GEOCHRONOMETRIA Vol. 23, pp 9-14, 2004 – *Journal on Methods and Applications of Absolute Chronology*

APPLICATION OF A GENETIC ALGORITHM TO FINDING PARAMETER VALUES FOR NUMERICAL SIMULATION OF QUARTZ LUMINESCENCE

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Key words: SENSITIVITY CHANGES, TAC, THERMOLUMINESCENCE, QUARTZ, SIMULATIONS, GENETIC ALGORITHM

Abstract: **The paper presents an application of a Genetic Algorithm (GA) to finding appropriate parameter values for the numerical simulation of quartz thermoluminescence (TL) and Optically Stimulated Luminescence (OSL). It is shown that using a genetic algorithm it is possible to achieve a very good match between simulated and experimentally measured thermal activation characteristics of fired quartz.**

1. INTRODUCTION

Quartz is one of the main materials used for the dating of geological sediments and fired pottery. A large body of research has been carried out into its properties in the recent years. The accurate knowledge of its properties will allow the development of more robust and reliable dating techniques.

The thermal activation characteristics (TAC) describes how the sensitivity to radiation of a given sample changes with its heating. The sensitivity changes influence the protocols used for the estimation of the absorbed dose. Therefore, the knowledge of their precise character allows the employment of relevant corrections which increase the precision and accuracy of the methods employed.

Numerical modelling can be an important tool in investigating the sensitivity changes in quartz. Potentially, it allows a comprehensive description of the luminescence phenomena in this material.

The investigated model is a band model of quartz with a number of electron and hole trapping centres. It was first suggested by (Zimmerman, 1971). The observed thermoluminescence and optically stimulated luminescence and the sensitivity changes are a result of charge transport between the centres via the conduction and valence bands. Mathematically, the model comprises a set of several coupled first-order differential equations (Bailey, 2001; Adamiec, 2005), which describe the time evolution of electron and hole populations within quartz under various conditions like irradiation, light illumination, heating. The problem encountered in accurately simulating luminescence using this approach (assuming the basic equations correctly describe the physical system), is finding a set of numerical parameter values that yield model output which accurately reflects the behaviour of real samples. Until now, parameter selection was made by the modeller, by manually changing the model parameters and comparing the outcome of the model with the measured values – a very time-consuming process. Using this long process, a good general model, that predicts most of the key features of quartz luminescence, has indeed been developed (Bailey, 2001).

In the present work we apply, for the first time, a genetic algorithm (GA) to the problem of finding parameter values for the model. We are primarily concerned with the simulation of pre-dose sensitisation in quartz represented by the above mentioned TAC. An attempt to simulate the shape of the TAC was presented in (Adamiec, 2005), achieving qualitative resemblance of the simulated and experimental TACs. Here we show that it is possible to find parameter values providing a significantly better match of the simulated and experimental results by means of an automatic parameter search.

2. EXPERIMENTAL DETAILS

The TL measurements presented here were performed with the use of an automated reader produced by the Risø National Laboratory type TL-DA-12 (Bøtter-Jensen and Duller, 1992). The reader was fitted with the EMI 9635QA PM tube. The emitted light was filtered with a combination of filters Schott BG-39 and Hoya HA-30 (maximum transmission was 55% @ 360 nm, FWHM 60 nm). The heating rate employed was 2 Ks^{-1} during the TL readout and 5 Ks^{-1} during activation stages.

For all the measurement the fraction of 90-125 μm was used. Geological samples were subjected to treatment with HCl and 48% HF and density separation to eliminate heavy minerals in case should they be present. The summary of samples used is given in **Table 1**. Sample Ark (a natural crystal) was crushed and the 90-125 μm fraction was extracted. The grains of all samples were then annealed in air at 1200°C for 10 h.

The samples were irradiated with a β-dose of 20 Gy prior to the measurement of the TAC. Subsequently they were preheated at 180°C. The sensitivities were measured using a test dose of 20 mGy and heating to 180°C. The activation temperatures were in the range of 220°C and 610°C in 30°C steps.

The heating rate employed was 2 Ks^{-1} during the TL readout and 5 Ks^{-1} during activation stages.

Table 1. *A List of samples used in the current work*

Sample symbol	Site	Description
Ark	Unknown	Arkansas crushed single crystal
BDH	N/A	Commercial grained quartz supplied by BDH
SOT	Stoke-on-Trent	Quartz extracted from sand used for porcelain production at Royal Doulton, Stoke-on-Trent
Merck	N/A	Commercial grained quartz supplied by Merck

3. NUMERICAL MODEL

In the present model it is assumed that hole and electron traps are characterised by the depth E_i, frequency factor $s_{e,i}$ ($s_{h,i}$), concentration of traps N_i, concentration of trapped charge n_i and trapping probability $A_{t,i}$. Hole traps are additionally characterised by the recombination probability $A_{r,i}$ (i is the number of trap) and electron traps are additionally characterised by optical detrapping factor g_i (the probability of optical detrapping of an electron per second; it is dependent on the photon flux and the interaction cross section). The populations of charge in the conduction band and valence band are denoted n_c and n_v respectively. It is assumed that the eviction probability of a trapped hole into the valence band per unit of time is governed by the Arrhenius equation and is proportional to $s_h \cdot e^{-\frac{E_h}{kT}}$.

If there are n electron traps (indexed from 1 to n) and k hole traps (indexed from $n+1$ to $n+k$) then the equations describing the charge traffic can be written as follows:

for electron traps

$$\frac{dn_i}{dt} = n_c \cdot (N_i - n_i) \cdot A_{t,i} - n_i \cdot s_i \cdot e^{-\frac{E_i}{kT}} - n_i \cdot g_i \cdot F \qquad (3.1)$$

for hole traps

$$\frac{dn_i}{dt} = n_v \cdot (N_i - n_i) \cdot A_{t,i} - n_i \cdot s_i \cdot e^{-\frac{E_i}{kT}} - n_c \cdot n_i \cdot A_{r,i} \qquad (3.2)$$

for the conduction band

$$\frac{dn_c}{dt} = G - \sum_{i=1}^{n} [n_c \cdot (N_i - n_i) \cdot A_{t,i} - n_i \cdot g_i - n_i \cdot s_i \cdot e^{-\frac{E_i}{kT}}] \qquad (3.3)$$

for the valence band

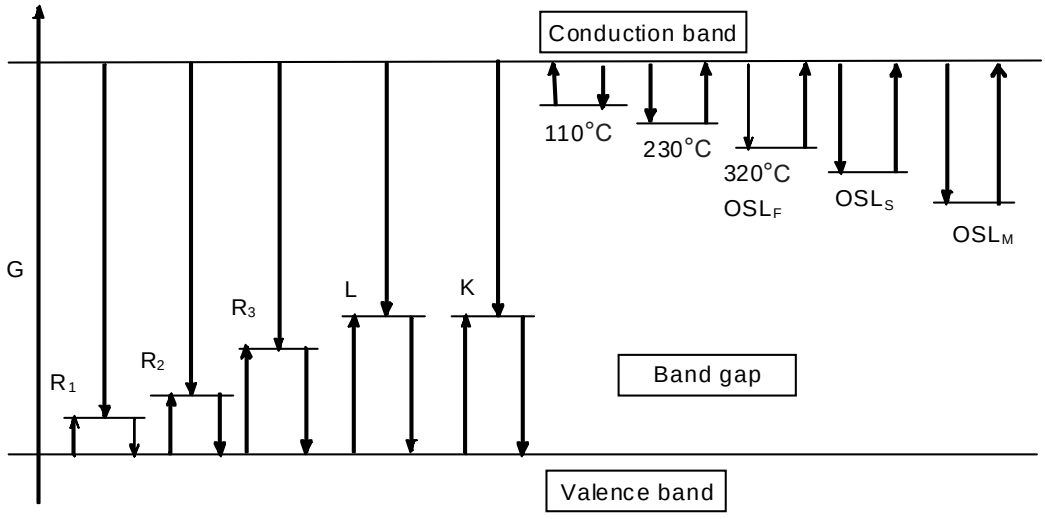

Fig. 1. *A scheme of the investigated model. R_1, R_2, R_3, L and K are hole trapping centres and the remaining ones are electron trapping centres. G indicates the charge pair generation under irradiation*

$$\frac{dn_v}{dt} = G - \sum_{i=n+1}^{n+k} [n_v \cdot (N_i - n_i) \cdot A_{t,i} - n_i \cdot s_i \cdot e^{-\frac{E_i}{kT}}] \qquad (3.4)$$

where G is the charge generation rate (rate of electron-hole production by ionising radiation) during irradiation and F is the photon flux during optical stimulation (not used in the current investigation).

The rate of recombination of electrons with holes, or emission rate, in trap i is calculated as

$$I = n_c \cdot n_i \cdot A_{r,i} \qquad (3.5)$$

The scheme of the general model can be seen in **Fig. 1**.

4. THE GENETIC ALGORITHM

The GA (Holland, 1975) is a method of optimisation inspired by the biological mechanisms of evolution. It starts with a population of individuals randomly generated in a defined search space. In our case, each individual consists of a set of parameter values for the model chosen to simulate the shape of the TAC.

To form the next generation of the population, individuals are selected according to their fitness – the fittest individuals stand a better chance of survival. For each parameter set, the system of differential equations is solved simulating the experimental sequence. Then, the sum of the squared differences (SSD) between experimental and simulated TAC is calculated and defined as the fitness of the individual.

Because selection cannot introduce new individuals, i.e. new points in the search space, two other mechanisms are used with that aim: crossover and mutation.

Crossover combines the genomes of the selected parents to produce an offspring. It is performed on a fixed number of individuals randomly chosen with a probability proportional to their fitness. We used two different crossover operators. The first one obtains the parameters of the offspring genome as a weighted average of the corresponding parents' parameters[1] using random weights (other weighing is also possible, e.g. with weights inversely proportional to the SSD). The second operator swaps random parameters or entire traps in randomly selected pairs of individuals.

The mutation operator modifies, with a low probability, a random parameter from a random individual.

The GA is iterated, creating new generations of constant size, until the fitness function of the best individual in the population does not change significantly over several generations.

There are many advantages of the genetic algorithm. First, GA's are not constrained by assumptions about the search space such as continuity or the existence of derivatives. Secondly, GA's are much less prone to getting stuck in local minima than the traditional local search algorithms because they conduct parallel searches over the whole search space thus improving the quality of the whole population. This feature is particularly valuable in complex search spaces, where a large number of parameters need to be adjusted (in our case 20 or more parameters).

5. IMPLEMENTATION AND RESULTS

The software used here consists of three modules written in C and Borland Delphi.

The first module is the program for editing simulation sequences, which allows creation and modification of simulation sequences. For each step the initial temperature, final temperature, duration, light flux, and dose rate can be specified.

The second module solves the charge transport equations. The inputs are the model parameters and the simulation sequence as well as initial charge concentrations if desired. The solver of the differential equations used was the stiff solver from the libraries supplied by NAG Inc. (Numerical Algorithms Group). The stiff solver allows rapid solution of the charge traffic equations, even when simulating heating to a few hundred degrees Celsius.

The third module implements the genetic algorithm. The algorithm is customizable by a number of parameters provided in a configuration file, for example number of generations, crossovers and mutations per iteration.

New individuals were generated randomly from the defined search space (the ranges of the parameters were chosen based on experience). Instead of using the trap depth E, the equivalent peak temperature was used. The trap depth was then calculated using the peak temperature and the frequency factor s using the first order TL peak formula assuming a heating rate of $2Ks^{-1}$. Using the equivalent peak temperature allowed a more intuitive control of the behaviour of the traps. For parameters s, A_r, A_t and N their logarithms were randomly generated from a uniform distribution from the given range. The logarithms were generated because initially the ranges of the parameters often spanned 4-5 orders of magnitude.

Taking into account the Zimmerman's model, and the results of (Bailey, 2001) and (Adamiec, 2005) models consisting of several traps was used. For sample SOT the same model was used as in (Adamiec, 2005). For the remaining samples

- five electron traps:
  - T_1 responsible for the '110°C peak',
  - T_2 responsible for the TL 230 °C peak,
  - T_3 responsible for the OSL emission (fast) and TL peak in the region of 320°C,
  - T_4 corresponding to the medium OSL component
  - T_5 corresponding to the slow component
- five hole traps:
  - non-luminescent centres R_1 and R_2 responsible mainly for the first sensitisation stage,
  - non-luminescent R_3 responsible for the high temperature sensitivity increase
  - luminescent centre L
  - K-centre

[1] Here the regular weighted average is calculated for the trap depth and optical detrapping probability. For the other parameters, the weighted geometric average is used to allow many orders of magnitude of their values. If w_1 and w_2 are the weights and x_1 and x_2 are the values than the new value is calculated as $\hat{x} = exp\left[\frac{w_1 \cdot \ln(x_1) + w_2 \cdot \ln(x_2)}{w_1 + w_2}\right]$

The description of electron traps T_4 and T_5 should not be taken literally since no additional checks were performed to confirm their labelling.

Bailey (2001) suggests the inclusion of a larger number of electron traps but here it was decided that a TAC curve alone was insufficient to estimate the model parameters for those traps and it was advantageous to decrease the number of searched parameters. It was thought that this would not affect the conclusion about the applicability of the genetic algorithm to the current problem. It can be expected that allowing the presence of additional traps will improve the model's applicability.

In the TAC simulations the dose of 1 Gy was set to produce a charge of $2.5 \cdot 10^{10}$ electron-hole pairs. The dose rate was 0.4 $Gy \cdot s^{-1}$. The sensitivity was measured using a test-dose of 20 mGy and heating to 180°C. All irradiations were performed at 20°C and were followed by a 5 s pause to allow system relaxation (decrease of the charge populations in the conduction and valence bands). After each heating the cooling stages were also simulated to reflect the experimental situation as closely as possible.

To start with, parameter ranges were defined based on previous experience. The GA was then executed for several generations to obtain an initial fit. After the initial fit was obtained sometimes it was necessary to re-adjust the parameter ranges. Also the initial fits were performed with a smaller number of traps and after a fit was obtained the parameters were used to define a new, smaller search space. Also at this stage an additional trap was added to reproduce some of the more detailed features of the TAC. For example in the sample Ark, initially the trap responsible for the slow component was not included. After a fit was reached, the trap OSL_S was included so that its presence would account for the sensitivity decrease in the range 400-500°C.

The calculations were performed for four TACs of fired quartz. The population size was 300 individuals. In each iteration, there were 30 to 60 random pairs and 10-20 best pairs crossed over (1st individual with 2nd, 2nd with 3rd and so on), the best individual was crossed with 10-20 next best individuals, and in 15 random pairs the traps or single parameters were swapped. In each generation, in 20 random individuals random mutations in a random parameter were introduced. There were up to 200 iterations necessary to reach the presented fits.

The experimental and simulated TACs are given in **Fig. 2** and the obtained parameter values are shown in **Table 2**. It can be seen that a very good agreement between experiment and simulations was achieved for all the investigated samples. It can be seen that the degree of sensitisation varies largely between the samples. For sample SOT the maximum sensitisation is around 3 whereas for sample Merck it is close to 300. The differences can be accommodated for by the relevant choice of trap populations and trapping and recombination probabilities.

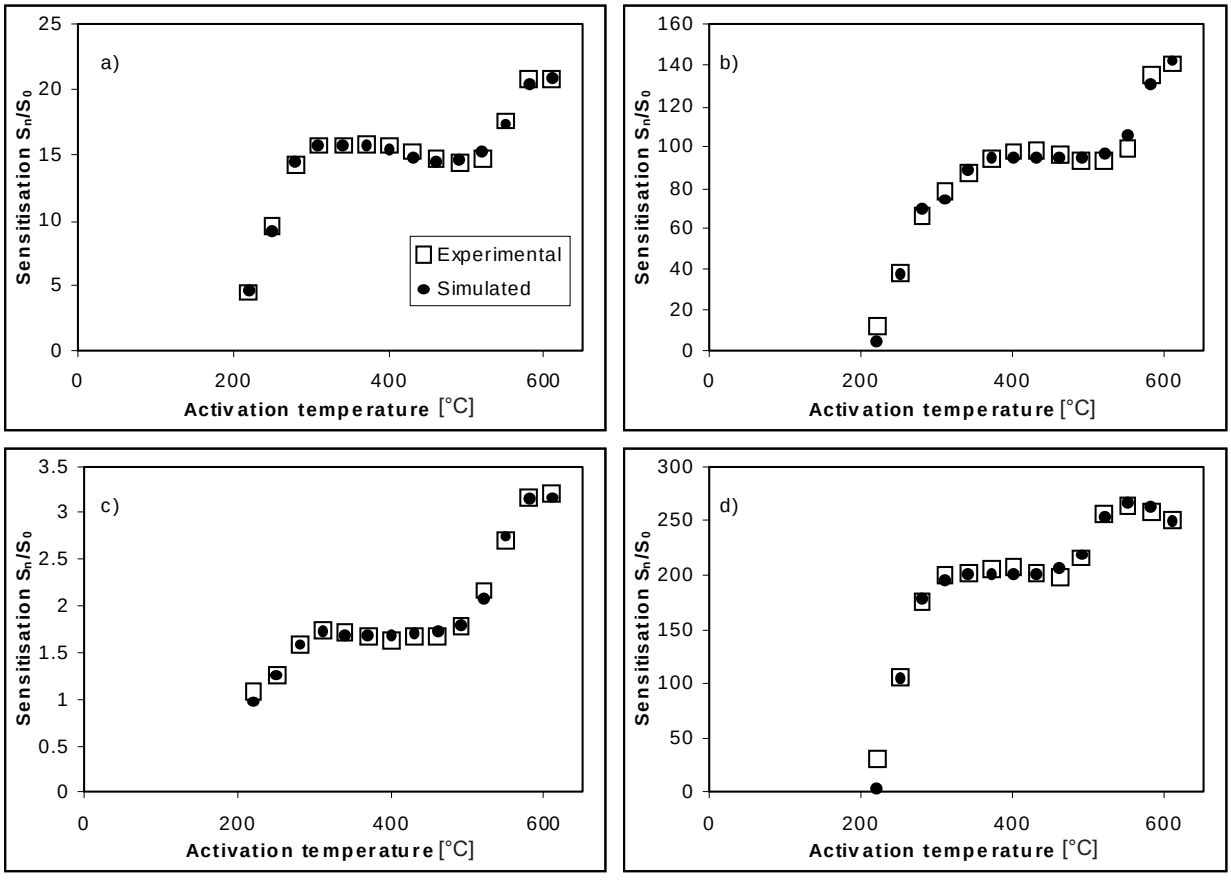

Fig. 2. *The measured and simulated thermal activation characteristics of the investigated samples. a) Ark, b) BDH, c) SOT, d) Merck*

Table 2. A list of parameters obtained for the samples under investigation. The first column contains the calculated equivalent peak temperature as described in the main text

	Equivalent peak temp. (ºC)	Trap depth E (eV)	Frequency factor s	Population	Trapping probability A (s^{-1})	Recombination probability A_r (s^{-1})
Ark						
110º peak	90	0.99	$1.00 \cdot 10^{13}$	$5.28 \cdot 10^{11}$	$5.68 \cdot 10^{-7}$	
230º peak	230	1.55	$5.00 \cdot 10^{14}$	$4.67 \cdot 10^{8}$	$9.01 \cdot 10^{-9}$	
Fast component	312	1.70	$5.00 \cdot 10^{13}$	$4.45 \cdot 10^{11}$	$1.54 \cdot 10^{-10}$	
Medium component	320	1.60	$4.41 \cdot 10^{12}$	$3.52 \cdot 10^{13}$	$1.50 \cdot 10^{-10}$	
Slow component	458	1.91	$1.21 \cdot 10^{12}$	$2.24 \cdot 10^{13}$	$1.75 \cdot 10^{-13}$	
Hole centre R_1	201	1.44	$2.92 \cdot 10^{14}$	$9.09 \cdot 10^{12}$	$2.75 \cdot 10^{-10}$	$6.21 \cdot 10^{-12}$
Hole centre R_2	265	1.38	$1.03 \cdot 10^{12}$	$5.10 \cdot 10^{14}$	$2.26 \cdot 10^{-13}$	$2.09 \cdot 10^{-12}$
Hole centre R_3	573	2.82	$5.58 \cdot 10^{15}$	$4.53 \cdot 10^{11}$	$1.57 \cdot 10^{-10}$	$1.58 \cdot 10^{-16}$
Luminescence centre	1500	5.07	$1.00 \cdot 10^{13}$	$2.76 \cdot 10^{12}$	$2.69 \cdot 10^{-11}$	$1.75 \cdot 10^{-15}$
K centre	1500	5.07	$1.00 \cdot 10^{13}$	$3.77 \cdot 10^{11}$	$1.02 \cdot 10^{-11}$	$1.95 \cdot 10^{-12}$
BDH						
110º peak	90	0.99	$1.00 \cdot 10^{13}$	$2.00 \cdot 10^{11}$	$1.00 \cdot 10^{-10}$	
230º peak	230	1.55	$5.00 \cdot 10^{14}$	$1.46 \cdot 10^{8}$	$1.89 \cdot 10^{-9}$	
Fast component	325	1.69	$1.88 \cdot 10^{13}$	$3.73 \cdot 10^{10}$	$6.11 \cdot 10^{-12}$	
Medium component	446	1.86	$9.28 \cdot 10^{11}$	$6.01 \cdot 10^{13}$	$1.12 \cdot 10^{-12}$	
Slow component	466	2.32	$6.16 \cdot 10^{14}$	$8.10 \cdot 10^{10}$	$3.26 \cdot 10^{-12}$	
Hole centre R_1	206	1.56	$4.68 \cdot 10^{15}$	$5.43 \cdot 10^{13}$	$8.14 \cdot 10^{-10}$	$9.71 \cdot 10^{-11}$
Hole centre R_2	342	1.76	$3.15 \cdot 10^{13}$	$3.23 \cdot 10^{12}$	$6.82 \cdot 10^{-11}$	$2.14 \cdot 10^{-11}$
Hole centre R_3	585	3.09	$1.35 \cdot 10^{17}$	$6.86 \cdot 10^{12}$	$4.91 \cdot 10^{-11}$	$4.51 \cdot 10^{-14}$
Luminescence centre	1500	5.42	$1.00 \cdot 10^{14}$	$3.48 \cdot 10^{13}$	$1.31 \cdot 10^{-11}$	$7.23 \cdot 10^{-15}$
K centre	1500	5.42	$1.00 \cdot 10^{14}$	$1.34 \cdot 10^{11}$	$9.72 \cdot 10^{-11}$	$1.36 \cdot 10^{-13}$
SOT						
110º peak	89	0.70	$7.07 \cdot 10^{8}$	$1.00 \cdot 10^{8}$	$2.00 \cdot 10^{-9}$	
230º peak	252	1.55	$5.00 \cdot 10^{14}$	$1.00 \cdot 10^{7}$	$1.00 \cdot 10^{-8}$	
Fast component	308	1.60	$8.50 \cdot 10^{12}$	$1.00 \cdot 10^{9}$	$3.00 \cdot 10^{-10}$	
Deep electron trap		4.00		$5.00 \cdot 10^{10}$	$2.00 \cdot 10^{-11}$	
Hole centre R_1	243	1.06	$2.06 \cdot 10^{9}$	$1.81 \cdot 10^{13}$	$2.77 \cdot 10^{-12}$	$1.43 \cdot 10^{-12}$
Hole centre R_2	295	1.86	$4.44 \cdot 10^{15}$	$1.62 \cdot 10^{12}$	$1.08 \cdot 10^{-11}$	$1.20 \cdot 10^{-13}$
Hole centre R_3	551	2.48	$1.26 \cdot 10^{14}$	$1.76 \cdot 10^{13}$	$2.96 \cdot 10^{-13}$	$4.35 \cdot 10^{-12}$
Luminescence centre		3.20		$4.44 \cdot 10^{11}$	$1.20 \cdot 10^{-11}$	$1.40 \cdot 10^{-11}$
Merck						
110º peak	90	0.99	$1.00 \cdot 10^{13}$	$1.80 \cdot 10^{11}$	$1.00 \cdot 10^{-10}$	
230º peak	230	1.55	$5.00 \cdot 10^{14}$	$5.69 \cdot 10^{8}$	$8.23 \cdot 10^{-9}$	
Fast component	312	1.70	$5.00 \cdot 10^{13}$	$7.29 \cdot 10^{10}$	$2.40 \cdot 10^{-11}$	
Medium component	451	2.23	$3.20 \cdot 10^{14}$	$2.20 \cdot 10^{10}$	$2.48 \cdot 10^{-14}$	
Slow component	597	2.28	$1.19 \cdot 10^{12}$	$2.95 \cdot 10^{11}$	$4.40 \cdot 10^{-10}$	
Hole centre R_1	193	1.41	$2.40 \cdot 10^{14}$	$9.01 \cdot 10^{13}$	$2.21 \cdot 10^{-10}$	$1.58 \cdot 10^{-9}$
Hole centre R_2	269	1.57	$5.40 \cdot 10^{13}$	$1.68 \cdot 10^{12}$	$6.31 \cdot 10^{-11}$	$2.50 \cdot 10^{-13}$
Hole centre R_3	524	2.68	$9.49 \cdot 10^{15}$	$3.25 \cdot 10^{12}$	$4.70 \cdot 10^{-11}$	$2.84 \cdot 10^{-14}$
Luminescence centre	1500	5.07	$1.00 \cdot 10^{13}$	$2.28 \cdot 10^{13}$	$2.09 \cdot 10^{-11}$	$2.99 \cdot 10^{-13}$
K centre	1500	5.07	$1.00 \cdot 10^{13}$	$2.54 \cdot 10^{11}$	$2.82 \cdot 10^{-14}$	$1.45 \cdot 10^{-11}$

The calculations were very intensive and lasted approximately 24-72 hour operation of a PC with a Pentium IV processor. Although it cannot be excluded that in the future it might be possible to reduce the calculation time using specialised versions of the genetic algorithm, as well as using more experimental data.

6. DISCUSSION AND FINAL CONCLUSIONS

The work is at initial stages but the initial results are very encouraging. The obtained match is very good. It shows that the shape of the thermal activation characteristics can indeed be reproduced by the investigated model – at least in fired quartz.

In the current work only the form of the TAC was considered. For future work there exists the need to take into account more experimental data.

In the case of using a multitude of experimental data an appropriate definition of the fitness function used in the GA plays a crucial role so that the different experimental results have an appropriate influence on the final result.

It could be advantageous to fit some parameters separately thus allowing even better performance of the genetic algorithm. For example it could help to employ the isothermal sensitisation curves, dose growth curves and similar measurements. The isothermal sensitisation curves could allow fitting of some of the parameters, for example the trap depths and frequency factors of traps R_1 and R_2 and later from other data the remaining parameters.

It might also be possible to employ traditional minimisation algorithms once the fit obtained using the GA is close, similarly as in (Garcia-Talavera and Ulicny, 2003).

One of the problems not dealt with so far is the thermal lag during the heating stages. Although the model parameters reproduce experimentally observed behaviour, they are not necessarily the "real" values of the parameters because they are affected by the thermal lag (this will be especially true for the high temperature data). One of the possibilities to deal with the problem would be the introduction into the differential equations of a term allowing for the thermal lag – such an equation would have to be determined experimentally for the used measuring system.

Despite that the results presented here are only preliminary, it can be expected that approaches like the genetic algorithm may help to further improve future models of luminescence in quartz and allow finding parameters for a wide range of samples and therefore allowing better possibilities for estimating various absorbed dose estimation techniques.

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ACKNOWLEDGEMENTS

G.A. would like to acknowledge the Marie-Curie fellowship (project ref. No. HPMD-GH-00-00032-01), which he was receiving during the research presented.

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