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Modelling thermal transfer in optically stimulated luminescence of quartz

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Abstract
A previously published kinetic model for the production of luminescence signals in quartz is used to investigate the production of thermally transferred optically stimulated luminescence (TT-OSL) signals. This paper provides a mathematical description of the thermal transfer mechanism for two different phenomena that have been observed in previously published experiments (Aitken and Smith 1988 *Quat. Sci. Rev.* **7** 387–93). The starting point is the model proposed by Bailey (2001 *Radiat. Meas.* **33** 17–45). The numerical values of some of the parameters are varied so that they match the experimental data. The effect caused by varying these values is investigated.

The first of these phenomena takes place after storing optically bleached samples at room temperature; this involves the traps responsible for the 110 °C thermoluminescence (TL) peak of quartz acting as a refuge trap. The second phenomenon concerns OSL signals that are induced by heating the samples after the bleaching of the OSL signal and involves a putative TL peak at ∼230 °C associated with the refuge trap; specifically, the paper presents a simulation of the temperature dependence of the OSL signal measured by successively heating the quartz samples to higher temperatures up to ∼400 °C.

1. Introduction

The accurate dating of sedimentary quartz using the optically stimulated luminescence (OSL) signal depends upon the ability to use this light-sensitive signal to determine the radiation dose that the grains have received since their last exposure to sunlight before incorporation in the sediment (Huntley *et al* 1985, Aitken 1998). Since the OSL signal is derived from light-sensitive electron traps, it is assumed that quartz grains that have been exposed to light, either in the natural environment or in the laboratory, and not exposed to any subsequent irradiation would give rise to zero OSL signal, and thus to a zero OSL age. However, when a thermal treatment is introduced into the dating procedure after laboratory bleaching, a non-zero OSL signal may be observed. The process that gives rise to this secondary signal is known as thermal transfer. The thermal transfer of electrons from light-insensitive to light-sensitive traps (i.e. those that would give rise to an OSL signal after natural or laboratory bleaching) was mentioned in the first publication on dating using the OSL signal (Huntley *et al* 1985).

For a sand of the Jurassic age that had been exposed to light, Smith *et al* (1986) found that when the OSL signal was measured at room temperature, an increase in the OSL signal was observed both after storage at room temperature in the dark or after heating to successively higher temperatures. For storage at room temperature, the traps related to the thermoluminescence (TL) at 110 °C play a role in the thermal transfer process (e.g. Kaylor *et al* 1995,

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Murray 1996, Alexander *et al* 1997, Bailey 1997 and Wintle and Murray 1997). Aitken and Smith (1988) reported more detailed studies of the dependence of the thermally transferred OSL intensity on both the time of storage, and the temperature reached, in two sets of experiments on a Moroccan dune sand. For these raised temperature experiments, they inferred that a deeper, more thermally stable, trap played a role in the thermal transfer process. In this paper, the experimental data for these two experiments are compared with the computer-generated output of a mathematical model based on the kinetic model proposed by Bailey (2001).

In the first dating studies, OSL measurements were performed with the sample held at room temperature during the optical stimulation. Under such conditions, Smith and Rhodes (1994) showed phototransfer of electrons from the OSL trap into the traps of the 110 °C TL peak. In addition, Wintle and Murray (1997) and Bailey (1997) showed that the traps giving rise to the 110 °C TL peak were optically sensitive; thus, Wintle and Murray (1997) recommended that OSL measurements were to be made with the sample held at 125 °C, thus keeping the 110 °C TL peak traps empty during optical stimulation. Measurement of the OSL signal at 125 °C has now been widely adopted in the dating of quartz using single aliquots (Murray and Wintle 2000, Wintle and Murray 2006).

Using the concept of fast, medium and slow OSL components proposed by Bailey *et al* (1997), Aitken (1998) considered the TT-OSL to come from the fast component trap. According to Aitken (1998), the recuperated OSL phenomenon is a double transfer effect, involving both a phototransfer of electrons during the bleaching of the OSL signal related to the fast component, and a thermal transfer of a proportion of these electrons during subsequent heating back into the fast OSL component traps. Rhodes (2000) also interpreted the TT-OSL as coming from the fast component trap. However, Jain *et al* (2003) concluded from their studies of the linearly-modulated OSL signals from quartz that there was negligible recuperation of the fast component, but significant recuperation of the medium and slow components.

Thus, there is much to learn about TT-OSL signals in quartz. The aim of running the Bailey (2001) computer model and comparing the output with published experimental data was to throw light on the mechanisms behind its production.

2. The model

During the past five years significant advances have been made in the mathematical modelling of TL and OSL phenomena in quartz (Bailey 2001, 2004, Bøtter-Jensen *et al* 2003, Adamiec *et al* 2004, 2006). Such advances include the development of comprehensive theoretical models that attempt to simultaneously describe TL and OSL production. The mathematical model used here is the original comprehensive kinetic model of Bailey (2001). The model consists of five electron traps and four hole centres.

For the sake of easy reference, we also provide a brief description of the various energy levels in the model. The electron traps are listed as levels 1 to 5. Level 1 is the 110 °C TL level, which is a relatively shallow electron trapping state, giving rise to a TL peak at ∼110 °C when measured at a heating rate of 5 °C s^{-1}. According to Wintle and Murray (1997), these traps are optically sensitive. Level 2 is a ‘medium stability’ electron trapping state, yielding a simulated TL peak at ∼230 °C at a heating rate of 5 °C s^{-1}. In the double transfer mechanism (Aitken 1998), level 2 acts as the ‘refuge trap’ into which the electrons are transferred by optical stimulation. Levels 3 and 4 represent the fast and medium OSL components (Bailey *et al* 1997) and yield TL peaks at ∼330 °C, as well as being possible sources of the TT-OSL electrons. In the present simulations and in the experiments discussed in this paper, only the initial few seconds of the OSL signal are used as a measure of the OSL intensity. This initial OSL signal is almost entirely due to the fast component of OSL in quartz, which is typically two orders of magnitude larger than the medium and slow OSL components in the initial part of the OSL decay curves measured under continuous stimulation. Level 5 represents a deep thermally disconnected and very stable electron centre.

The Bailey (2001) model has four hole trapping centres (levels 6–9), with transitions of electrons from the conduction band being allowed for, as well as additional transitions of holes from the valence band into these levels. Levels 6 and 7 are thermally unstable non-radiative recombination centres similar to the hole reservoir ‘R-centres’ first introduced by Zimmerman (1971). These levels are known to be important in explaining the thermal activation characteristics of quartz (Chen and Li 2000, Chen *et al* 2000, Li and Chen 2001). Level 8 is a thermally stable radiative recombination centre (‘L-centre’), in which electrons recombine with trapped holes to produce the OSL signal. Level 9 is a thermally stable non-radiative recombination centre, representing all other recombination centres in quartz.

In terms of OSL traps, the model used in this paper contains traps for the fast and medium OSL components ($\mathrm{OSL_F}$ and $\mathrm{OSL_M}$, levels 3 and 4 in the Bailey (2001) model), but does not contain traps that describe the slow OSL components. Such additional levels were introduced later in a more complex version of the model (Bailey 2004). The absence of these additional energy levels does not limit the applicability of the present model, since the phenomena described here involve exclusively the fast OSL component.

The computer code is written in *Mathematica* and was tested for consistency by successfully reproducing several of the simulation results in Bailey (2001). The parameters are as defined by Bailey (2001); N_i are the concentrations of electron traps or hole centres (cm^{-3}), n_i are the concentrations of trapped electrons or holes (cm^{-3}), s_i are the frequency factors (s^{-1}), E_i are the electron trap depths below the conduction band or hole trap depths above the valence band (eV), A_i are the valence band to trap transition probability coefficients ($\mathrm{cm}^3\ \mathrm{s}^{-1}$) and B_i are the conduction band to hole trap transition probability coefficients ($\mathrm{cm}^3\ \mathrm{s}^{-1}$). Other parameters related to the optically sensitive traps are the photo-eviction constant θ_{01} (s^{-1}) at $T = \infty$, the thermal assistance energy E_i^{th} (eV) and P representing the stimulation photon flux.

Table 1. The Qtz-A$_1$ parameters of Bailey (2001) are shown together with their modified values used in the simulation shown in figure 2. The modified values are shown in bold.

Levels		Parameters N_i (cm^{-3})		E_i (eV)		s_i (s^{-1})		A_i (cm^3 s^{-1})		B_i (cm^3 s^{-1})	θ_{0i} (s^{-1})	E_i^{th} (eV)
1	(110 °C TL)	1.5e7	**5.1e9**	0.97		5e12		1e − 8		—	0.75	0.1
2	(230 °C TL)	1e7		1.55		5e14		1e − 8	**1.0e−9**	—	—	—
3	(OSL$_\mathrm{F}$)	1e9	**1e11**	1.7	**1.73**	5e13	**36e13**	1e − 9	**0.5e−9**	—	6	0.1
4	(OSL$_\mathrm{M}$)	2.5e8		1.72	**1.8**	5e14	**1.5e13**	5e − 10		—	4.5	0.13
5	(Deep)	5e10		2		1e10		1e − 10		—	—	—
6	(R$_1$-centre)	3e8		1.43		5e13		5e − 7		5e − 9	—	—
7	(R$_2$-centre)	1e10		1.75		5e14		1e − 9		5e − 10	—	—
8	(L-centre)	1e11	**1e8**	5		1e13		1e − 9		1e − 10	—	—
9	(K-centre)	5e9		5		1e13		1e − 10		1e − 10	—	—

Table 2. The Qtz-A_1 parameters of Bailey (2001) are shown together with their modified values used in the simulation shown in figure 6. The modified values are shown in bold.

Levels		Parameters N_i (cm^{-3})		E_i (eV)		s_i (s^{-1})		A_i (cm^3 s^{-1})		B_i (cm^3 s^{-1})	θ_{0i} (s^{-1})	E_i^{th} (eV)
1	(110 °C TL)	1.5e7	**1.25e9**	0.97		5e12	**1e14**	1e − 8		—	0.75	0.1
2	(230 °C TL)	1e7	**1e8**	1.55		5e14		1e − 8	**1.0e−9**	—	—	—
3	(OSL$_\mathrm{F}$)	1e9	**5e10**	1.7	**1.73**	5e13	**36e13**	1e − 9	**0.5e−9**	—	6	0.1
4	(OSL$_\mathrm{M}$)	2.5e8		1.72	**1.8**	5e14	**1.5e13**	5e − 10		—	4.5	0.13
5	(Deep)	5e10		2		1e10		1e − 10		—	—	—
6	(R$_1$-centre)	3e8		1.43		5e13		5e − 7		5e − 9	—	—
7	(R$_2$-centre)	1e10		1.75		5e14		1e − 9		5e − 10	—	—
8	(L-centre)	1e11	**1e8**	5		1e13		1e − 9		1e − 10	—	—
9	(K-centre)	5e9	**5e10**	5		1e13		1e − 10		1e − 10	—	—

Table 3. Comparison of the simulated and experimental results obtained by Bailey (2001) with the corresponding results obtained using the set of kinetic parameters in tables 1 and 2.

Simulated quantity		Using original parameters in Bailey (2001)	Using parameters in table 1	Using parameters in table 2	Experimental values from Bailey (2001)
PTTL/OSL ratio		0.041	0.44	0.17	0.041 ± 0.008
Optical desensitization	$\chi_\mathrm{I}/\chi_\mathrm{o}$ %	4.94	3.0	1.5	4.23 ± 2.70
Dose quenching	$\chi_\mathrm{q}/\chi_\mathrm{o}$ %	23.0	8.0	11.0	20.46 ± 3.47
Optical sensitization	$\chi_\mathrm{R}/\chi_\mathrm{q}$ %	0.33	0.4	0.4	1.33 ± 0.59

The equations used in this study are as follows:

$$\frac{\mathrm{d}n_i}{\mathrm{d}t} = n_\mathrm{c}(N_i - n_i)A_i - n_i P\theta_{oi}\mathrm{e}^{(-E_i^{\mathrm{th}}/k_\mathrm{B}T)} - n_i s_i \mathrm{e}^{(-E_i/k_\mathrm{B}T)}, \quad (i = 1, \ldots, 5), \tag{1}$$

$$\frac{\mathrm{d}n_j}{\mathrm{d}t} = n_\mathrm{v}(N_j - n_j)A_j - n_j s_j \mathrm{e}^{(-E_j/k_\mathrm{B}T)} - n_\mathrm{c} n_j B_j, \quad (j = 6, \ldots, 9), \tag{2}$$

$$\frac{\mathrm{d}n_\mathrm{c}}{\mathrm{d}t} = R - \sum_{i=1}^{5}\left(\frac{\mathrm{d}n_i}{\mathrm{d}t}\right) - \sum_{j=6}^{9}(n_\mathrm{c} n_j B_j), \tag{3}$$

$$\frac{\mathrm{d}n_\mathrm{v}}{\mathrm{d}t} = \frac{\mathrm{d}n_\mathrm{c}}{\mathrm{d}t} + \sum_{i=1}^{5}\left(\frac{\mathrm{d}n_i}{\mathrm{d}t}\right) - \sum_{j=6}^{9}\left(\frac{\mathrm{d}n_j}{\mathrm{d}t}\right). \tag{4}$$

The luminescence is defined as

$$L = n_\mathrm{c} n_8 B_8 \eta(T) \tag{5}$$

with R denoting the pair production rate and η representing the luminescence efficiency, given by

$$\eta(T) = \frac{1}{1 + K\mathrm{e}^{-W/kT}}. \tag{6}$$

Here $K = 2.8 \times 10^7$ is a dimensionless constant and $W = 0.64$ eV is the activation energy (Bailey 2001, p 23)

The initial numerical values for the parameters in the model were those chosen by Bailey (2001) on the basis of experimental data from a number of samples; they are given in normal typeface in tables 1, 2 and 3. Some of these values needed to be modified in order to provide a match with the experimental data that form the basis of the three simulations listed in this section. The modified values, obtained by a systematic variation in the parameters in the model, are shown in bold typeface in tables 1, 2 and 3.

In a recent publication, Singarayer and Bailey (2003) obtained slightly different values of the kinetic parameters E and s for the fast and medium OSL components, namely they obtained $E_3 = 1.74$ eV, $s_3 = 8.9 \times 10^{13}$ s^{-1}, $E_4 = 1.8$ eV, $s_4 = 1.5 \times 10^{13}$ s^{-1} in their study of a Sri Lankan dune deposit. We have used these values, with small modifications where necessary, in all simulations presented in this paper. Specifically a value of $E_3 = 1.73$ eV instead of $E_3 = 1.74$ eV was used in all simulations. In addition, a value of $s_3 = 3.6 \times 10^{14}$ s^{-1} was used instead of $s_3 = 8.9 \times 10^{13}$ s^{-1} in simulations #1 and #2. The values of E_4 and s_4 used in all three simulations were the same as the original Singarayer and Bailey (2003) values.

Charge neutrality was checked through the simulation in order to check the accuracy of the calculations and of the numerical integrations. The observed deviation from neutrality was less than 1 part in 10^{11} charge units, which is comparable to the charge accuracy in Bailey (2001).

2.1. Simulation #1: recuperated OSL at room temperature

This simulation describes the experimental behaviour of sample A described by Aitken and Smith (1988), a Moroccan dune sand with an equivalent dose of 14 Gy that was not given any additional irradiation or heat treatments. Aitken and Smith (1988) used the 514 nm line from an argon ion laser to bleach their sample and to make the OSL measurements with the sample held at a temperature of 17 °C. They found that, if the sample was left for a period of time after the end of the bleaching, the subsequent OSL was higher than it was immediately after bleaching had finished. This was referred to as the recuperation effect by Aitken and Smith (1988).

The mechanism proposed to give rise to the recuperated OSL involves several charge transfer steps. During bleaching of the OSL signal in the laboratory, electrons are released from the main OSL traps (levels 3 and 4 in the Bailey model) into the conduction band. Some of these electrons are captured in competing electron traps, such as the 110 °C TL traps (level 1). During storage of the quartz sample at room temperature for long periods of time, some of the electrons captured at the 110 °C TL trap (level 1) are gradually thermally released into the conduction band and may be retrapped into the main OSL traps (levels 3 and 4). Measurement of the OSL signal after a few days using short optical stimulations results in an increased recuperated OSL signal due to the increased concentration of trapped electrons in the main OSL traps (levels 3 and 4).

Figure 1 is an outline of the computer simulation of the thermally transferred OSL phenomenon that is observed at 20 °C. The first five steps are a simulation of the ‘natural’ quartz sample as suggested by Bailey (2001); the remaining steps are a simulation of the experimental procedure of Aitken and Smith (1988). The simulation contains additional stages not shown in figure 1, namely 60 s intervals of relaxation after each irradiation stage as well as appropriate cooling-down periods after each heating stage in the simulation. We have repeated the calculations with a relaxation time of 1 s and the results remained the same. The relaxation periods are necessary to allow the concentrations of electrons and holes in the conduction and valence band to drop to zero, before proceeding to subsequent stages in the simulation.

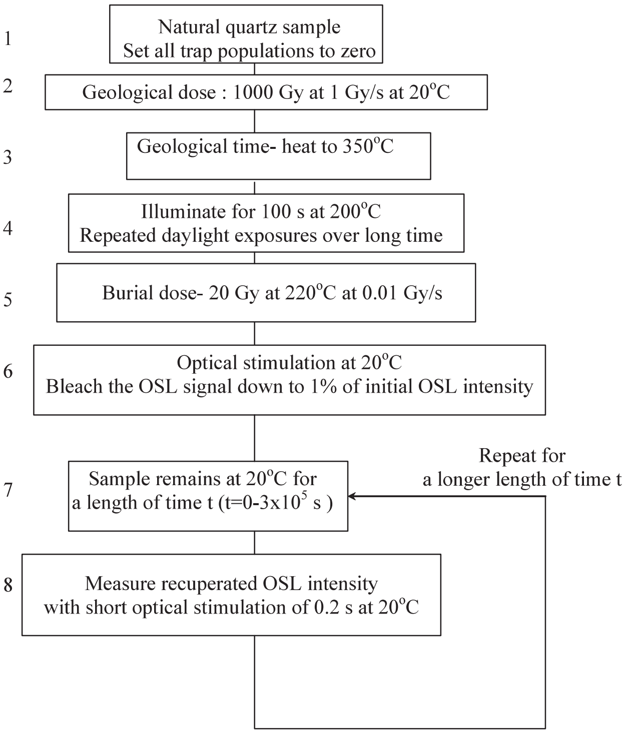

Figure 1. Outline of the experimental procedure of Aitken and Smith (1988), which is simulated using the Bailey model (2001) and with the set of modified parameters shown in table 1. The first five steps are proposed by Bailey (2001).

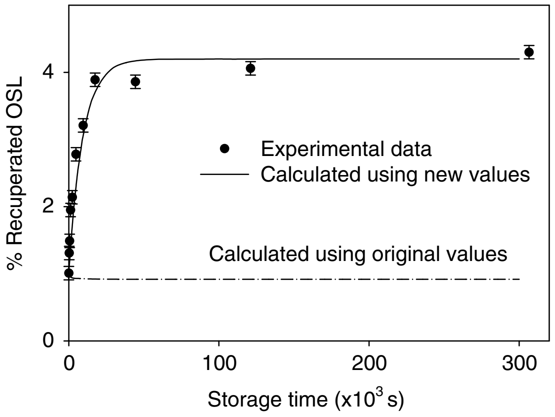

Figure 2. The results of simulating steps 6, 7 and 8 of the procedure shown in figure 1 and using the selected parameters (table 1). The per cent recuperated OSL signal is shown as a function of the elapsed time *t* after the end of the bleaching process (i.e. step 7 in figure 1). The dashed line shows the result of the same calculation made using the original Bailey (2001) parameters (also given in table 1). The experimental data from figure 1 of Aitken and Smith (1988) are redrawn for direct comparison with the model. The experiment runs over the equivalent of five days.

Aitken and Smith (1988) measured the recuperated OSL signal as a function of storage time over three days and their data are shown in figure 2. They found that the OSL signal increased with storage time from an initial value of 1% of the OSL signal before bleaching, up to a value between 4% and 5% after several days of storage time at room temperature. The results of the initial simulation of the steps in figure 1 using the original parameters in Bailey (2001) are shown as a dashed line in figure 2 and did not match the trend of the experimental data. This output implied that the recuperated OSL signal would have been fixed at a value that was 1% of the OSL signal before bleaching. Changing the values of E and s for levels 3 and 4 to those of Singarayer and Bailey (2003), as discussed in the previous section, made negligible difference to this output. However, a good fit was obtained using the numerical values shown in bold in table 1 and the output is given as the solid line in figure 2. Altogether, it was necessary to change only five parameters (N_1, A_2, N_3, A_3 and N_8) of the

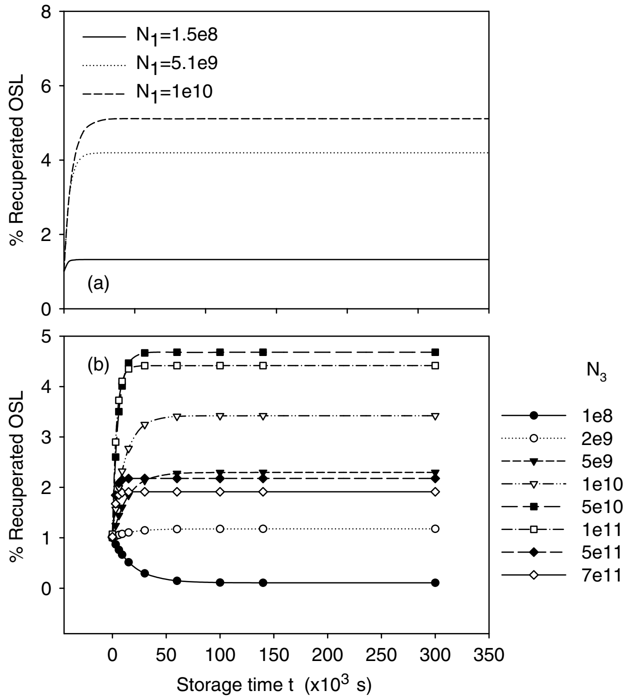

Figure 3. Numerically simulated graphs obtained by varying concentration parameters (*a*) N_1 (the 110 °C trap) and (*b*) N_3 (the fast OSL component).

original total of 43, in order to get a reasonably good fit to the experimental data.

The value of N_1 was increased by over two orders of magnitude (from 1.5×10^7 to $5.1 \times 10^9\,\mathrm{cm^{-3}}$) in order to increase the probability of thermal transfer from the OSL traps (level 3) into the 110 °C TL trap (level 1) during the bleaching process. The value of A_2 (probability coefficient of electron capture in competing level 2) was decreased by an order of magnitude (from 1×10^{-8} to $1.0 \times 10^{-9}\,\mathrm{cm^3\,s^{-1}}$) in order to decrease the competition for electrons from the 230 °C TL trap (level 2). The value of N_3 was increased by two orders of magnitude (from 1×10^9 to $1 \times 10^{11}\,\mathrm{cm^{-3}}$) in order to increase the probability of electron capture into the OSL traps (level 3), as compared with the competition from the 230 °C TL trap (level 2). Similarly, the value of N_8 was decreased by three orders of magnitude (from 1×10^{11} to $1 \times 10^8\,\mathrm{cm^{-3}}$) in order to have comparable probabilities of electron capture into levels 1, 3 and 8.

No changes were necessary for the original Bailey (2001) values of $E_1 = 0.97\,\mathrm{eV}$ and $s_1 = 5 \times 10^{12}\,\mathrm{s^{-1}}$. This value of E_1 is well within the range of $0.86 \pm 0.12\,\mathrm{eV}$ cited by Pagonis *et al* (2002). The value is slightly lower than the model value of 0.99 eV obtained by Adamiec *et al* (2004) who used a genetic algorithm along with a model that had five electron traps and five hole traps; the extra trap corresponded to a slow OSL component, used in addition to the levels in the Bailey (2001) model.

Figure 3(*a*) shows the effect on the room temperature recuperation effect of changing the concentration N_1 of available 110 °C TL traps. The value of N_1 affects the rate at which the recuperated signal approaches equilibrium. Also, as the value of N_1 is increased, the maximum per cent recuperation signal increases.

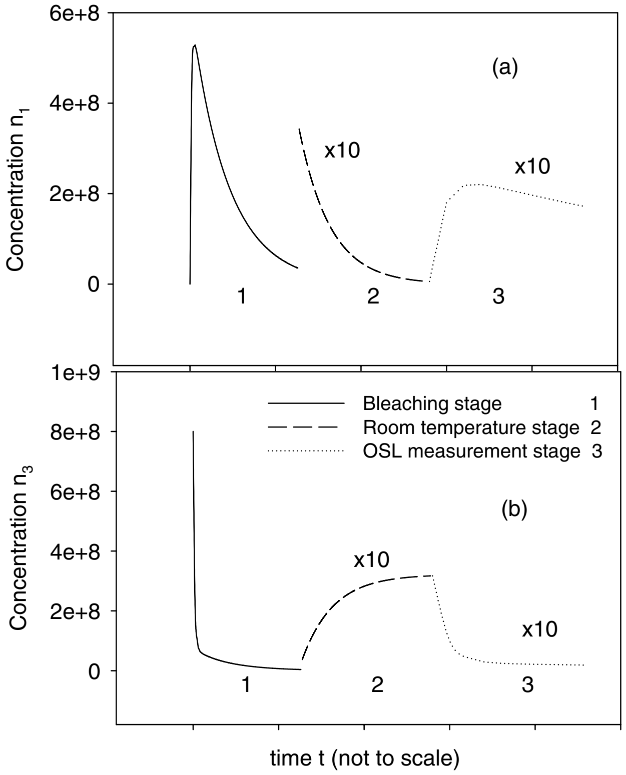

Figure 4. Simulated electronic charge transfer occurring during the Aitken and Smith (1988) procedure, leading to the thermal transfer effect involving the 110 °C TL traps of quartz: (*a*) concentration of electrons in the 110 °C refuge traps (n_1) and (*b*) concentration of electrons in the fast OSL traps (n_3).

Figure 3(*b*) shows the effect on the room temperature recuperation effect of changing the concentration N_3 of available fast OSL traps. The value of N_3 affects the rate at which the recuperated signal approaches equilibrium in a complex, non-monotonic, manner. Furthermore, as the value of N_3 is increased, the maximum per cent recuperation signal decreases. This result makes physical sense, since an increase in N_3 means an increased probability of electrons being retrapped at the OSL traps during the bleaching stage, instead of these electrons being transferred into the refuge trap (level 2). This means that fewer electrons will be available for thermal transfer from the refuge trap into the main OSL traps during sample storage; this will result in a smaller measured recuperated OSL signal.

Figure 4 shows a schematic diagram of the simulated electronic charge transfer occurring during the experimental procedure of Aitken and Smith (1988), leading to a thermal transfer effect involving the 110 °C TL peak of quartz. During the bleaching of the OSL signal (stage 1 in figure 4(*a*) and (*b*), some of the electrons transfer from the OSL traps into the 110 °C TL trap (level 1). The concentration of electrons in the fast OSL trap (n_3) gets reduced to zero by the bleaching (figure 4(*b*)), while the concentration of electrons (n_1) in the refuge traps initially increases from zero to $\sim 5.5 \times 10^8\,\mathrm{cm^{-3}}$, with a subsequent decrease to a value of $\sim 3.5 \times 10^7\,\mathrm{cm^{-3}}$ as bleaching continues. This shows that electrons in the 110 °C TL traps are optically bleachable, but to a much lesser extent than the electrons in the fast OSL traps. By examination of the electron concentrations shown at the end of stage 1 in figure 4, we estimate that during the bleaching process ~4% of the electrons released from the deep OSL traps (n_3) get recaptured in the 110 °C TL level (n_1). This estimate is of the same order of magnitude as the estimates given by Murray and Wintle (1999) based on the data of Smith and Rhodes (1994) and Wintle and Murray (1997). These authors calculated that between 10% and 23% of the electrons ejected from a deep OSL trap during continuous stimulation for 100 s are retrapped by shallow traps (Murray and Wintle 1998, p 77).

During the subsequent storage stage of the sample at room temperature (stage 2 in figure 4), some of the electrons in the refuge 110 °C TL trap transfer thermally back into the main OSL traps (level 3); the concentration N_3 increases from zero to a value of $n_3 \sim 3 \times 10^7$ cm^{-3} during storage at room temperature (stage 2 in figure 4(*b*)), while the concentration n_2 of the refuge traps drops to zero. These transferred electrons in the main OSL traps lead to an increased recuperated OSL signal during subsequent OSL measurements (stage 3 in figure 4). Since this experiment involves storage at room temperature on a laboratory time scale, there are no significant changes relating to levels 6 and 7, the non-radiative recombination centres that are related to changes in luminescence sensitivity.

In summary, although good agreement can be obtained between the model and this single set of experimental results (figure 2), it does not mean that this is really the correct set of parameters. There are perhaps other sets of parameters that can give such comparable fits for a given experimental result. Nevertheless, the success of the model lies in the fact that it can provide a mathematical description of experimental data relating to thermal transfer phenomena; thus, it provides useful information for understanding the mechanisms involved in thermal transfer occurring at room temperature.

2.2. *Simulation #2: temperature dependence of recuperated OSL*

2.2.1. Measured OSL signal. In their second experiment, Aitken and Smith (1988) measured the effect on the recuperated OSL signal of heating to successively higher temperatures and then immediately cooling to 17 °C. The sample had been bleached previously using an argon ion laser, causing electrons to be transferred to refuge traps from which they could be removed by subsequent heating. The recuperated OSL signal was measured using a brief laser stimulation at 17 °C between each heating. The outline of our computer simulation is shown in figure 5, where the first five steps are again a simulation of the 'natural' quartz sample according to Bailey (2001); the remaining steps are a simulation of the steps in the experimental procedure of Aitken and Smith (1988).

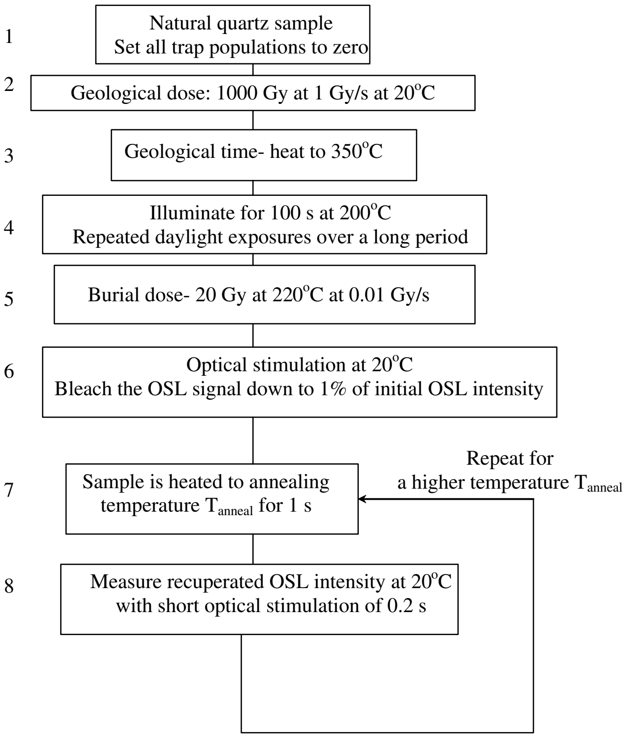

Figure 5. Outline of the experimental OSL/annealing procedure of Aitken and Smith (1988), which is simulated using the parameters of table 2.

The modified set of parameters used in this part of the simulation is given in table 2. Altogether, it was found necessary to change a total of eight parameters (N_1, N_2, N_3, N_8, N_9, A_2, A_3 and s_1) out of 43 available parameters in the Bailey model, in order to get good agreement with the experimental data. The modified values of A_2, A_3 and N_8 given in table 2 were chosen to match the values of these parameters from table 1. The value of N_1 was increased by two orders of magnitude (from 1.5×10^7 to 1.25×10^9 cm^{-3}) in order to match the low temperature behaviour of the recuperated OSL signal between room temperature and 200 °C; this is similar to the value of 5.1×10^9 cm^{-3}, the parameter used in the first simulation (table 1). E_1 was unchanged at 0.97 eV and s_1 was increased by a factor of 10 to 1×10^{14} s^{-1} in order to match the rapid increase in the OSL signal between room temperature and 120 °C. The value of N_2 was increased by an order of magnitude (from 1×10^7 to 1×10^8 cm^{-3}) in order to match the small peak in the experimental data around 280 °C. (Note that level 2, the trap for TL at 230 °C, did not play a role in the room temperature simulation and was thus not changed in table 1.)

The results of simulating the steps in figure 5 are shown in figure 6(*a*), where the per cent recuperated OSL signal is shown as a function of the annealing temperature T. The experimental data from figure 4(i) of Aitken and Smith (1988) are redrawn for direct comparison with the model. The good agreement is achieved by the selection of the parameters (given in table 2) in order to obtain a good fit. However, several features can be seen in both the experimental and the computer-generated data in figure 6(*a*).

Firstly, the increase in the recuperated OSL after heating to temperatures up to ~80 °C is caused by successive thermal transfer from the 110 °C TL peak, from level 1 to level 3, as seen in the simulation for room temperature storage (section 2.1).

Secondly the recuperated OSL remains constant for additional heating to temperatures between 80 and 180 °C. Since no further charge can be transferred from level 1, the use of short optical stimulations (step 8 in figure 5) results in charge remaining in level 3.

Thirdly, an increase in the recuperated OSL is seen between 220 and 280 °C. This increase is primarily due to

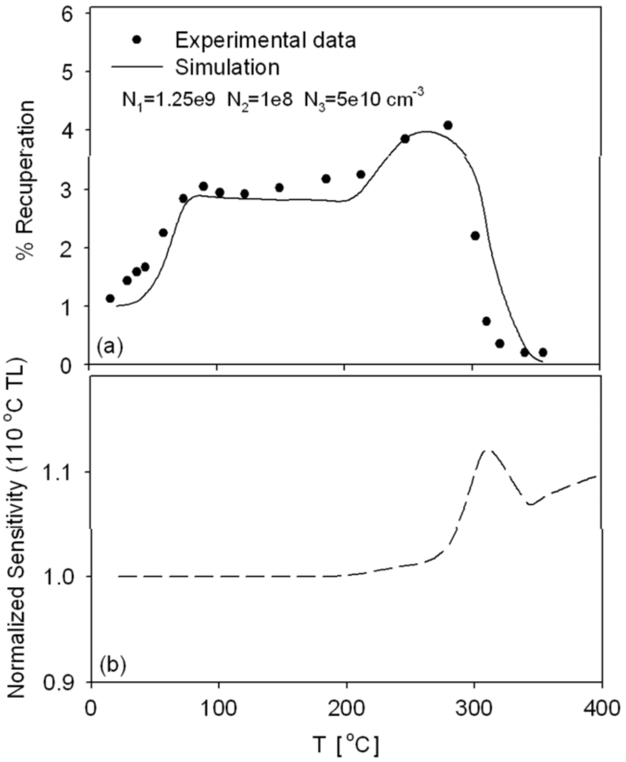

Figure 6. (*a*) The results of simulating the steps shown in figure 5. The per cent recuperated OSL signal is shown as a function of the anneal temperature T. The experimental data from figure 4(i) of Aitken and Smith (1988) are redrawn for direct comparison with the model. (*b*) Simulated sensitivity monitored using the height of the 110 °C TL peak.

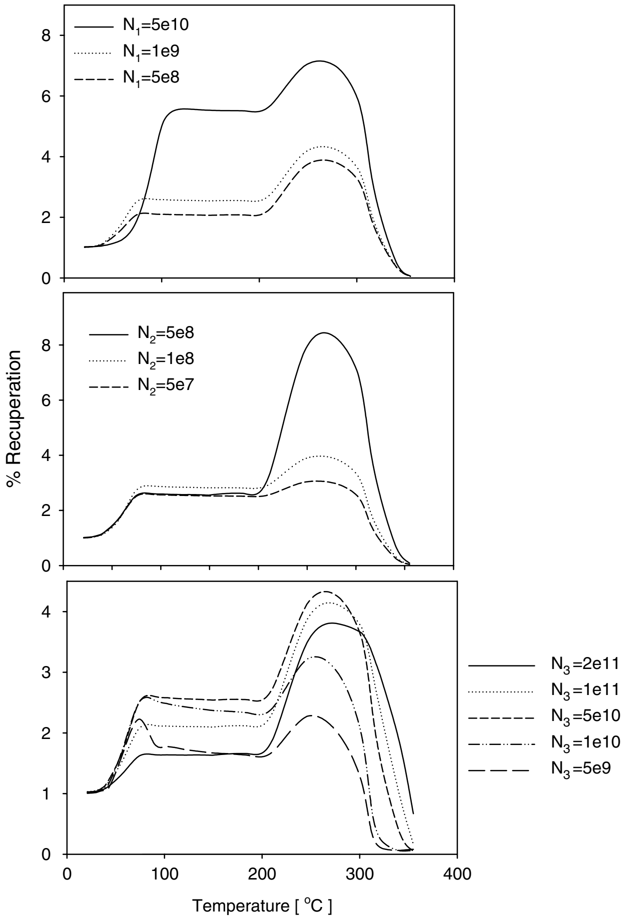

Figure 7. Numerically simulated graphs for the temperature dependence of the recuperated OSL signal obtained by varying concentrations (*a*) N_1, (*b*) N_2 and (*c*) N_3.

the transfer of charge from the TL trap at 230 °C (i.e. level 2) into the OSL traps (levels 3 and 4). However, heating to these temperatures will also give rise to a change in luminescence sensitivity (Wintle and Murray 2000). Indeed, using both an OSL response to a test dose and the response of the 110 °C TL peak, Aitken and Smith (1988) reported an increase in luminescence sensitivity over this temperature range, but no data were shown. An increased signal from the 110 °C TL peak was reported by Wintle and Murray (1998) when their naturally irradiated sample was heated to successively higher temperatures from 200 to 280 °C. In order to investigate the effect of such sensitivity changes on the data in figure 6(*a*), the response of the 110 °C TL peak was simulated using the model. The simulation uses the first five steps of the simulation in figure 5, but the sample is then optically bleached. A simulated dose of 0.2 Gy was used to measure the 110 °C TL peak height and the response as a function of the annealing temperature is shown in figure 6(*b*). The results for this simulation can be compared with the experimental data of Wintle and Murray (1999) who showed only a small OSL sensitivity increase for a bleached sample that had received no additional dose after bleaching (except for the test dose). The sensitivity increase between 220 and 280 °C is very small (<3%), thus confirming that the peak in figure 6(*a*) is mainly caused by thermal transfer of electrons and not by increased luminescence sensitivity.

Finally, the drop in recuperated OSL signal after heating to ∼300 °C is due to the thermal release of electrons from the main OSL traps. These traps become thermally unstable and release the electrons at this temperature range, leading to reduced concentrations of electrons being available at levels 3 and 4.

2.2.2. Sensitivity to parameter changes of recuperated OSL as a function of annealing temperature. It is interesting to note that Bailey (2001) identifies the importance of concentrations in obtaining better fits. Since three of the major changes to the parameters of the original model (Bailey 2001) relate to the concentrations of levels 1, 2 and 3 (i.e. N_1, N_2 and N_3), it is important to demonstrate the effect on the simulated OSL output of changing these concentrations one by one. Figure 7(*a*) shows the effect of the concentration N_1 (the 110 °C TL traps) on the temperature dependence of the recuperation effect. As the value of N_1 is increased, the recuperated signal increases at all temperatures. This is due to the fact that as N_1 increases, more electrons are trapped at the 110 °C TL trap during the bleaching process, leading to an increased number of electrons being transferred thermally into the OSL traps. To obtain the output in figure 6, a value for N_1 of 1.25×10^9 (cm^{-3}) was chosen, close to the middle value of 1×10^9 (cm^{-3}) shown in figure 7(*a*). Changing this parameter has no effect on the overall shape of the curve above 100 °C.

Figure 7(*b*) shows the effect of the concentration N_2 (the 230 °C refuge traps) on the recuperation effect. As might be expected on physical grounds, the value of N_2 affects only the higher temperature region of the data between 200 °C and 400 °C. As the value of N_2 is increased, more electrons are trapped in the 230 °C TL trap than in the 110 °C TL trap, leading to an increase in the recuperated OSL signal at higher temperatures. To obtain the output in figure 6, a value for N_2 of 1.5×10^8 (cm^{-3}) was chosen, close to the value of 1×10^8 (cm^{-3}) shown in figure 7(*b*).

Figure 7(*c*) shows the effect of the concentration N_3 (the fast OSL traps) on the recuperation effect. As the value of N_3 is increased, the maximum per cent recuperation signal increases, but the shape of the curve remains similar above 100 °C. For values of N_3 above 5×10^{10} cm^{-3}, the value used to obtain the output in figure 6, the decrease above 300 °C is insufficient to match the experimental data.

3. Comparison with output using Bailey's parameters

Bailey (2001) performed a series of experiments on a number of sedimentary sands to empirically constrain various parts of the model. We have repeated several of his simulations using both his original parameters and the parameters in tables 1 and 2 in order to ascertain whether the new set of parameters in this paper are consistent with the previous experiments and simulations (Bailey 2001).

The first data set is related to measurements of phototransfer efficiency as a function of illumination time; this monitors any possible changes in the available charge transfer pathways in quartz. Bailey (2001,section 3.1.1) measured the ratio PTTL/OSL of phototransferred TL (PTTL) to the OSL signal during 0.1 s illumination, using the 110°C TL peak. The simulations and experiments consisted of successive partial removal of the OSL signal, inducing a PTTL peak with OSL (0.1 s) illumination, and heating up to 160 °C at 2 °C s^{-1} to measure the PTTL peak. The values obtained for the PTTL/OSL ratios obtained for five samples ranged from $\sim$ 0.03 to 0.05, similar to the value of 0.05 reported by Murray and Wintle (1998). In a previous study, Bailey (1997) had reported that the transfer efficiency ratio PTTL/OSL remained constant as a function of the illumination time.

By using the parameters in tables 1 and 2 we found that the ratio PTTL/OSL indeed remains constant in our simulations, even when the OSL signal has been bleached down to 1% of its initial value. This verifies that no changes occur in the charge transfer pathways when using the new set of parameters in tables 1 and 2. However, the values obtained for the PTTL/OSL ratio in our simulations (given in table 3) were rather high, a value of 0.44 for the parameters in table 1 and 0.17 for those in table 2. These values are 4 times and 11 times larger than the value of the ratio PTTL/OSL $= 0.041 \pm 0.008$ obtained in the experiment by Bailey (2001) and the value of 0.041 obtained by us using his parameters (table 3).

Bailey also assessed changes in the sensitivity of the 110 °C TL peak due to irradiation, annealing and illumination of quartz samples (Bailey 2001, figures 2 and 5).

The second data set he chose was related to the optical de-sensitization of the 110 °C TL peak. The peak obtained in response to a small beta dose was measured before (χ_0) and after (χ_I) optical stimulation at 160 °C. Bailey's experimental data set showed a reduction in signal between 0% and 10%, with a mean value for this optical desensitization (χ_I/χ_0) of 4.46±3.13%. Using his model parameters, we obtained 4.94% in agreement with his simulated value of 4.74%, but values of only 3% when using parameters from table 1 and 1.5% using parameters from table 2. Optical de-sensitization is not significantly affected by changes in concentration parameters (N_i), and is primarily dependent on A_i and B_i.

The third data set was related to dose quenching, the name given to the phenomenon of reduced sensitivity caused by irradiation, observable when the 110 °C TL peak is measured before (χ_0) and after (χ_q) a further small dose and optical sensitization measured before (χ_q) and after (χ_R) 100 s illumination at 160 °C. These data were obtained using a single sequence (Bailey 2001, figure 5) and he obtained a simulated value of $\chi_\mathrm{q}/\chi_0 = 23\%$ for the percent quenching due to a beta dose of 20 Gy. Using the parameters in table 1, our simulations yield a value of $\chi_\mathrm{q}/\chi_0 = 8\%$ for a beta dose of 20 Gy, and using the parameters in table 2, $\chi_\mathrm{q}/\chi_0 = 11\%$. A simulated value of $\chi_\mathrm{R}/\chi_\mathrm{q} = 0.33\%$ was found for the per cent recovery of optically activated sensitivity from the quenched state due to OSL measurement at 160 °C. Our simulations gave $\chi_\mathrm{R}/\chi_\mathrm{q} = 0.4\%$ for both sets of parameters. Once again, these two ratios (χ_q/χ_0 and $\chi_\mathrm{R}/\chi_\mathrm{q}$) are hardly affected by the trap concentrations (N_i) for $i = 1$–5, but are more affected by the concentrations of recombination centres (N_i) for $i = 6$–9.

The fourth data set was related to the subsequent recovery of sensitivity when the quartz is heated, measured as the thermally activated sensitivity change (χ_T/χ_0). Our simulations of the changes in thermal sensitivity (table 3) gave very similar results to the results by Bailey 2001, figure 7) when holding the sample at 220 °C for a fixed period of time.

4. Conclusions

This paper presents the first results of using the kinetic model of Bailey (2001) to obtain simulated data sets for TT-OSL signals measured at room temperature (17 °C).

The first simulation presented in this paper provides a mathematical description of the charge transfer processes which can take place when quartz is stored at room temperature, after it has been optically bleached. The simulation shows that the 110 °C TL traps act as the refuge trap for this thermal transfer process. Parameters taken from the model of Bailey (2001) needed to be changed, in order for the results to be in agreement with the published experimental data of Aitken and Smith (1988).

The second simulation provides a mathematical description of the temperature dependence of the recuperated OSL signal, obtained by heating quartz up to successively higher temperatures, and then monitoring the recuperated OSL signal using short optical stimulations at room temperature in between each step. The model output shows the general shape of the dependence found by Aitken and Smith (1988); however, by further modification of the parameters, the simulation can be made to be in even closer agreement.

The new parameter values were used to calculate a series of sensitivity changes, suggested by Bailey (2001) as being tests of his model. There is general reasonably good agreement between the sensitivity values obtained by Bailey (2001) and the values obtained using the parameters in tables 1 and 2 in the present simulations. However, when either set of new parameters is used to calculate the PTTL/OSL ratio, one of the tests proposed by Bailey (2001), substantially different values were found. This demonstrates the importance of parameter selection and suggests that the parameters suggested in this paper require further modification, particularly the concentrations of the traps giving rise to the 110 °C TL and the fast component of the OSL. However, use of the original Bailey (2001) parameters did not result in the correct behaviour for TT-OSL as a function of storage time at room temperature.

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