the second-order kinetics by formula (4) [11]. Table 4 illustrates the values of the frequency factors calculated for the peaks listed in Table 3. The calculation of activation energy and frequency factor was performed using the user-defined function in the Matlab program.

Table 4. Calculated frequency factor values of the isolated peaks for the unheated feldspar irradiated at 34.12 Gy (For the punctuation of the peaks, please, refer to Fig.4)

Peak identity	Values calculated for τ method		Values calculated for δ method		Values calculated for ω method	
	1st order	2nd order	1st order	2nd order	1st order	2nd order
1	6.64E+12	6.22E+12	2.17E+12	2.02E+12	3.79E+12	3.50E+12
2	3.49E+9	3.20E+9	1.15E+10	1.05E+9	8.51E+9	7.82E+9
3	2.45E+9	2.60E+9	5.94E+9	5.45E+9	4.43E+9	4.07E+9
4	4.43E+9	4.07E+9	5.94E+9	5.45E+9	5.95E+9	5.45E+9

Table5. TL parameters of the isolated peaks for the feldspar heated at 600ºC and irradiated at 34.12 Gy(For the punctuation of the peaks, please, refer to Fig.5)

Peak identity	T_m, K	T_1, K	T_2, K	ω	δ	τ	μ
1	423	399	448	49	25	24	0.51
2	394	372	419	47	25	22	0.53
3	405	376	437	61	32	29	0.52
4	406	377	439	62	33	29	0.53

Table 6. Activation energy values of the isolated peaks for the feldspar heated at 600ºC and irradiated at 34.12 Gy (For the punctuation of the peaks, please, refer to Fig.5)

Peak identity	E_τ (eV)	E_δ (eV)	E_ω (eV)
1	1.00	1.01	1.01
2	0.98	0.97	0.98
3	0.75	0.76	0.77
4	0.75	0.77	0.76

Table 7. Calculated frequency factor values of the isolated peaks for the unheated feldspar irradiated at 34.12 Gy (For the punctuation of the peaks, please, refer to Fig.4)

Peak identity	Values calculated for τ method		Values calculated for δ method		Values calculated for ω method	
	1st order	2nd order	1st order	2nd order	1st order	2nd order
1	8.81E+13	8.29E+13	2.12E+13	2.12E+13	3.75E+13	3.52E+13
2	2.91E+9	2.67E+9	3.92E+9	3.60E+9	3.92E+9	3.60E+9
3	2.29E+9	2.09E+9	4.16E+9	3.82E+9	3.09E+9	2.83E+9
4	5.03E+12	4.71E+12	2.74E+12	2.56E+12	3.71E+12	3.47E+12

CONCLUSIONS

The decay of the luminescence curve with time was considered to be an isothermal decay at ambient temperature, and the proposed procedure makes it possible to identify peaks in the low-temperature region of the TL luminescence curve. An analysis of the values of the symmetry factor suggests that bimolecular mechanisms are based on the kinetics of decay processes since the values of this parameter vary within 0.52. The values of the calculated activation energy range between 0.7 to 1.1 eV, and frequency factor values of the isolated peaks change within the order of 10^9 to $10^{13}s^{-1}$.

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ПАРАМЕТРИ ТЕРМОЛЮМІНЕСЦЕНЦІЇ ОПРОМІНЕНОГО К-ПОЛЬОВОГО ШПАТУ

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Досліджено ізотермічний спад кривої світіння ТЛ при температурі навколишнього середовища. Нагрівання польового шпату до 600ºC призводить до підвищення чутливості зразків при опроміненні на всьому діапазоні кривої світіння. Загалом ми спостерігаємо підвищення чутливості приблизно в п'five разів. Згасання кривої світіння спостерігається на низькотемпературній ділянці кривої світіння при зберіганні в темряві при постійній температурі навколишнього середовища. Через певний проміжок часу, приблизно через 40-50 днів, низькотемпературна область кривої світіння згасає, а високотемпературна частина залишається незмінною. Піки в низькотемпературній області кривої світіння ТЛ виділені методом віднімання кривої. Розраховано параметри енергії активації та частотного фактору ізольованих піків з урахуванням кінетики першого та другого порядку. Значення розрахованої енергії активації змінюються в межах від 0,7 до 1,1 еВ, а значення коефіцієнта частоти ізольованих піків змінюються в межах порядку від 109 до 1013 $с^{-1}$. Значення μ чітко вказують на те, що всі ізольовані піки, швидше за все, є кінетикою другого порядку.

Ключові слова: *польовий шпат; ізотермічний розпад; енергія активації; частотний фактор*