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Thermoluminescence glow curves in preheated feldspar: A Monte Carlo study

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ABSTRACT

Thermoluminescence (TL) glow curves from feldspars have been the subject of numerous experimental and modeling studies, because of their importance in dosimetry and luminescence dating. Recently there also has been increased interest in using these signals for temperature sensing and for thermochronometry studies. It is now generally accepted that these materials exhibit anomalous fading phenomena due to quantum mechanical tunneling, and that several of their luminescence signals can be described by localized energy transitions taking place in a randomly distributed system of trapped electrons and recombination centers. Our recent modeling work showed that the TL signals of freshly irradiated feldspar samples can also be described from a completely microscopic point of view, by using Monte Carlo methods. This paper extends this recent work, and shows how the Monte Carlo method can also describe TL signals from thermally pretreated feldspars. Specifically, the simulations show that the Monte Carlo method can describe several types of TL experiments for irradiated samples that underwent partial thermal cleaning, and for samples that underwent more complex multistage isothermal procedures. The results from the Monte Carlo simulations are compared quantitatively with experimental data from several types of feldspars, which were preheated at temperatures above 200–300 °C. Common experimental characteristics are pointed out for these preheated feldspars, and the experimental data suggest the possibility of a universal description of the thermal behavior of TL glow curves in feldspars. Specifically, it is found that the shape and width of the experimental TL glow curves do not change significantly for different preheat temperatures, and also do not change when different preheat times are used at a fixed preheat temperature. The relevance of these results for dosimetric and thermochronometry studies is discussed.

1. Introduction

Thermoluminescence (TL) signals from feldspars have been studied extensively both experimentally and by modelling, due to their importance in dosimetry and luminescence dating. During the past few years, there has also been increased interest in using TL glow curves in feldspars as the basis of temperature sensing and in thermochronometry studies (Brown et al. [1]; Yukihara et al. [2]; Biswas et al. [3]; and references therein). These studies have been based on changes taking place on the properties of TL glow curves when samples have been exposed to different thermal or optical treatments, for both naturally and laboratory irradiated samples.

It is now generally accepted that luminescence signals in feldspars originate from a random distribution of trapped electrons and acceptors in these materials, and that the luminescence process involves localized transitions (Jain et al. [4]).

From a modeling point of view, two different approaches have been developed which are based on a *macroscopic* versus a *microscopic* description of the luminescence process.

In the *macroscopic* point of view based on differential equations, it is assumed that the number of trapped electrons is much smaller than the number of acceptors, and therefore the system can be described by a constant number density of acceptors . Based on this assumption and on the quasi-equilibrium conditions, Kitis and Pagonis [5] obtained analytical solutions of the system of differential equations in the model by Jain et al. [4], and these equations have been used to analyze a wide variety of luminescence signals in feldspars and other dosimetric materials ([6–8], and references therein).

In the alternative *microscopic* point of view, a Monte Carlo approach has been used to describe the luminescence process based on quantum tunneling interactions of a small number of defects in a nanometer-sized volume [9–13]. In this second approach, it is not necessary to assume that the number density of acceptors is constant, and the results of the simulations depend on the relative concentrations of electrons

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and acceptors. A significant advantage of the Monte Carlo method is that it can easily be extended to include additional phenomena of interest to the luminescence dosimetry and dating community. Specifically, the Monte Carlo method can also be used to study irradiation phenomena, the presence of multiple types of traps, and retrapping phenomena [10–12].

In a typical TL experiment, one measures two different types of signals. In the first type of measurement, a freshly irradiated sample is immediately heated with a linear heating rate, and one measures the *prompt* TL signal. In the second type of measurement, the freshly irradiated sample undergoes a thermal or optical treatment, before measurement of the *remnant* TL signal.

The recent modeling work by Pagonis and Truong [13] showed that *prompt* TL signals of freshly irradiated feldspar samples can be described from a completely *microscopic* point of view, by using Monte Carlo methods. By using the alternative *macroscopic* differential equation approach, Polymeris et al. [7] showed that it is possible to simulate complex experimental protocols in feldspars, which involve multiple stages required to measure the *remnant* TL signal.

The general purpose of this paper is to extend the work of Polymeris et al. [7] and Pagonis and Truong [13], by using the alternative Monte Carlo method to describe TL glow curves in preheated feldspar samples.

The specific goals of the simulations in this paper are:

- To simulate the well-known fractional glow method of analyzing TL glow curves in feldspars, by using a Monte Carlo technique.
- To investigate by simulation the effect of varying the preheat temperature and preheat time on the TL glow curves, and to compare with experimental data in a quantitative manner.
- To examine in detail the changes occurring in the shape and position of the TL glow curves, and to look for universal behaviors in both the simulated glow curves, and in available experimental data.

2. Experimental

Two sets of experimental data in this paper are analyzed in this paper, and these were previously described by Pagonis et al. [8,14] and by Polymeris et al. [7]. What is new in this paper is the analysis of these previously published data using Monte Carlo methods, based on the microscopic description of tunneling phenomena.

The first set of experimental data shown in Fig. 1a, concerns a museum specimen of feldspar (laboratory code FL3), which was placed in the plagioclase feldspar series by the detailed analysis presented in Morthekai et al. [15]. The analysis included elemental concentrations using ICP-MS measurements and X-ray Diffraction analysis. The 90–150 μm size fraction of the samples were used without any further chemical treatment, and a Risø TL/OSL Reader DA-20 was used for all measurements. 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), transmitting in the wavelength region (395 ± 50) nm. The heating rate was 1.8 °C/s in a nitrogen atmosphere. The TL glow curves of the feldspar samples shown in Fig. 1a 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 *remnant* 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} = 210–400 °C.

Fig. 1b shows the same results as in Fig. 1a, with the TL glow curves shifted along the temperature axis, and scaled to the maximum intensity of the first TL glow curve. After this scaling and temperature-shifting procedure, the twenty remnant TL glow curves coincide with each other within experimental error, indicating that *the preheating procedure did not significantly change the TL glow curve*. This is a rather remarkable experimental result, which to the best of our knowledge has not been pointed out previously for feldspars, and which is discussed further later in this paper.

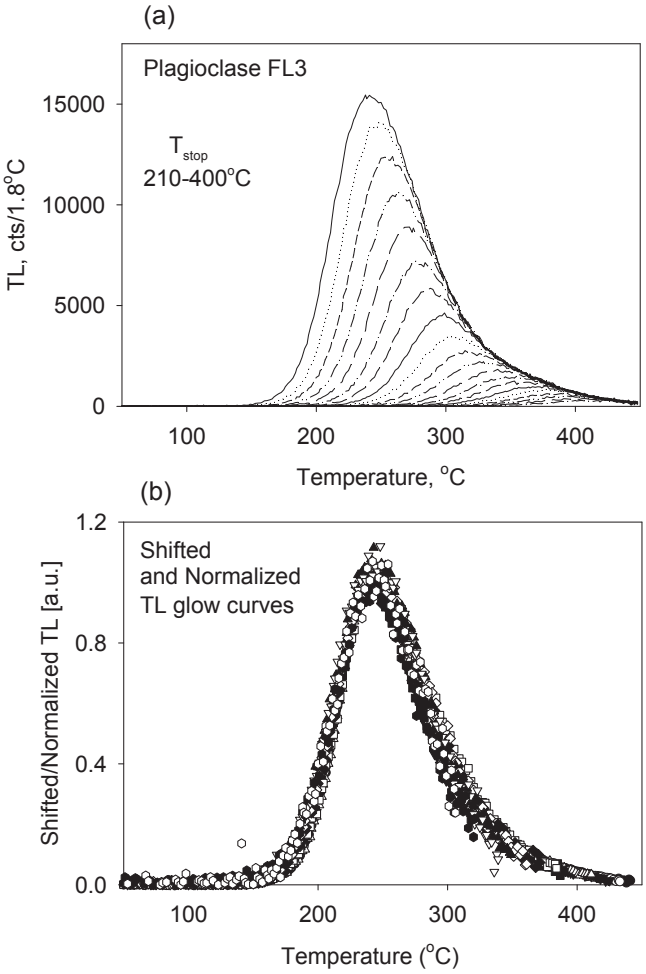

Fig. 1. (a) Remnant TL glow curves of a plagioclase feldspar sample FL3, obtained using a T_{MAX}–T_{STOP} thermal cleaning procedure. This sample was previously studied in Refs. [14,15]. (b) The same data as in (a), after the TL glow curves are shifted along the temperature axis, and scaled to the maximum intensity of the first TL glow curve. This scaling and temperature-shifting procedure shows that the preheating procedure did not significantly change the shape of the TL glow curve.

The second set of experimental data analyzed in his paper concerns a set of K-feldspar samples shown in Fig. 2a and 3a of this paper, and previously studied by Pagonis et al. [8], and Polymeris et al. [7,16]. These previous experimental studies demonstrated the possibility of using TL for structural characterization of ten K-feldspar samples consisting of 3 sanidine, 4 orthoclase and 3 microcline feldspars. A good correlation was shown between TL sensitivity and individual K-feldspar structure, and it was suggested that these samples are ideal for investigating various basic luminescence signals in feldspars. Data from five of the ten samples described in Polymeris et al. [16] are analyzed in this paper, namely microcline samples KST4 and LED1, sanidine sample SAM3, orthoclase sample VRS3 and sanidine sample BAL21. The luminescence measurements were carried out using a Risø TL/OSL reader (model TL/OSL-DA-15), equipped with a $^{90}Sr/^{90}Y$ beta particle source, delivering a nominal dose rate of 0.075 Gy/s. A 9635QA photomultiplier tube was used with a 7.5 mm Hoya U-340 filter (~340 nm, FWHM ~80 nm). A combination of Pilkington HA-3 heat absorbing and Corning 7–59 (320–440 nm) blue filter were used for light detection,

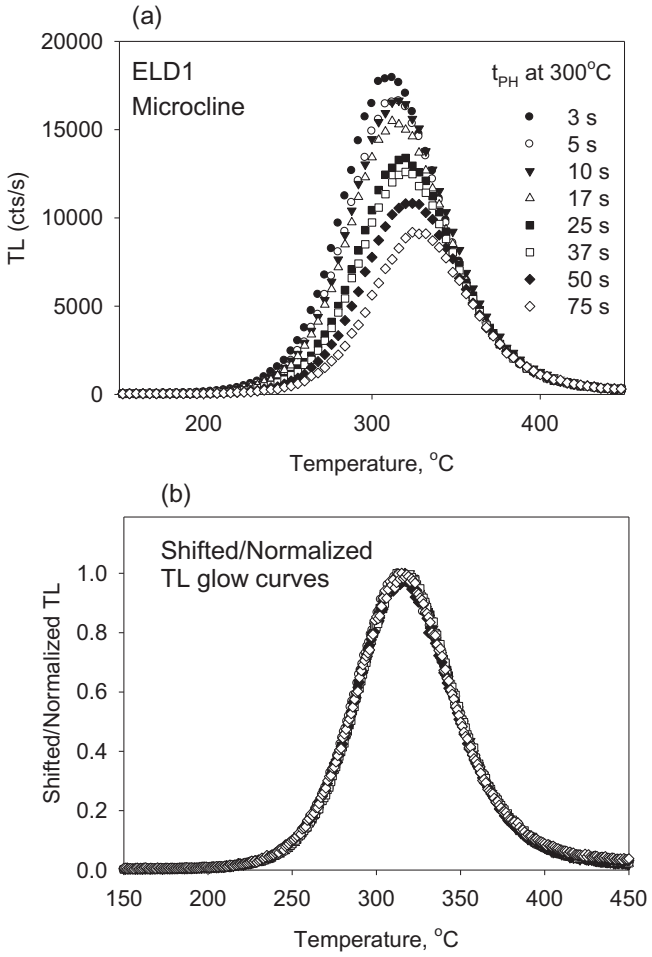

Fig. 2. (a) TL glow curves for microcline sample ELD1, irradiated and subsequently at a temperature T_{PH} = 300 °C for various preheat times t_{PH} = 0–75 s, followed by measurement of its *remnant* TL signal. This sample was previously studied in Ref. [7]. (b) The same data as in (a), shifted along the temperature axis and normalized to their maximum TL intensity, showing that a multistage preheating procedure does not change significantly the shape and width of the TL glow curves.

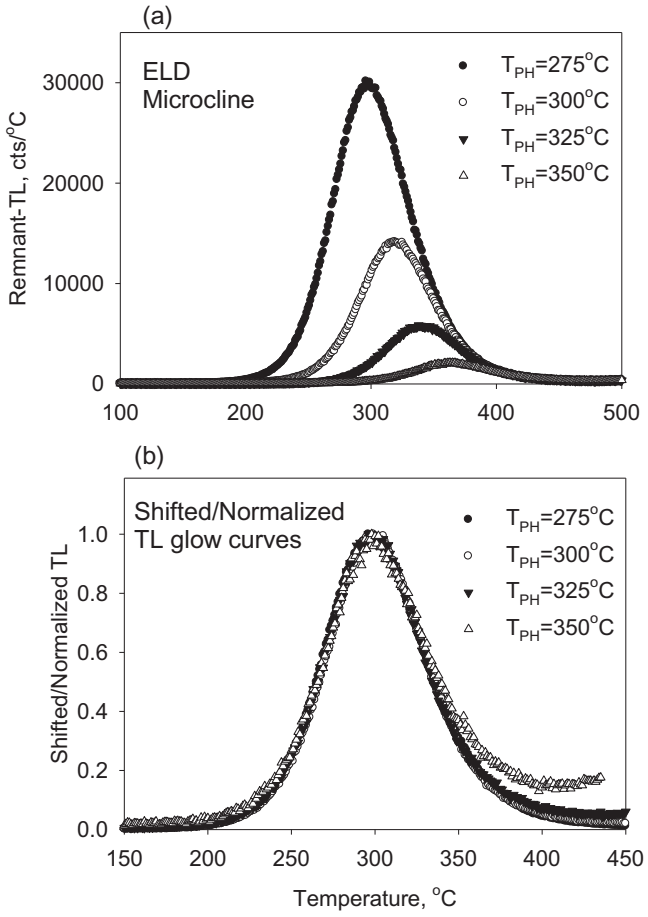

Fig. 3. (a) TL glow curves for microcline sample ELD1, irradiated and subsequently preheated to a variable preheat temperature T_{PH} , then kept at this temperature for a fixed preheating time t_{PH} = 10 s, followed by measurement of its *remnant* TL signal. This sample was previously studied in Ref. [7]. (b) The same data as in (a), shifted along the temperature axis and normalized to their maximum TL intensity.

and all measurements were performed in a nitrogen atmosphere up to the maximum temperature of 500 °C with a low constant heating rate of 1 K/s, in order to avoid significant temperature lag.

In the experimental data of Fig. 2a for microcline sample ELD1, the sample is irradiated with a test dose of 40 Gy and then preheated at a temperature T_{PH} = 300 °C for various preheat times t_{PH} = 0–75 s, and finally heated to 500 °C to measure its remnant TL signal. Similar results were obtained for microcline sample KST4, sanidine SAM3, orthoclase VRS3 and sanidine BAL21, and these were previously analyzed using the differential equation approach in Polymeris et al. [7]. Fig. 2b shows the same data as Fig. 2a, shifted along the temperature axis and normalized to their maximum TL intensity. This figure shows clearly that complex/multistage preheating procedure does not change significantly the shape and width of the TL glow curves.

Fig. 3a shows a third type of experimental data, previously shown in Polymeris et al. [7], for microcline sample ELD1. In the experiments the sample is irradiated with a test dose of 40 Gy and subsequently preheated to a variable preheat temperature T_{PH} = 275, 300, 325 and 350 °C; it is then kept at this temperature for a fixed preheating time t_{PH} = 10 s, followed by measurement of its *remnant* TL signal. Fig. 3b shows the same data as Fig. 3a, shifted along the temperature axis and normalized to their maximum TL intensity. Once more, it is clear that the preheating procedure does not change significantly the shape and width of the TL glow curves.

As in the previous experimental studies by Polymeris et al. [16], a sensitivity test was performed by measuring the TL signals before and after the end of the above protocols. It was found that the sensitivities of all samples showed an excellent stability, and that the reproducibility of the signals was better that 2%. It was concluded that there was no need for applying sensitivity corrections in these experiments.

The unchanging shape of the TL glow curve in preheated feldspar samples shown in Figs. 1b-2b-3b is an important new conclusion from the experimental data, and it has important implications for practical use of feldspars for temperature sensing and thermochronometry. This is further considered later in the DISCUSSION section of this paper.

3. Monte Carlo simulations of remnant TL glow curves in preheated feldspar samples

There are several recent Monte Carlo simulations in the literature, which are based on a random distribution of electrons and positive ions/holes. (Larsen et al. [9], Pagonis and Kulp [10], Pagonis et al. [11–13]). Recently Pagonis and Truong [13] simulated the localized transition model introduced by Jain et al. [4] for TL processes. In this model, one considers three possible transitions, as shown in Fig. 4a. These are thermal excitation of electrons from the ground state into the excited state of the trap (transition D in Fig. 4a), de-excitation from the excited to the ground state (transition T in Fig. 4a), and the

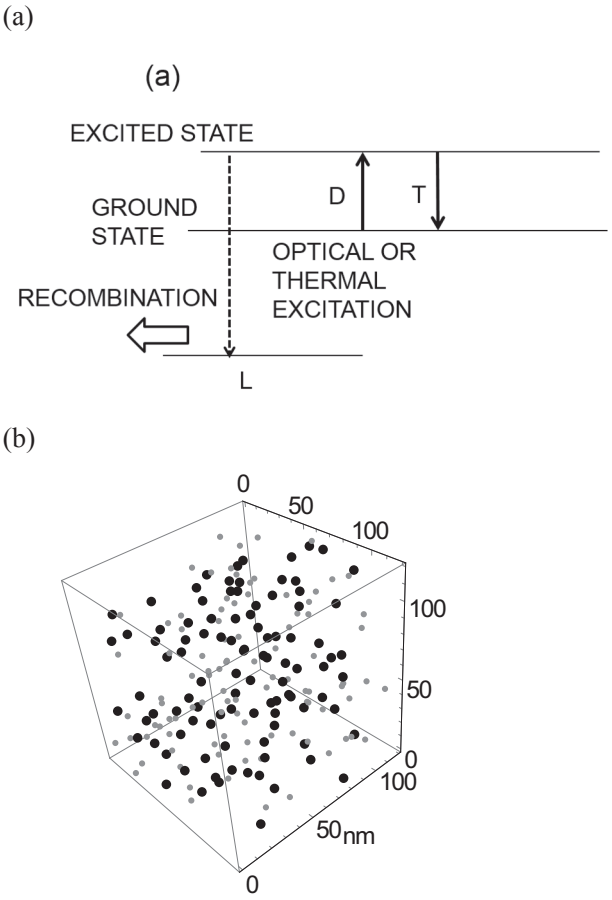

Fig. 4. (a) The model of Jain et al. [4], showing three possible transitions. Thermal or optical excitation of electrons from the ground state into the excited state of the trap (transition D), de-excitation from the excited to the ground state (transition T), and recombination from the excited state of the trapped electron to the recombination center (transition R). (b) The microscopic description of the model in (a), showing a cubic volume containing random initial concentrations of electrons and positive ions. For details of the model see Ref. [13].

recombination process which takes place from the excited state of the trapped electron to the recombination center (transition R in Fig. 4a).

In the Monte Carlo simulations typically one considers a cubic (or spherical) volume of side R as shown in Fig. 4b, containing initial numbers of electrons and positive ions n_o, m_o [10,13]. When the initial number of electrons (n_o) is much smaller than the initial number of positive ions (m_o), the random distribution of charges can be characterized by an almost constant density of positive ions $\rho = m_o/R^3$.

In this paper, we extend the recent work of Pagonis and Truong [13], to include multiple stages involved in a typical measurement of *remnant* TL glow curves in preheated samples. The new simulations involve either a TL process (heating the sample with linear heating rate), or an isothermal process (heating the sample at a constant temperature and for a specific amount of time). As discussed in Ref. [13], in each step of the Monte Carlo simulation, the times t_i of each allowed transition are generated for all carriers in the system, and can be evaluated from the integral Eqt. (Mandowski and Świątek [17], their Eq. (5)):

$$\int_0^{t_i} \lambda(t')dt' = -\ln(a_i) \tag{1}$$

where a_i is a homogeneous normalized random variable from the interval (0, 1) and $\lambda(t)$ (in s^{-1}) is the appropriate transition rate for the process under consideration. In the case of TL experiments Eq. (1) must be solved numerically, as described previously in Refs. [18–20], and recently in Ref. [13].

In a typical TL process the sample is heated with a constant heating rate $\beta(K/s)$ from a starting temperature T_o up to a high temperature around 500 °C, so that the temperature varies with time t as $T(t) = T_o + \beta t$. Jain et al. [21] and previously Thioulouse et al. [22] and Chang and Thioulouse [23], demonstrated that for this type of experiment the transition rate $\lambda(r,t)$ is given by the following Arrhenius type of expression (Jain et al. [21], their Eqs. (3) and (6)):

$$\begin{aligned}\lambda_{TL}(r,\,t) &= s\exp[-r/a]\exp[-E/(k_BT)] \\ &= s\exp[-r/a]\exp[-E/\{k_B(T_o+\beta t)\}]\end{aligned} \tag{2}$$

where a is the tunneling length, E is the thermal activation energy between the ground state and the excited state of the trapped electron, k_B is the Boltzmann constant and s is the frequency factor characterizing tunneling taking place from the excited state of the system.

During the Monte Carlo simulation of the TL process, each of the remaining electrons $n(t)$ in the nanometer-sized volume is examined, and the nearest neighbor distances r_{MIN} are calculated. The recombination times t_i are calculated for each electron in the system by solving numerically the equation (Ref. [13]):

$$s\exp[-r_{MIN}/a]\frac{k_BT^2}{\beta E}\exp[-E/(k_BT)]\left(1-\frac{2k_BT}{E}\right) = -\ln(a_i);\; T = T_o + \beta t_i \tag{3}$$

Only the event corresponding to the shortest of all the possible times t_i in Eq. (3) happens, i.e. the electron-ion pair corresponding to this shortest time is allowed to recombine. Close-by pairs are more likely to recombine first, and further away pairs are likely to recombine later. After this pair is removed from the system in the simulation, the distances between each electron and the positive ions are re-evaluated, and the minimum t_i time is used to update the total time elapsed from the beginning of the simulation. This process is repeated until there are no more electrons left in the system. The program keeps track of the number of recombinations taking place as a function of the elapsed time t, and this number is proportional to the experimentally observed TL intensity.

Some of the simulations in this paper involve an isothermal process, in which one holds the sample at a constant temperature for a specific amount of time. In such a typical *isothermal* process, the sample is heated at a constant temperature T_{PH} for a specific preheat time t_{PH}. In this type of process, the transition rate λ becomes:

$$\lambda_{ISO}(r,\,T_{PH}) = s\exp[-r/a]\exp[-E/(k_BT_{PH})] \tag{4}$$

For such isothermal processes Eq. (1) has a simple analytical solution, since the transition rates $\lambda_{ISO}(r,\,T_{PH})$ do not depend on time. Hence in these isothermal simulations, the recombination times t_i are calculated by modifying Eq. (3) into the following simple analytical equation:

$$s\exp[-r_{MIN}/a]\exp[-E/(k_BT_{PH})]t_i = -\ln(a_i) \tag{5}$$

In these multistage simulations, the final positions of electrons and acceptors at the end of each stage are used as the initial positions of the charges for the next stage in the simulation.

4. Results from the Monte Carlo simulations

4.1. Simulation of the T_{MAX}-T_{STOP} technique when concentration of electrons is much smaller than concentration of holes

In a first test of the Monte Carlo algorithm, we simulate the T_{MAX}-T_{STOP} experimental method, which produces sets of TL glow curves similar to Fig. 1a. In these simulations, the concentration of electrons is taken to be much smaller than the concentration of acceptors. The sample is preheated to a temperature of T_{STOP} = 25–100 °C, followed by measurement of the *remnant* TL signal. Specifically the simulation

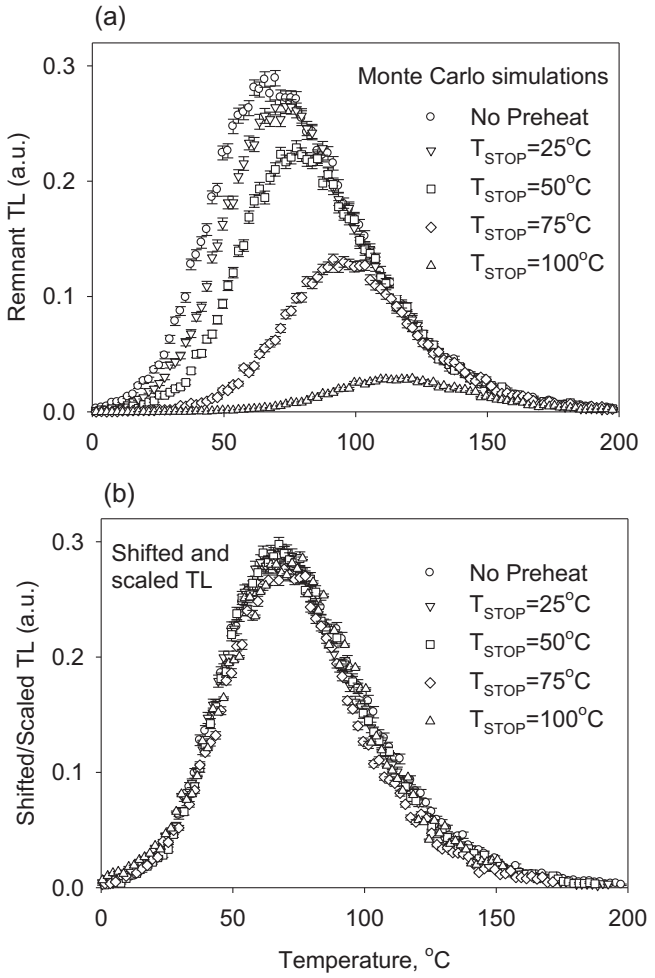

Fig. 5. Monte Carlo simulation of the T_{MAX}-T_{STOP} experimental method, with the sample preheated to a temperature of T_{STOP} = 25–100 °C, followed by measurement of the *remnant* TL signal. The concentration of electrons is taken to be much smaller than the concentration of acceptors, and the parameters of the model are given in the text. (b) The simulation results in (a) with the TL glow curves shifted along the temperature axis, and scaled to their maximum intensity. The scaling and temperature-shifting procedure does not change the shape and width of the remnant TL glow curves.

involves the following distinct stages:

Step 1: The electrons and acceptors are placed at random positions inside the cube.

Step 2: Sample is heated up to a preheat temperature T_{STOP} with a constant heating rate, then immediately cooled to room temperature.

Step 3: The sample is heated from room temperature up to 500 °C with a constant heating rate $\beta = 1 K/s$ in order to measure the remnant TL signal (R-TL). The whole process in steps 1–3 is then repeated for a higher preheat temperature T_{STOP}.

The result of these simulations is shown in Fig. 5a, and is typical of behavior observed in experimental situations with feldspar samples. As the temperature T_{STOP} is increased, the TL glow curves gradually become less intense, and the temperature T_{MAX} corresponding to the maximum TL intensity also increases. Most interestingly from a physical point of view, the preheating process affects only the lower temperature side of the TL glow curve, while the higher temperature side remains unaffected. The physical explanation is that at low T_{STOP} values, only nearby electron-acceptor pairs recombine, while faraway pairs remain unaffected.

The simulated cubic distribution has a radius $R = 71.5\,nm$, tunneling length $a = 0.71$ nm, constant density of positive ions

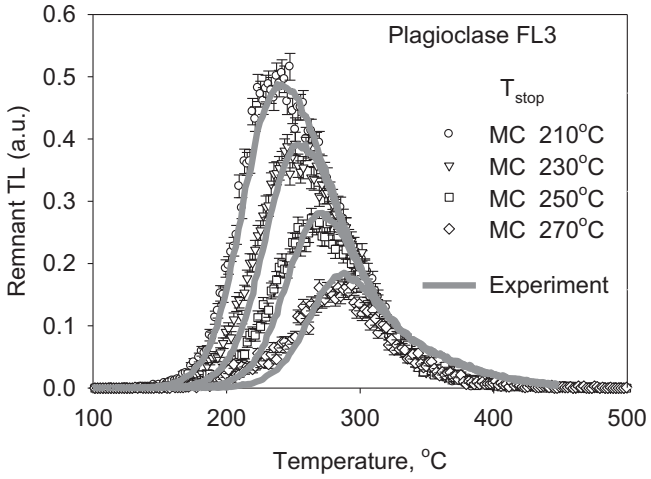

Fig. 6. Monte Carlo simulation of the experimental data in Fig. 1a for sample FL3, with the parameters given in the text. Good quantitative agreement is seen between the Monte Carlo results and the small differences are most likely due to the small number of particles used in the Monte Carlo simulations.

$\rho = 3 \times 10^{24}\,m^{-3}$, activation energy $E = 0.8$ eV, and a tunneling frequency $s = 3 \times 10^{12}\,s^{-1}$. The Monte Carlo data shown in Fig. 5a is the average of 6000 Monte Carlo runs, with each run consisting of an initial number of $n_o = 10$ electrons and $m_o = 1100$ acceptors inside the cube, i.e. the concentration of electrons is 110 times smaller than the concentration of positive ions.

Fig. 5b shows the same simulated results as in Fig. 5a, with the TL glow curves shifted along the temperature axis, and scaled to their maximum intensity. After this scaling and temperature-shifting procedure, the remnant TL glow curves coincide with each other, in agreement with the experimental behavior shown in Fig. 1a for sample FL3.

Fig. 6 shows a direct comparison of some of the experimental data in Fig. 1a with a Monte Carlo (MC) simulation based on the following parameters: cube radius $R = 45.7$ nm, tunneling length $a = 0.5$ nm, constant density of positive ions $\rho = 1.05 \times 10^{25}\,m^{-3}$, activation energy $E = 1.20$ eV, and a tunneling frequency $s = 3.5 \times 10^{12}\,s^{-1}$. The Monte Carlo data shown in Fig. 6 is the average of 1000 Monte Carlo runs, with each run consisting of an initial number of $n_o = 30$ electrons and $m_o = 1000$ acceptors inside the cube. Fig. 6 shows good quantitative agreement between the Monte Carlo results and the experimental data, with the error bars indicating the standard deviation of 1000 Monte Carlo runs. The small differences are most likely due to the small finite number of particles used in the Monte Carlo simulations.

4.2. Simulations of remnant TL glow curve from sample preheated at $T_{PH} = 300$ °C, and for variable time $t_{PH} = 1$–75 s

In a second test of the Monte Carlo algorithm, we simulate the T_{MAX}-T_{STOP} experimental method, which produces sets of TL glow curves similar to Fig. 2a. Specifically the simulation involves the following distinct stages:

Step 1: The electrons and acceptors are placed at random positions inside the cube.

Step 2: The sample is heated from room temperature up to a preheat temperature T_{PH} = 300 °C with a linear heating rate

Step 3: The sample is kept at the preheat temperature T_{PH} = 300 °C for a preheat time t_{PH} = 10 s, then cooled down to room temperature. This is an *isothermal* process, simulated on the basis of Eq. (5).

Step 4: The sample is heated from room temperature up to 500 °C with a linear heating rate in order to measure the remnant TL signal (R-TL). The whole process in steps 1–3 is then repeated for a higher preheat time t_{PH} = 1–75 s.

Fig. 7 shows a direct comparison of some of the experimental data in

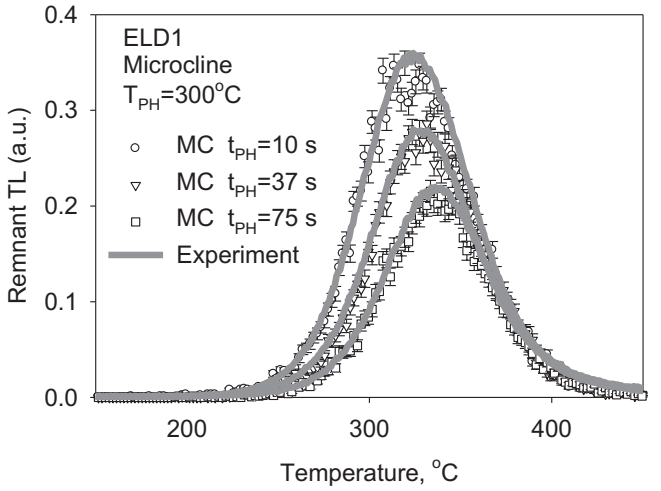

Fig. 7. Monte Carlo simulation of the experimental data in Fig. 2a for microcline sample ELD1 and for a variable preheating time, with the parameters given in the text.

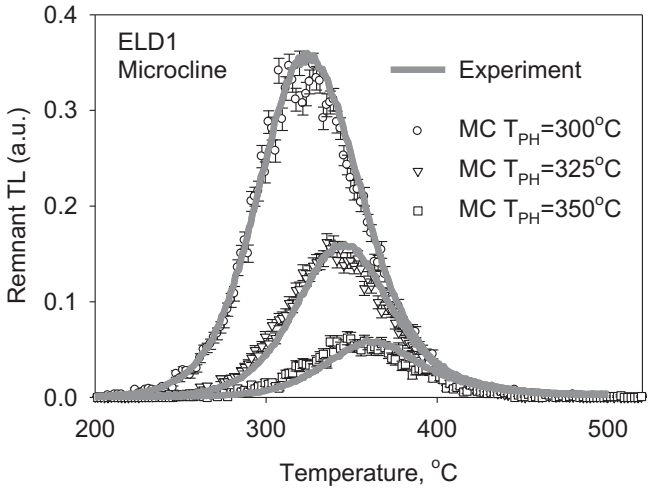

Fig. 8. Monte Carlo simulation of the experimental data in Fig. 3a for microcline sample ELD1 and for a variable preheating temperature. The same model parameters are used in Figs. 7 and 8.

Fig. 2a with a Monte Carlo (MC) simulation based on the following parameters: cube radius $R = 41.2$ nm, tunneling length $a = 0.50$ nm, constant density of positive ions $\rho = 2.85 \times 10^{25}\ m^{-3}$, activation energy $E = 1.45$ eV, and a tunneling frequency $s = 3.5 \times 10^{12}\ s^{-1}$. The Monte Carlo data shown in Fig. 7 is the average of 1500 Monte Carlo runs, with each run consisting of an initial number of $n_o = 30$ electrons and $m_o = 2000$ acceptors inside the cube.

4.3. Simulations of remnant TL glow curve from sample preheated for 10 s at T_{PH} = 300–350 °C

In a third application of the MC algorithm, we simulate the T_{MAX}-T_{STOP} experimental method, which produces sets of TL glow curves similar to Fig. 3a. The simulation involves the following stages:

Step 1: The electrons and acceptors are placed at random positions inside the cube.

Step 2: Sample is heated from room temperature up to a preheat temperature T_{PH} = 300 °C with a linear heating rate.

Step 3: The sample is kept at the preheat temperature T_{PH} = 300 °C for a fixed preheat time t_{PH} = 10 s, then cooled down to room temperature.

Step 4: The sample is heated from room temperature up to 500 °C with a constant heating rate in order to measure the remnant TL signal (R-TL). The whole process in steps 1–3 is then repeated for a higher preheat temperatureT_{PH} = 325 °C, 350 °C.

Fig. 8 shows a direct comparison of some of the experimental data in Fig. 3a with a MC simulation. The model parameters are the same as the ones used for the MC simulations in Section 4.2, and they are the same as the parameters used by Polymeris et al. [7] in their study based on numerical solutions of the differential equations in the model.

4.4. Simulation of the T_{MAX}-T_{STOP} technique for equal initial concentration of trapped electrons and acceptors

One of the powerful features of Monte Carlo algorithms is that they can also be used when no analytical equations are available for the physical situation. We have repeated the simulation of the T_{MAX}-T_{STOP} method in Fig. 5, with the initial concentration of electrons taken to be equal to the concentration of acceptors. No analytical equation is available for this case, and Monte Carlo is currently the only method available for studying such a system.

The results of the simulation are shown in Fig. 9, and the parameters were: cube radius $R = 36.8$ nm, tunneling length $a = 0.5$ nm, constant

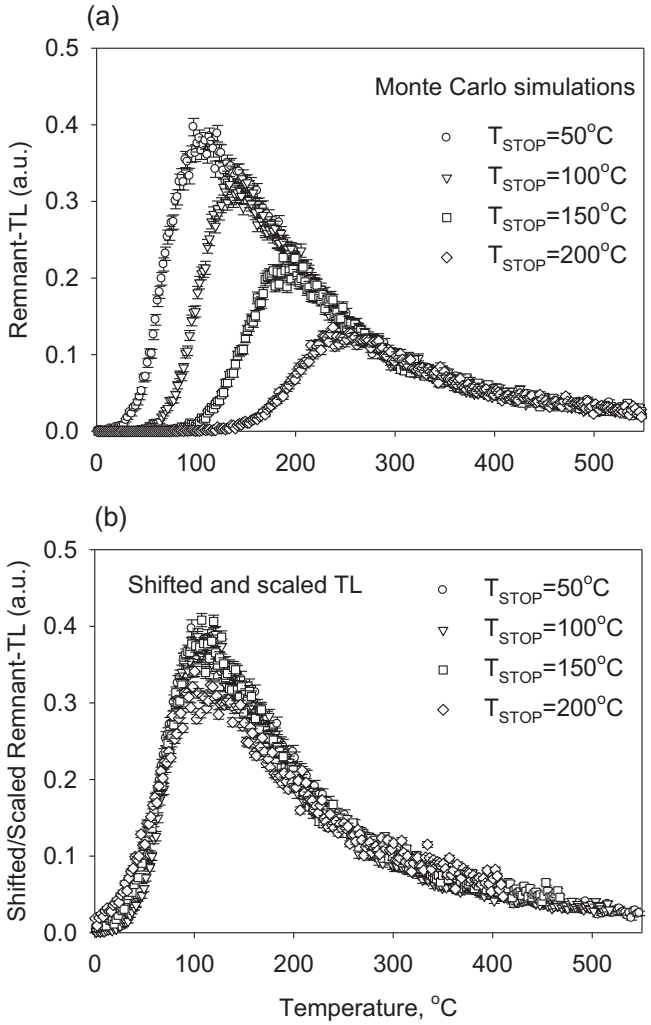

Fig. 9. (a) Monte Carlo simulation of the T_{MAX}-T_{STOP} experimental method, with the initial concentration of electrons taken to be equal the concentration of acceptors. This figure can be compared with the simulations in Fig. 5, in which the initial concentration of electrons is taken to be much smaller than the concentration of acceptors. The parameters of the model are given in the text. (b) The simulation results in (a) with the TL glow curves shifted along the temperature axis, and scaled to their maximum intensity. The scaling and temperature-shifting procedure does not change the shape and width of the remnant TL glow curves.

density of positive ions $\rho = 1.0 \times 10^{24}\,m^{-3}$, activation energy $E = 0.80$ eV, and a tunneling frequency $s = 3 \times 10^{12}\,s^{-1}$. The Monte Carlo data shown in Fig. 6 is the average of 1000 Monte Carlo runs, with each run consisting of an initial number of $n_o = 50$ electrons and $m_o = 50$ acceptors inside the cube. By comparing the shape of the TL glow curves in Figs. 5 and 9, it is clear that the tunneling process in Fig. 9a is slower, resulting in overall broader TL glow curves. The physical explanation for this slower tunneling process is that in the case of equal initial concentrations of electrons and acceptors, each electron can access acceptors that are more distant within the nanometer sized cube.

Once more, Fig. 9b shows the same simulation results as in Fig. 9a, with the shifted and normalized TL glow curves from Fig. 9a, verifying that preheating the samples did not change significantly the shape of the TL glow curve.

5. Discussion and conclusions

The experimental results shown in this paper and the Monte Carlo simulations lead to the same general conclusion: preheating of feldspar samples for various preheat temperatures T_{PH} above 300 °C and for various preheat times t_{PH}, does not change significantly the shape of the remnant TL glow curves.

Fig. 10ab shows additional experimental data on the full width at half maximum (FWHM) for five of the 10 samples studied by Polymeris et al. [16]. Specifically the experiments in Figs. 2 and 3 were repeated for microcline samples KST4 and LED1, sanidine sample SAM3, orthoclase sample VRS3 and sanidine sample BAL21. These experiments produced a total of N = 66 remnant TL glow curves. Fig. 10a shows the distribution of FWHM for the variable preheat time experiments, and Fig. 10b shows the corresponding distributions for the variable preheat temperature experiment. The two distributions are almost identical, with the average FWHM = (77 ± 8) K (1σ, N = 41), and FWHM = (7 ± 8) K (1 σ, N = 25) correspondingly, verifying that the multistage preheating procedures do not alter the shape and width of the TL glow curves. In addition, the range of the distributions of FWHM in Fig. 10ab is rather narrow, despite the very different types of feldspars being studied.

These results are important for thermochronometry studies using feldspars, since they point to the possibility of a universal behavior by preheated feldspar samples. The unchanging shape of the TL glow curve means that it is possible to carry out detailed and reliable temperature sensing experiments on the scale of seconds, and possibly to carry out thermochronometry studies on much larger geological scales (see for example Brown et al. [1]; Yukihara et al. [2]; Biswas et al. [3]). Since the shape of the TL glow curves does not change, one can use the position of maximum TL intensity T_{MAX} as an accurate indicator of the thermal history of the samples. Alternatively, one can also use the temperature $T_{1/2}$ of half-maximum TL intensity, as was done in the detailed thermochronometry work by Brown et al. [1].

Additional current work in progress has provided a theoretical explanation for the unchanging shape of the TL glow curves, based on the differential equations approach, and will be presented elsewhere.

The results of the MC method presented here are consistent and in complete quantitative agreement with the mathematically more complex approaches, which use differential equations (see for example Polymeris et al. [7]).

The MC method was shown here to be an attractive alternative, since it does not require numerical integrations, and it can simulate a larger range of physical phenomena by varying the model parameters in the microscopic point of view. Finally, the Monte Carlo method can also be used when no analytical equations are available to describe the physical system.

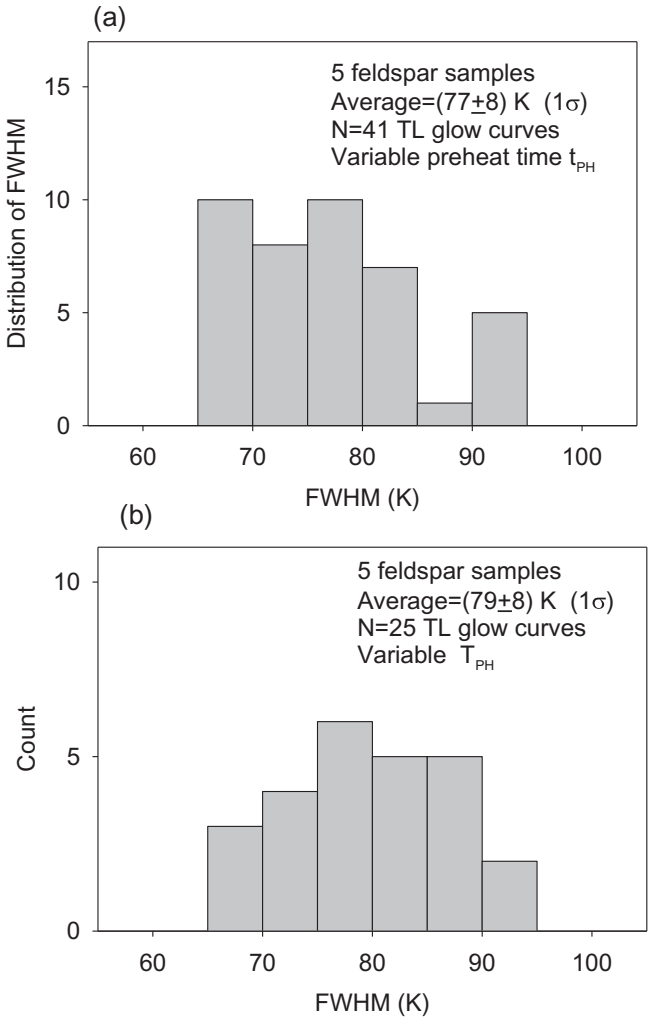

Fig. 10. Results of analysis of N = 66 remnant TL glow curves obtained by repeating the experiments in Figs. 2 and 3 for five feldspar samples. (a) Distribution of the FWHM obtained with a variable preheat temperature and (b) The corresponding FWHM distributions obtained with a variable preheating time.

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