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GEOCHRONOMETRIA 41(2) 2014: 168–177
DOI 10.2478/s13386-013-0148-z

Available online at
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KINETIC ANALYSIS OF THERMOLUMINESCENCE GLOW CURVES IN FELDSPAR: EVIDENCE FOR A CONTINUOUS DISTRIBUTION OF ENERGIES

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Received 18 August 2013 Accepted 25 November 2013

Abstract: The thermoluminescence (TL) glow curves from feldspars have been the subject of numerous studies, because of their importance in luminescence dating and dosimetry. This paper presents new experimental TL glow curves in a plagioclase feldspar, measured using the T_{max}–T_{stop} technique of glow curve analysis. Kinetic analysis of the experimental results is carried out for a freshly irradiated sample, as well as for a sample which has undergone optical treatment using infrared light for 100 s at 50°C. Application of the initial rise method of analysis indicates that the TL signals from both samples can be characterized by a continuous distribution of energy levels. By subtracting the TL glow curves measured at successive T_{stop} values, a series of TL glow curves is obtained which are analyzed using the empirical general order kinetics. It is found that all TL glow curves obtained by this subtractive procedure can be described accurately by the same general order parameter $b \sim 1.7$. In a second attempt to analyze the same TL glow curves and possibly extract information about the underlying luminescence process, the shape of TL glow curves is analyzed using a recently proposed physical kinetic model which describes localized electronic recombination in donor-acceptor pairs. Within this model, recombination is assumed to take place via the excited state of the donor, and nearest-neighbor recombinations take place within a random distribution of centers. This recent model has been used recently to describe successfully several types of luminescence signals. This paper shows that it is possible to obtain good fits to the experimental data using either one of these two approaches.

Keywords: : thermoluminescence in feldspars, kinetic analysis, feldspar glow curves, random defect distribution.

1. INTRODUCTION

Luminescence signals from feldspars have been the subject of extensive experimental and modeling studies during the past 20 years, due to the possibility of extending the range of luminescence dating techniques (Bøtter-Jensen *et al.*, 2003; Chen and Pagonis, 2011). Despite rather extensive recent research efforts, the characteristics of the traps associated with the thermoluminescence (TL) and infrared stimulated luminescence (IRSL) signals from these materials are still not well known. As pointed out in the recent comprehensive experimental work by Kars *et al.* (2013), these uncertainties are due to the complex nature of the defect energy levels and also due to the

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ISSN 1897-1695 (online), 1733-8387 (print)

presence of multiple charge transport mechanisms which complicate interpretation of experimental results (e.g. Poolton *et al.*, 2009; Jain and Ankjærgaard, 2011; Kars *et al.*, 2013). It is generally accepted that these materials exhibit anomalous fading phenomena due to quantum mechanical tunneling (Wintle, 1977; Visocekas, 1985), and that they contain a lattice defect with an excited state characterized by a strong IR resonance located ~1.45 eV above the ground state. Hence electrons trapped at the ground state can be excited with 1.45 eV IR and recombine with a trapped hole emitting luminescence.

Quantitative description of the trap responsible for IRSL signals from feldspars has focused on two types of trap depth, namely the optical trap depth and the thermal trap depth. Kars *et al.* (2013) reviewed previous investigations of the IR-trap in feldspar, and specifically focused on estimating the *optical trap depth* of the IR trap. This depth represents the amount of photon energy required for an electron to move from the ground state of the defect into the conduction band. This optical trap depth in feldspars has been estimated using various methods, to be either ~2 eV or ~2.5 eV. The IR-trap is also associated with a *thermal trap depth* which represents the thermal energy required to excite an electron from the ground state into the conduction band. This thermal energy has been estimated between 1.6 and 1.8 eV, although some estimates have been close to ~2 eV or higher (Clark and Sanderson, 1994; Strickerson, 1985; Li *et al.*, 1997; Huntley and Lamothe, 2001; Baril and Huntley, 2003; Huntley and Lian, 2006; Murray *et al.*, 2009; Andersen *et al.*, 2012; Li and Li, 2013).

In addition to these types of studies which provided estimates of the optical and thermal trap depths associated with the IRSL trap(s), other studies have examined the kinetics and possible correlations between TL and IRSL signals from feldspars. These additional types of studies typically examine the kinetics of TL glow peaks, as well as the effect of IR illumination on the TL signal. Several of these previous studies are summarized in the next section. Despite these extensive studies of the TL glow curves in feldspars, the exact composition of these glow curves and their kinetics are still an open research question. Specifically it is not yet known whether the typically broad TL glow curves in feldspars consist of individual narrow TL peaks, or whether they are the result of an underlying continuous distribution of energy levels.

This paper represents a detailed systematic attempt to describe the kinetics of TL glow curves from a feldspar sample, in the temperature region between room temperature and 450°C.

The specific goals of the present paper are:

1) To carry out a detailed kinetic analysis of a series of TL glow curves for a feldspar sample which has undergone a gradual and systematic thermal bleaching of its TL signal using the well-known T_{max}–T_{stop} method. The series of TL glow curves is analyzed using empirical general order kinetics algorithms, in an effort to describe the changes occurring in the shape of the TL glow curve.
2) To compare the results of the kinetic analysis from two samples; the first sample underwent gradual thermal bleaching only, while the second sample underwent a combined optical (IR) and thermal bleaching of its TL signal.
3) To suggest a new method of analyzing the series of TL glow curves, in which the methods of kinetic analysis are applied to the signal obtained by subtracting successive TL glow curves.
4) To explore the possibility that the changes occurring in the TL glow curves due to either thermal or to combined thermal/optical bleaching, can be described using the recent kinetic model by Jain *et al.* (2012) for localized electronic transitions. This type of analysis is carried out in an effort to obtain useful information about the underlying luminescence mechanism in this material.

2. PREVIOUS STUDIES OF TL GLOW CURVES IN FELDSPARS

This section presents selected previous studies of the shape and kinetics of TL glow curves in feldspars, of possible correlation between TL and IRSL signals and of the effect of IR illumination on the TL signal. This list is not meant to be exhaustive, but rather emphasis is given to kinetic studies similar or relevant to the experimental work described in this paper.

Strickertsson (1985) examined the TL signals from microclines and identified six overlapping peaks between room temperature and 500°C. This author used the fractional glow technique to estimate the activation energy E as a function of the temperature T_{stop} at which the sample was preheated. The results in his Figure 6 show clearly a continuous distribution of energies between E ~1.0 eV at T_{stop} ~70°C and E ~1.7 eV at T_{stop} ~280°C. For higher T_{stop} values between 280°C and 370°C the activation energy E was found to be constant at a value of E ~1.75 eV. In another pioneering study Bøtter-Jensen *et al.* (1991) performed combined TL and IRSL studies in feldspars and examined the effect of preheat and of infrared stimulation on the TL glow curves. Grün and Packman (1994) studied the kinetics of TL glow curves from Na and K-feldspar samples, and showed that all their samples followed non-first order kinetics. They also concluded that the TL glow curves in their K-feldspar samples consisted of a continuum of signals, rather than a sum of a finite number of components.

In an extensive experimental study Duller (1995) studied the effect of exposing four feldspar samples to infrared stimulation on the TL signal, and in particular showed which part of the TL signal is removed by exposure to IR and which part is directly related to the IRSL signal. Duller used also pulse annealing measurements and found a linear relationship between the amount of

lost TL and the integrated IRSL signal measured. Visocekas *et al.* (1996) studied TL glow curves from several types of feldspars which were X-irradiated at LNT, and used initial rise and fractional glow technique to evaluate the activation energy E as a function of the preheat temperature T_{stop}. The experimental results in their Figure 3 showed unambiguously the presence of a continuous distribution of E values, linearly increasing with the temperature T_{stop}. These continuous distributions were found in both ordered and disordered feldspar samples, and the typical range of their E values was from E ~0.4 eV at a T_{stop} ~200 K, up to E ~1.4 eV at a T_{stop} ~500 K. The authors concluded that these results were consistent with delocalized transitions and with the "built in disorder" expected in feldspars.

Chruścińska (2001) applied the fractional glow technique to optically bleached samples of K-feldspars; the experimental data in her Figure 11 shows also clearly a continuous distribution of energies E between ~1.0 and ~1.7 eV. In additional experimental work Chruścińska *et al.* (2001) applied the fractional glow technique to optically bleached samples of K-feldspars, and reported on various groups of traps which are active at the same temperature region above 300°C. These authors explained the diversity of TL bleaching efficiency on the basis of the different kinetic trap parameters. Visocekas and Guérin (2006) studied far-red TL emission from feldspars and the associated anomalous fading phenomena. Guérin (2006) studied the kinetics of high temperature TL from plagioclase feldspars and fitted the TL glow curve in preheated samples by a single peak characterized by close to second-order kinetics. However their more detailed study of TL growth with radiation dose and of phosphorescence experiments, provided evidence for a Gaussian distribution of energies.

Murray *et al.* (2009) pointed out the difficulty of relating the IRSL signal from feldspar to a particular region of the TL curve, and investigated two possible explanations for the experimental data, namely the existence of a distribution of energies for the sensitive traps, and the possibility of changes occurring in the recombination probability induced by IR exposure. They concluded that the main dosimetry trap(s) in feldspar are not reduced significantly by preheats of 60 s at 320°C, and tentatively identified the source of this IRSL as the trap responsible for a TL peak located around 410°C; furthermore they concluded that shallow traps in feldspar do not give rise to significant IRSL signals. In another relevant recent work Panzeri *et al.* (2012) studied the effects of prior IR stimulation on the TL emission from three feldspar minerals, and observed different types of behaviors in the reduction of the TL signal.

These previous studies provided ample experimental evidence for the existence of a continuous distribution of energies in feldspars, with typical thermal activation energies E between ~0.4 eV and ~1.7 eV. The results presented in this paper expand on these previous studies by addressing the specific goals listed in the previous section.

3. SAMPLE CHARACTERISTICS AND EXPERIMENTAL DETAILS

A museum specimen of feldspar (FL3) was used in this study. X-ray Diffraction analysis indicated that the major fractions in FL3 were andesine, while diopside was also present in the sample. Elemental concentrations were estimated using ICP-MS measurements, and sample FL3 was placed in the plagioclase feldspar series and details about the samples are given in Morthekai *et al.* (2012). The samples were crushed gently using agate mortar and sieved to obtain the 90-150 μm size fraction, which was used without any further chemical treatment. A few mg of samples were mounted on a stainless steel disc using Silkospray silicone oil.

A Risoe TL/OSL Reader DA-20 was used for all measurements in this study (Morthekai *et al.*, 2012). The IR bleaching was achieved by IR LEDs (870 ± 40 nm) and the luminescence emission was detected using a photomultiplier tube (EMI 9235QB; 30% QE at ~395 nm) with a combination of optical filters BG-39 (2 mm) and Corning 7–59 (4 mm). These filter combinations transmitted photons in the wavelength region 395 ± 50 nm. The heating rate was 2°C/s unless it is mentioned specifically, and heating was done in nitrogen atmosphere.

The TL glow curves of the feldspar samples are obtained using a T_{max}–T_{stop} thermal cleaning procedure, as follows. A single aliquot of the material is irradiated with a beta dose of 21.3 Gy, then subsequently heated up to a temperature T_{stop}, and cooled to room temperature. Immediately after, the aliquot is heated all the way to a high temperature of 450°C and the remaining TL glow curve is obtained. The process is then repeated several times by irradiating with the same dose and heating the same aliquot to a slightly higher temperature T_{stop} each time, in steps of 10°C for the complete interval T_{stop} = 150–380°C. This procedure produces a series of TL glow curves shown in **Fig. 1**, essentially corresponding to a gradual thermal cleaning process of the overall TL glow curve for this sample. For purposes of clarity, this aliquot will be referred to as "unbleached" aliquot in the rest of this paper.

This complete T_{max}–T_{stop} experimental procedure is carried out also on the same aliquot (after a thermal and optical wash) which is irradiated with the same dose and then is exposed to infrared light for 100 s at 50°C. This aliquot will be referred to as "IR-bleached" aliquot in the rest of this paper. This procedure produces a series of TL glow curves shown in **Fig. 2**, essentially corresponding to a gradual thermal cleaning process of the overall TL glow curve for this IR-bleached sample. The inset to **Fig. 2b** shows an example of direct comparison of the bleached and unbleached TL curves at a T_{stop} temperature of 200°C.

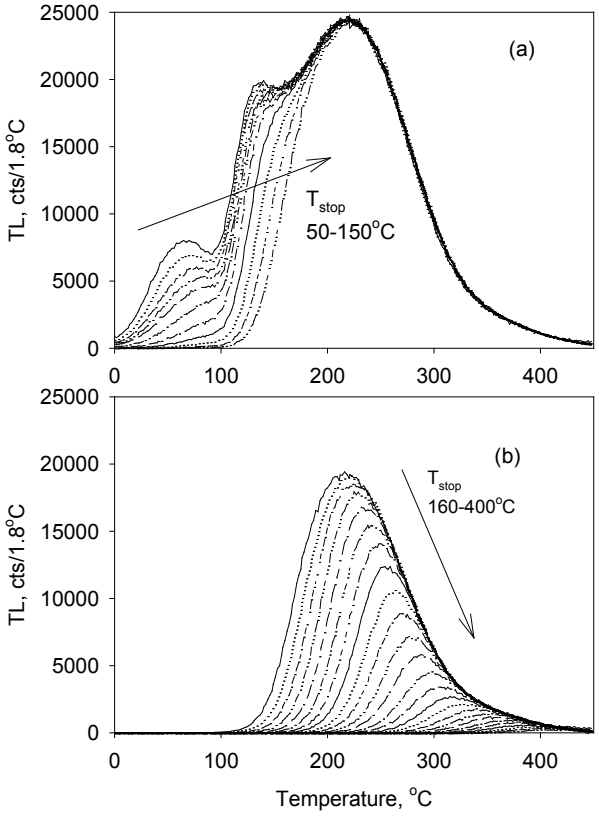

Fig. 1. *Experimental TL glow curves for unbleached samples at (a) low* T_{stop} *temperatures and (b) at higher* T_{stop} *temperatures.*

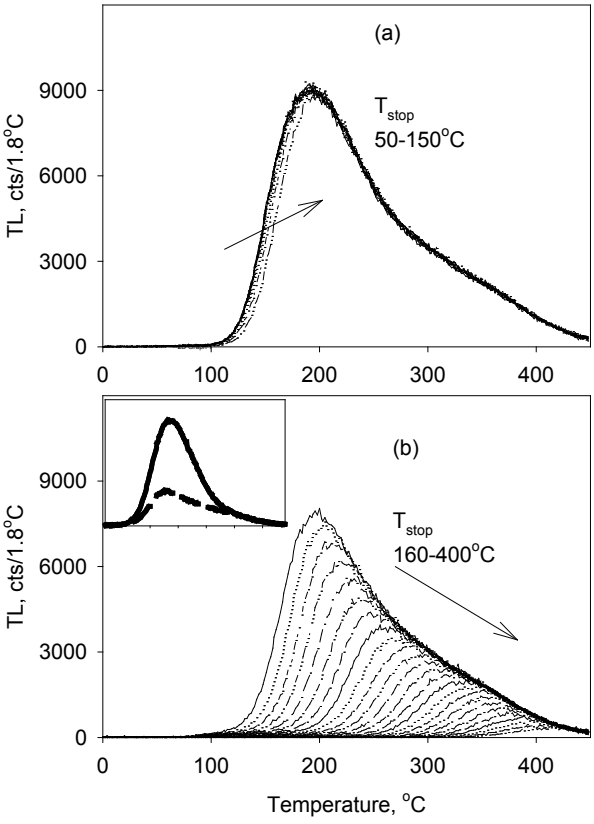

Fig. 2. *Same as Fig. 1, for IR-bleached samples. The inset to Fig. 2b shows an example of direct comparison of the bleached and unbleached TL curves at a* T_{stop} *temperature of 200°C.*

4. EXPERIMENTAL RESULTS

A closer look at the TL glow curves shown in **Figs. 1** and **2** indicates that as the material is heated towards progressively higher temperatures T_{stop}, the temperature of maximum TL intensity T_{max} shifts continuously towards higher temperatures; this behavior is known to be a strong indication of the presence of overlapping TL peaks, possibly forming a continuum of closely located energy states.

By performing an initial rise (IR) analysis on the TL glow curves in **Figs. 1** and **2**, the activation energy E is calculated for each temperature T_{stop}. A graph of E as a function of T_{stop} is shown in **Fig. 3**, for both the unbleached and IR-bleached samples. The shape of this E–T_{stop} graph for the unbleached sample (solid circles in **Fig. 3**), indicates the presence of a single energy level at E ~0.4 eV for low T_{stop} values, followed by an almost continuous gradual increase of the E values between 0.4 and ~1.5 eV up to T_{stop} ~260°C. The E values seem to change little at higher temperatures between T_{stop} = 260 and T_{stop} = 380°C, remaining constant at ~1.6 eV.

The distribution of E values shown in **Fig. 3** for the IR-bleached sample (open triangles) is slightly different, showing an almost constant energy E ~1.0 eV up to T_{stop} = 260°C, followed by a gradual increase of E between ~1.0 and ~1.5 eV. The E values for this sample also seem to change little between T_{stop} = 260 and T_{stop} = 380°C, as in the case of the unbleached sample.

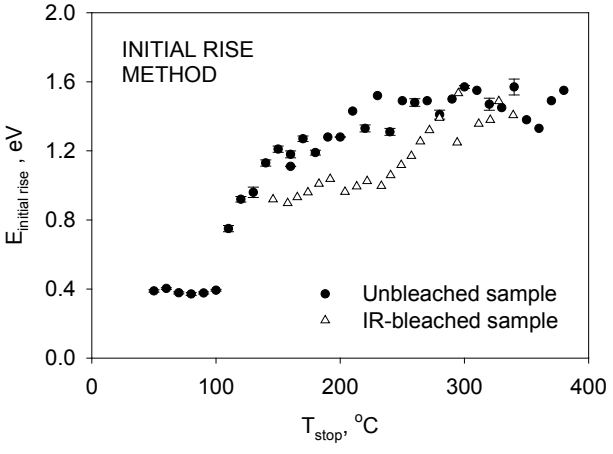

Fig. 3. E–T_{stop} *graph obtained by applying the initial rise method to the data in Figs. 1 and 2, for both unbleached and IR-bleached samples. The shape of this graph indicates the possibility of a continuous distribution of energy levels.*

The results from **Figs. 1–3** provide strong evidence for the possible presence of a continuum of E levels in both the unbleached and the IR-bleached samples. These results indicate that the TL glow curves in **Figs. 1** and **2** are in reality composites due to an underlying distribution of energy traps corresponding to different activation energies E and different amplitudes. In the next section we present a method which allows estimation of the amplitudes of these constituent TL glow curves.

A subtractive method of analyzing the TL glow curves

The continuum of energy states creates a very broad TL glow curve shown in **Figs. 1** and **2**. This broad glow curve cannot be analyzed as a single peak, but could possibly be described as the linear combination of many narrower TL peaks, each corresponding to a different but closely located energy level E. One possible method of identifying these closely located individual energy states is shown in **Figs. 4a** and **4b**, for the unbleached and IR-bleached samples correspondingly. Specifically **Figs. 4a** and **4b** show the results of subtracting the TL glow curve corresponding to $T_{stop} = 210°C$ from the glow curve for $T_{stop} = 200°C$ for the two samples.

The individual TL glow curves obtained in this subtractive procedure are shown in **Figs. 5a** and **5b**. Only every second subtractive result is shown in these figures, for purposes of clarity. The shape of the TL glow curves in **Fig. 5** can be described using the following empirical general order (GO) kinetic equation (Kitis *et al.*, 1998), in which the TL intensity $I(T)$ is given by:

$$I(T) = s'' n_0 \exp\left(-\frac{E}{kT}\right) \times \left[1 + \frac{s''(b-1)}{\beta}\int_{T_0}^{T}\exp\left(-\frac{E}{kT'}\right)dT'\right]^{\frac{b}{b-1}} \quad (4.1)$$

where E is the activation energy or trap depth (in eV), k — Boltzmann's constant (in $eV \cdot K^{-1}$), T — the absolute temperature (in K). The temperature varying as $T = T_o + \beta t$, where β — linear heating rate (in $K \cdot s^{-1}$), t — time (in s), and T_o is the temperature at time $t = 0$ (in K). The parameter n_0 represents the concentration of

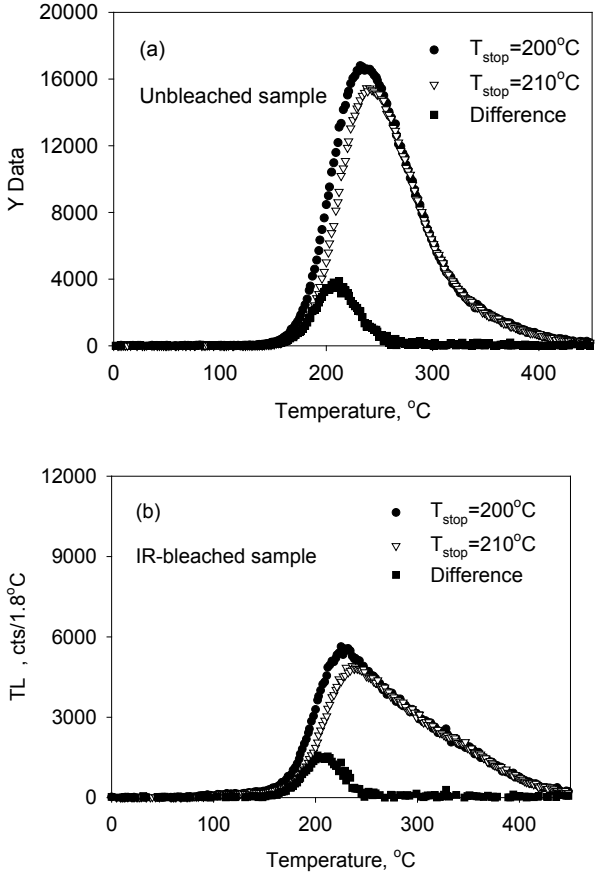

Fig. 4. *Subtraction of the TL glow curve corresponding to T_{stop} = 210°C from the glow curve for T_{stop} = 200°C for (a) the unbleached sample and (b) the IR-bleached sample. Some of the individual TL glow curves obtained using this subtractive procedure are shown in Figs. 5 and 7.*

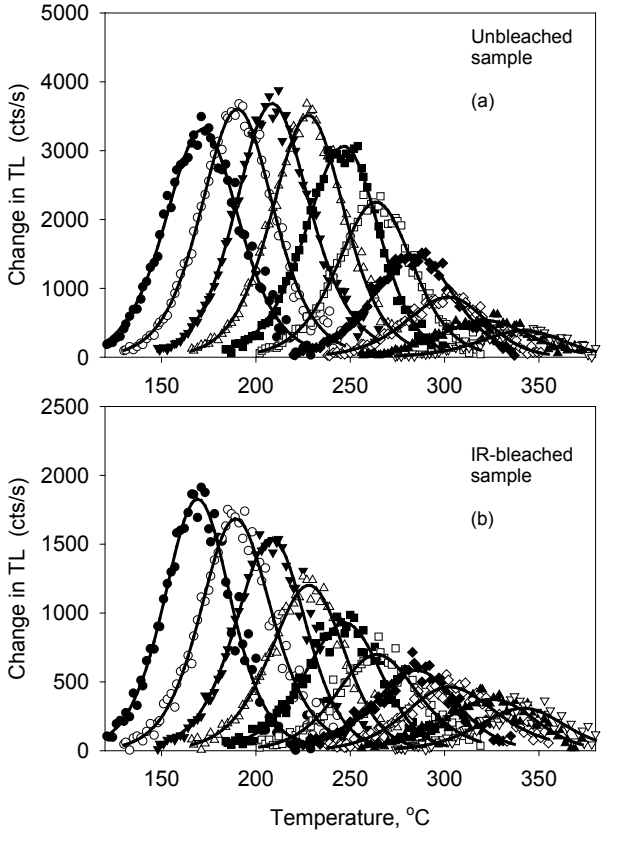

Fig. 5. *Empirical analysis of TL glow curves based on general order kinetics Eq. 4.2, for the (a) unbleached and (b) IR-bleached samples. Only every second subtractive result is shown in these figures, for purposes of clarity. The solid lines indicate the best fit using the empirical GO equation.*

trapped electrons at time $t = 0$ (in m^{-3}) and $s'' = s' \cdot n_0^{(b-1)}$ is an empirical parameter acting as an "effective" frequency factor for general order kinetics, b is the kinetic order parameter with values typically between 1 and 2.

By using a series approximation in **Eq. 4.1**, Kitis *et al.* (1998) showed that equation **Eq. 4.1** can be transformed into a convenient curve fitting algorithm, which contains two experimentally determined parameters, namely the maximum TL intensity I_M and the corresponding temperature T_M. These authors showed that **Eq. 4.1** can be written as:

$$I(T) = I_M \times b^{\frac{b}{b-1}} \times \exp\left(\frac{E}{kT} \times \frac{T - T_M}{T_M}\right) \times \left[1 + (b-1) \times \frac{2kT_M}{E} + (b-1) \times \left(1 - \frac{2kT}{E}\right) \times \left(\frac{T^2}{T_M^2} \times \exp\left(\frac{E}{kT} \times \frac{T - T_M}{T_M}\right)\right)\right]^{\frac{-b}{b-1}} \quad (4.2)$$

This equation is the fitting equation used to analyze the TL glow curves obtained by the subtractive procedure described above. There are only two free fitting parameters in **Eq. 4.2**, namely b and E, and the results of the best fitting procedure are shown as solid lines in **Fig. 5**.

The quality of the least square fits shown in **Fig. 5** (and also in **Fig. 7**) can be characterized by the R^2 value which varies from R^2=0.999 for low temperature glow curves, to R^2=0.990 for the high temperature curves. Alternatively, the quality of the fits can be expressed by the well-known Figure of Merit (FOM) which varies for a value of ~2% for the low temperature fits, up to a value of ~13% for the high temperature curves in **Figs. 5** and **7**.

The best fitting parameters are shown as a function of T_{stop} in **Fig. 6**. **Fig. 6a** shows the best fit E values obtained using **Eq. 4.2**, and the best fit activation energy E can be seen to increase continuously from E ~1.1 eV to E ~1.8 eV for the complete temperature range T_{stop} = 150–380°C and for both the unbleached and IR-bleached samples. **Fig. 6b** shows the best fit values of the kinetic order b, and the inset of this figure shows a histogram distribution of b values from all T_{stop} values and for both samples. It is concluded that all TL glow curves obtained by this subtractive procedure can be described accurately by the same general order parameter $b = 1.65 \pm 0.26 (1\sigma)$. Finally **Fig. 6c** shows the maximum height of the TL glow curves as obtained from the best fit procedure; it is seen that the distribution of heights I_{max} for the unbleached samples shows an initial increase in the interval T_{stop} = 150–200°C, followed by a gradual decrease to zero at higher T_{stop} temperatures.

By contrast, the behavior of the IR-bleached sample is different, and **Fig. 6c** shows a continuous decrease of I_{max} with T_{stop}, in the complete interval T_{stop} = 150–380°C. The difference between the two samples shown in **Fig. 6c** can be attributed to the preheat used before measurement of the CW-IRSL curves (100 s at 50°C). This preheating process can be expected to empty some of the shallower energy levels for T_{stop} values below ~260°C, as seen in **Fig. 6c**.

An alternative method of analyzing the data

In a second attempt to analyze the same TL glow curves and possibly extract physical information about the underlying luminescence process, the shape of TL glow curves is analyzed using a recently proposed physical kinetic model (Jain *et al.*, 2012). This model describes localized electronic recombination in donor-acceptor

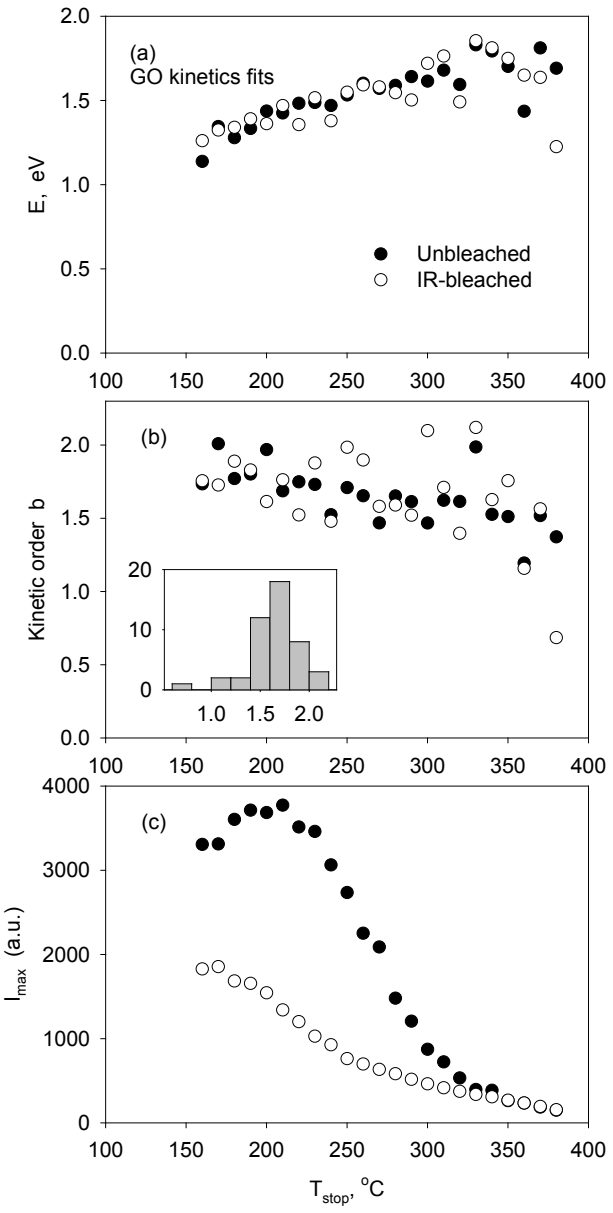

Fig. 6. *Results of fits with GO kinetics (a)* E vs. T_{stop} *(b)* b vs. T_{stop} *and (b)* I_{max} vs. T_{stop}, *for both unbleached and IR-bleached samples.*

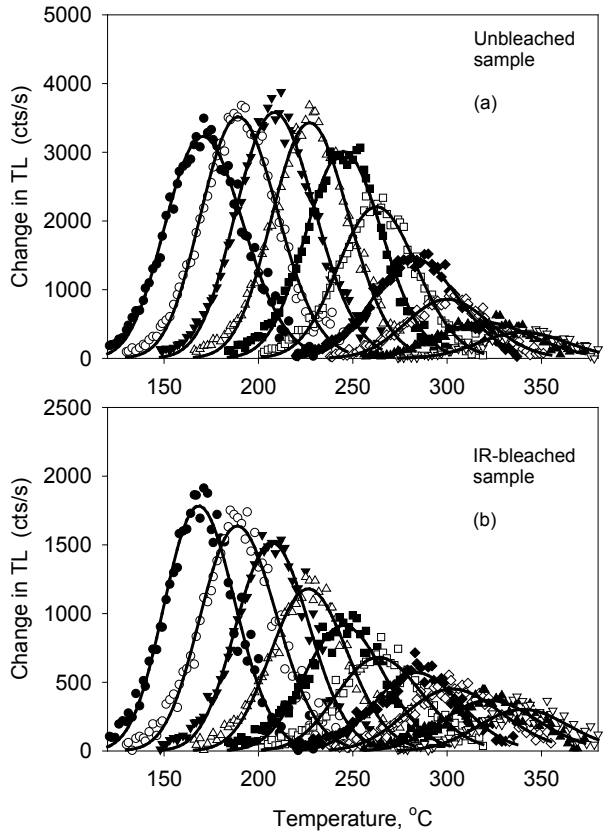

Fig. 7. *Analysis of TL glow curves based on the analytical Eqs. 4.3 and 4.4, for the (a) unbleached and (b) IR-bleached samples. Only every second subtractive result is shown in these figures, for purposes of clarity. The solid lines indicate the best fit using these equations.*

pairs, and is believed to be able to describe luminescence processes in feldspar. Within this model, recombination is assumed to take place via the excited state of the donor, and nearest-neighbor recombinations take place within a random distribution of centers. This recent model has been used successfully to describe several types of luminescence signals (Jain *et al.*, 2012; Kitis and Pagonis, 2013).

In this second type of analysis, it is assumed that there is a continuous distribution of *localized* energy levels E distributed somewhere below the conduction band, and that the difference between two subsequent TL curves can be described with the new equations for TL derived in the paper by Kitis and Pagonis (2013). It is assumed that these energy levels are all independent of each other, and that one can describe the TL glow curves as the sum of independent glow curves with different E-values, different s-values and the same ρ' value. Therefore in this model the TL glow curves are described as the sum of signals from a distribution of localized energy levels, and the assumption is that the conduction band is not involved in this process.

Jain *et al.* (2012) presented two versions of the model which were found to be in good agreement with each other, namely an exact model that evolves both in space and in time, and an approximate semi-analytical model evolving only in time. The model simulated successfully both thermally stimulated luminescence (TL) and optically stimulated luminescence (OSL), and also demonstrated the power law behavior for simulated OSL signals. Kitis and Pagonis (2013) showed that the system of simultaneous differential equations in the semi-analytical model can be approximated by a single differential equation which can be solved analytically. These authors obtained analytical expressions for four different experimental modes of stimulation: TL, OSL, linearly modulated OSL (LM-OSL) and isothermal TL processes. The analytical equations were tested by successfully fitting typical infrared stimulated luminescence (IRSL) and TL signals from feldspar samples, and the dimensionless number density of acceptors in the model was estimated from fitting the experimental data. The expression for TL intensity derived by Kitis and Pagonis (2013) is:

$$L_{TL} = \frac{3n_0 e^{-\rho'[F_{TL}(t)]^3} s\left(E^2 - 6k^2T^2\right) z\rho'\left[F_{TL}(t)\right]^2}{EksT^2 z - 2k^2 sT^3 z + e^{\frac{E}{kT}} E\beta}\beta \qquad (4.3)$$

where

$$F_{TL}(t) = \ln\left(1 + \frac{zskT^2}{\beta E} e^{-\frac{E}{kT}} \times \left(1 - \frac{2kT}{E}\right)\right) \qquad (4.4)$$

The experimental TL curves obtained using the subtractive method of **Fig. 4** were fitted using **Eqs. 4.3** and **4.4**. The fitting parameters in this method are the energy E, proportionality constant n_o, frequency s, and dimensionless concentration ρ'. The least square fitting routines based on the Levenberg-Marquardt algorithm were used in the commercial software *Mathematica*, with the results of the fitting procedure shown as continuous lines in **Fig. 7** for the unbleached and IR-bleached samples.

It is emphasized that the exact luminescence mechanism in this material is unknown, and that the analytical equations derived in the model of Jain *et al.* (2012) are used in this paper on a completely empirical basis, in an effort to describe empirically the shape of the broad TL glow curves in **Figs. 1** and **2**. The analysis described in this section does not imply that the model of Jain *et al.* (2012) is the correct physical picture, but rather that the equations from the model are used here on an empirical basis.

The best fitting parameters are shown as a function of T_{stop} in **Fig. 8**. **Fig. 8a** shows the best fit E values obtained using **Eq. 4.3**, and E can be seen to increase continuously from E ~1.1 eV to E ~1.8 eV for the complete temperature range T_{stop} = 150–380°C and for both the unbleached and IR-bleached samples. **Fig. 8b** shows the best fit values of the dimensionless density

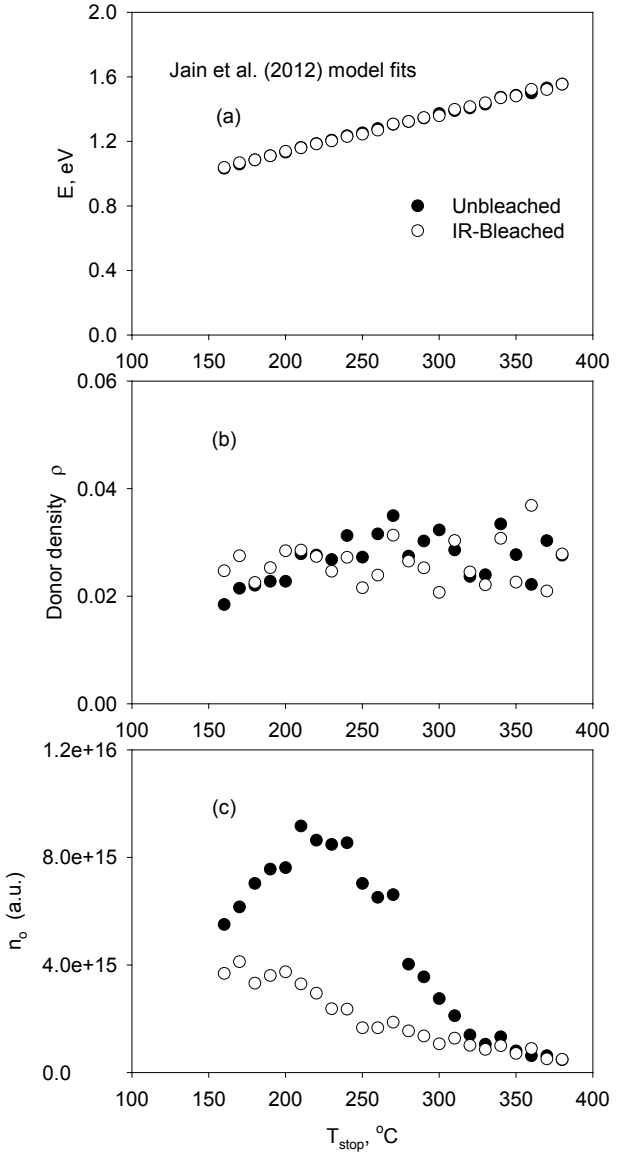

Fig. 8. *Results of fitting the TL data using the analytical Eqs. 4.3 and 4.4, for both unbleached and IR-bleached samples. (a)* E vs. T_{stop} *(b)* b vs. T_{stop} *and (b)* I_{max} vs. T_{stop}.

$\rho' = 0.0266 \pm 0.0004(1\sigma)$. Finally **Fig. 8c** shows the maximum height of the TL glow curves as obtained from the best fit procedure; the data from **Fig. 8c** are seen to be qualitatively similar to the distribution shown in **Fig. 8c** which was obtained using general order kinetics.

5. DISCUSSION

In the previous section it was shown that the T_{max}–T_{stop} method of analysis revealed the existence of an underlying continuous distribution of energy levels. This continuous distribution results in a very broad TL glow curve, a common characteristic for almost all feldspar samples. The broad TL glow curves of **Figs. 1** and **2** were analyzed in this paper by assuming that it can be described as the sum of several independent TL processes, each corresponding to closely located activation energies E. Two different methods were then used to describe the kinetics of these individual TL glow curves, namely the general order kinetics and the localized model of Jain *et al.* (2012). The results of these two methods are shown in **Figs. 6** and **8**.

Both methods of analysis are found to be consistent with a continuous distribution of energies from E ~1.1 eV to E ~1.8 eV for the complete temperature range T_{stop} = 150–380°C. Most notably, this distribution of energies is present for both the unbleached and IR-bleached samples. The shape of the individual TL glow curves can be described accurately by either the almost constant general order parameter $b = 1.65 \pm 0.26(1\sigma)$ shown in **Fig. 6b**, or by the almost constant values of the dimensionless density $\rho' = 0.0266 \pm 0.0004(1\sigma)$ shown in **Fig. 8b**.

The distributions of heights I_{max} obtained from the two methods are shown in **Figs. 6c** and **8c**. In both figures there are some differences in the observed values of I_{max} for the unbleached and IR-bleached samples; these differences could be due to the preheating process used for the IR-bleached sample, as mentioned above. Another possible explanation is that the thermal excitation may be accessing a slightly different set of energy levels than the combined IR-plus-thermal excitation.

The results from both methods of analysis used in this paper are consistent with each other and with the existence of a continuous distribution of energies. It is not possible to choose one method of analysis as preferable to the other, because they both provide a similar description of the experimental TL glow curves, and they both are used here on an empirical basis. Also it is not possible to decide whether the observed distribution of energies corresponds to a distribution in trapping levels or in a distribution of the band-tail states. Even though the role of band-tail states is not clear, there is strong recent experimental evidence pointing to the participation of band-tail states in luminescence production in feldspars (Poolton *et al.*, 2009, Jain and Ankjærgaard, 2011, Morthekai *et al.*, 2012; Pagonis *et al.*, 2012). In the current authors' opinion, the second method based on the analytical **Eq. 4.3** may be preferable, because it has its origin in the physical localized model of Jain *et al.* (2012), while the general order kinetics is a purely empirically based equation with no underlying physical mechanism to support it.

The possible existence of a continuous distribution of shallow states below the conduction band in feldspars, termed the band tail states, was discussed extensively in the papers by Poolton *et al.* (1995, 2002a, 2002b), and direct experimental evidence for their existence was provided later by Poolton *et al.* (2009).

The continuous distribution discussed in this paper may or may not be directly related to the band tail states,

and further work is required to ascertain if this continuous distribution is found in other types of feldspar samples.

The analysis of the distribution of energy levels used in this paper is based on a rather strong assumption, namely that the different energy levels are independent of each other and that no retrapping/redistribution of the electrons takes place between the energy levels. This is a rather strong and arbitrary assumption, which simplifies of course the data analysis. In general one should consider the effects of retrapping and redistribution of charge carriers between different energy levels. In principle, one should attempt to carry out a complete analysis of the assumed distribution of energy levels by using a method similar to the one presented in Hornyak *et al.* (1992). These authors modeled the TL characteristics of the 375°C glow peak in quartz by using a Gaussian distribution of trapping states, two recombination centers and a single active electron trap. Their method consists of solving the system of differential equations for charge carrier concentrations, by subdividing the Gaussian distribution into 96 separate equal narrow energy sections. The electron population in each section was then treated independently, each with its own kinetic parameters. In this paper such a detailed analysis was not attempted, since it would require several additional assumptions for the kinetic parameters in the model.

6. CONCLUSIONS

It is concluded that the broad TL glow curves in feldspar can be analyzed using the T_{max}–T_{stop} method in conjunction with successive subtractions of the overall glow curves. The composition of these narrower constituent glow curves can be analyzed using either the general order kinetics method, or the analytical **Eq. 4.3** derived on the basis of the model of Jain *et al.* (2012). In general, the methods of analysis described in this paper can be a valuable tool for describing the shape and behavior of the broad TL glow curves in feldspars, and for obtaining some useful information about the kinetic parameters describing the underlying luminescence mechanism.

ACKNOWLEDGEMENT

This work was financially, partially, supported by the Department of Science and Technology, India (SR/FTP/ES-56/2011).

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