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Simulation of an Anomalous Behavior of Thermoluminescence Glow Peak of Quartz from Nigeria

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ABSTRACT

Mechanism of the experimentally observed anomalous shift of a thermoluminescence (TL) glow peak that varied with irradiation dose is yet to be fully established. A theory that the anomalous peak must have contained more than one first order composite peaks each with different TL dose characteristics has been one major explanation proposed to explain this observation. This work was undertaken to simulate the anomalous glow peak by using a modified version of a previously proposed model and numerically solving sets of simultaneous differential equations governing the stages of TL phenomena (excitation, relaxation and heating). In the modified model, two additional electron trapping centers were incorporated in order to simulate accurately this glow curve. Computerized glow curve deconvolution (CGCD) analyses was carried out on the simulated glow peak in attempt to retrieve back the electron trapping center energies and to identify their respective peak positions. The outcome of this confirmed the peak to be possibly composite in nature comprising of three overlapping glow peaks at 288, 300 and 317ºC with respective energy gaps of 1.70, 1.73 and 1.75eV. It is therefore further substantiated that this kind of temperature shift of peak with dose resulting from composite glow peaks is possible.

Keywords: Thermoluminescence (TL), Simulation, Anomalous glow peak, Quartz, CGCD analyses

1. Introduction

Nature is rich in quartz and the study of its thermoluminescence (TL) characteristics has made it a very important component for dating and retrospective dosimetry applications [1, 8, 28]. However, the major challenge of quartz is the issue of its luminescence mechanisms that are very complex [22, 28]. This majorly concerns its sensitivity/sensitisation variability and some provenance characteristics that make some luminescence features to be sample dependent. Although existing luminescence dating protocols are based on luminescence features that are universal, many of the important characteristics that are sample dependent are taken care of by existing dating procedures. This is evident in the universal acceptability and reliability of the technique in spite of this setback. However, the increasing recent reports on luminescence basic studies (experimental, theoretical and numerical modeling) reveal the importance and necessity of more basic research on luminescence characteristics for better understanding of its mechanisms and improved applications [23].

Quartz is known to exhibit a number of glow peaks when heated from room temperature to about 500ºC with a constant heating rate. This is one of the provenance dependent characteristics of quartz that is known to be complex. Quartz of different origins have associated glow peaks ranging from 95-110, 150-180, 200-220 as well as 305-325°C, depending on the heating rate used. Similar results, but not exact, are also available in the literature for different quartz samples as well as different detection optics [17, 21, 28, 34, 36]. The influence of excitation dose and thermal treatment on the TL properties of glow peaks in quartz is widely studied [17, 21, 24]. Studies of TL characteristics of quartz [20], CaF_2:Dy [37], muscovite mica [33] among other materials have shown glow curves having overlapping peaks. Peak shifts with variation in radiation dose have also been reported. Such peak shifts had been reported in ThO_2 [31] which was later confirmed numerically possible [18]. In the experimental work of [21]. an anomalous shift in the fourth peak of the glow curve of a Nigerian quartz with respect to irradiation dose relative

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to the first three peaks was reported. They suggested the reason for this to be the fact that the peak may actually be a complex one consisting of multiple first order glow peaks each with different TL dose behaviour.

In an attempt to understand this better, Ogundare and Chithambo [20] carried out computerized glow-curve deconvolution analysis (CGCD) analysis which indicated the presence of at least five peaks at about 80, 146, 199, 295 and 350 °C meaning that the apparently single peak IV may consist of at least two overlapping glow peaks. Generally, glow curves obtained from TL measurements, of quartz in particular, are usually of several overlapping glow peaks in nature. This consequently makes separation of the composite peaks to their individual glow peak difficult, since this is required for analyses. In reality, the 110 °C TL peak of quartz is always isolated and glaring, but the thermally stable peaks that are mostly used for dating are the major problem of overlapping.

What is desirable in basic research and luminescence applications is the individual peak/component of the glow or OSL curve. This is due to the reason that evaluation of the charge stored in respective trap, which corresponds to each peak/component, may be obtained from the area under each peak/component if all the traps make significant contribution to the glow/OSL curves [19]. Furthermore, important information, like trapping parameters, about the traps and subsequently about the crystal in general may be deduced from the shape of each glow peak or OSL component. Therefore, it is essential to separate each glow or OSL curve into their individual glow peak or component respectively. This is mostly necessary when the peak in question is thermally stable, that is, those that are routinely employed for dosimetry or dating purposes [3, 15, 16]. The interaction between traps may however, have significant effect on the shape of glow curves [7].

The computerized glow-curve deconvolution analysis (CGDA) is the general term used for doing this. CGCD has proved to be promising and more easily applied than the experimental approach that is used in determining the number of peaks in a complex TL glow curve of quartz [3, 13, 15, 16]. There is no available experimental procedure for isolating the components of OSL curve.

Another approach towards understanding the complex nature of quartz is by way of simulation/modelling. This has proved very viable in explaining many luminescent phenomena for various materials generally [4, 5, 6, 12, 14, 25, 26]. This process is something identical to virtually synthetizing, say, a TL material with some predetermined properties and subsequently subjecting it to traditional experimental procedures numerically. The results are then processed like experimental observables. The major task in recording achievements in this line is a successful development of relevant models and appropriate selection of input parameters within the experimental range to reproduce experimental results effectively. The success of an undertaking like this, establishes the employed model for the particular phenomena investigated. Different models for each phenomenon has been considerably used; one example is one trap one recombination center model (OTOR) that has been used to simulate many experimental behaviors [9, 29, 30, 32] despite the fact that it is a hypothetical system that has not been replicated in natural materials. Simulation of a complete quartz luminescence system using a single model has not been achieved. However, there exist many models in form of simultaneous differential rate equations that have been used to simulate good numbers of experimental system by selectively varying the trappings parameters [1, 10, 11, 22] an approach that is merely synonymous to taking a synthetic TL material back to the factory to redesign! An all-encompassing single TL model is hoped to emerge in the future when absolute TL mechanisms of samples are unequivocally comprehended with the same set of trapping parameters.

More light could be shed on the complex nature of the quartz reported by [20] and [21] by simulating the model proposed by these authors to confirm if their model will yield the experimentally observed glow curves numerically. Thus, this work is an attempt to simulate this complex glow curve and also to show that the peak under study is a complex one as proposed by these authors.

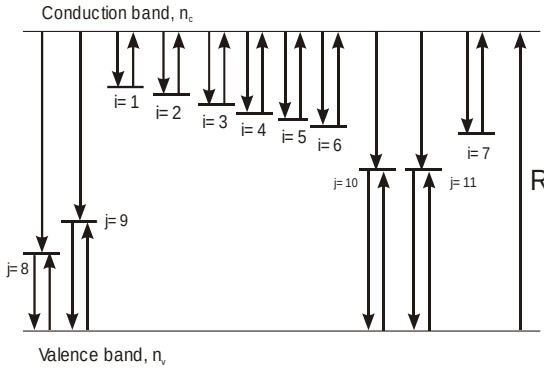
2. Model

The model proposed by [4] describes the general kinetics for OSL and TL stimulated luminescence of quartz. The model consists of five electron-trapping states and four hole trapping states with recombination allowed at these hole-trapping centres. According to [4], Level 1 is a relatively shallow electron trapping centre that gives rise to a TL peak at approximately 100^{o}C at a heating rate of 5^{o}C s^{-1}. Level 2 is included as a medium stability electron trapping centre and it yields a simulated TL peak at approximately 230^{o}C. Levels 3 and 4 are fast and medium OSL components at approximately 330^{o}C and they give rise to photo-luminescence signals. Level 5 represents a stable trap that is extremely thermally stable. Its thermal stability is higher than that of levels 3 and 4. Bailey [4] represents levels 6-9 as hole trapping centres. Levels 6 and 7 depict 'reservoir centres' (R-centre) and are significant in dose quenching and thermal activation characteristics of quartz. Luminescence happens in level 8 as a result of recombination and this level is termed 'luminescence centre' (L-centre). Level 9 is a non-radiative recombination centre though thermally stable (K-centre). Several simulations of TL and OSL processes in quartz have been carried out using this model [4, 22, 25, 27]. For this present simulation, a modified version of [4] model is employed in order to numerically verify the claim by [21] in a bid to explain their experimental observation. This contains two additional electron

trapping centres as depicted by Fig. 1. This is to seemingly account for the experimentally observed multiple overlapping first order glow peaks.

Fig. 1 *Band diagram of the simulated system. Electron trapping centers are denoted by the suffix* i *and the hole trapping centers by* j*. The vertical position of each center indicates the energy* $E_{i,j}$ *of trapped charge.*

The simultaneous differential rate equations to be solved numerically are given below. For each of the $q(q=7)$ electron trapping centers, the change in population with time is,



$$\frac{dn_i}{dt} = n_c(N_i - n_i)A_i - n_i s_i \exp\left(\frac{-E_i}{k_B T}\right) \quad (1)$$

$$i = (1,....,7)$$

The corresponding equation for each of the $r(r=4)$ hole trapping centers is,

$$\frac{dn_j}{dt} = n_v(N_j - n_j)A_j - n_j s_j \exp\left(\frac{-E_j}{k_B T}\right) - n_c n_j B_j \quad (2)$$

$$j = (8,.....,11)$$

$$\frac{dn_c}{dt} = R - \sum_{i=1}^{q}\left(\frac{dn_i}{dt}\right) - \sum_{j=q+1}^{q+r}(n_c n_j B_j) \quad (3)$$

$$(i = 1,.....,7;\ j = 8,.....,11)$$

$$\frac{dn_v}{dt} = R - \sum_{j=q+1}^{q+r}\left(\frac{dn_j}{dt}\right) - \sum_{j=q+1}^{q+r}(n_c n_j B_j) \quad (4)$$

$$j = (8,.....,11)$$

The rate of temperature change is constant,

$$\frac{dT}{dB} = \beta$$

giving $T = T_o + \beta t$ (5)

where T_o is initial temperature. The intensity of the emitted light during recombination was calculated using equation (6) given below

$$L = n_c n_{10} B_{10} \quad (6)$$

where L is the instantaneous luminescence. In the above equations according to [4], N_i is the concentration of electron traps (cm^{-3}); s_i is the frequency factor (s^{-1}); E_i is electron trap depth below the conduction band (eV); N_j is the concentration of hole traps (cm^{-3}); n_j is the concentration of trapped holes (cm^{-3}); E_j is the hole depth above the valence band (eV); k_B is Boltzmann's constant (eVK^{-1}); T is the absolute temperature (K); A_i is the conduction band to electron trap transition probability (s^{-1}); A_j is the valence band to hole trap transition probability (s^{-1}); B_j is the conduction band to hole trap transition probability (s^{-1}); β is the rate of change of temperature (K s^{-1}); R is ionization (pair production) rate (s^{-1}); t is the time (s).

2.1 Numerical Solution

The set of Equations [1 – 6] were solved numerically for certain sets of the parameters using the ode15s Matlab package solver that is designed to deal with 'stiff' sets of equations like the present set of rate equations. In order to simulate more accurately the experimental procedures, an intermediate stage of 'relaxation' in which the irradiation has ceased was added in the simulation. These set of equations is solved for this period with $R = 0$ for a period of time so as to bring n_c and n_v down to negligible values. The final values of n_i, n_j, n_c and n_v at the end of the relaxation time were used as initial values for the heating stage. Excitation and relaxation stages were performed at room temperature (21°C) while the stimulation stage was heated using equation (5) by adopting a constant heating rate (1°C/s).

3. Computerized Glow Curve Deconvolution Analysis

The CGCD of the numerically generated glow curve of the present work was included in this study for easy comparison with the original experimental glow curve of [21]. The difference between the essence of this section and the immediate previous paragraph (section 1.2.1) is essential in order to follow the whole study. While the numerical simulation of section 1.2.1 is synonymous to synthetizing a particular quartz sample with predetermined trapping parameters of Table 1, in this section, attempts are made to numerically retrieve some of the supplied parameters by following the routine procedures of deconvolution. Analytical expressions used for CGCD are based on the TL and OSL kinetic equations. However, the expressions are transformed to the forms that have some experimental measurable parameters unlike the conventional kinetic equations: n_o and s are replaced with I_m and T_m in TL kinetics, whereas, n_0 and s replaced with I_m and t_m in LM-OSL kinetics. T_m is the temperature at glow-peak maximum intensity, I_m while t_m is the time, t, at component maximum intensity, I_m for LM-OSL. General order kinetics for both TL and LM-OSL are often used for CGDA . This is because both general and mixed orders reduce to first order form for TL and LM-OSL cases when the order of kinetics b =1.00001. The transformed expressions of general order for TL are as follows:

$$I(T) = I_m b^{\frac{b}{b-1}} \exp\left(\frac{E}{kT}\frac{T-T_m}{T_m}\right) \times \left[(b-1)(1-\Delta)\frac{T^2}{T_m^2}\exp\left(\frac{E}{kT}\frac{T-T_m}{T_m}\right) + Z_m\right]^{\frac{b}{b-1}} \quad (7)$$

$2kT/E, \quad \Delta_m = 2kT_m/E, \quad Z_m = 1+(b-1)\Delta_m$ and the frequency factor s is calculated using

$$s = \frac{\beta E}{kT_m^2}\exp\left(\frac{E}{kT_m}\right)\left[1+(b-1)\frac{2kT_m}{E}\right]^{-1} \quad (8)$$

The quality of the fit produced by fitting any of these equations to experimental curve is normally tested with figure of merit (FOM) defined by Balian and Eddy (1977) as

$$FOM(\%) = 100 \times \frac{\sum_j |I_j - I(T_j)|}{\sum_j I_j} \quad (9)$$

where I_j and $I(T_j)$ are the experimental and fit intensities in channel j respectively. The background signal is usually simulated by an equation of the form

$$BKG_{LM} = \alpha + ct \quad (10)$$

where α is the average in the first few seconds of a zero dose LM-OSL measurement, and c is a constant.

4. Results and Discussion

The set of equations (1) – (6) were solved as described above with the prime objective of numerically generating the experimentally observed anomalous glow peak of quartz from Nigeria that was reported by [21] (Fig. 2a). The major task that is required for this study is to select appropriate trapping parameters that are within existing experimental range for the simulation that will, at the same time, numerically reproduce the experimentally observed glow curves of Fig.2a. By starting with parameters earlier presented by [4] (though not exactly the same), and after tedious, time consuming and rigorous trial and error of different sets of parameters in the range of experimental values (Table 1), a convincing glow curve of Fig. 2a was obtained. This is shown in Fig. 2b. The irradiation dose was 50 Gy at a heating rate of 1^oCs^{-1}.

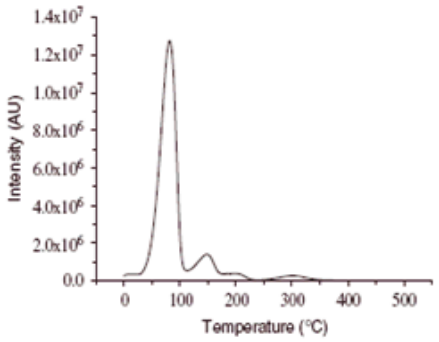


Fig. 2(a) *Experimental result (Ogundare et al., 2006). The four clear peaks are shown with associated temperatures.*

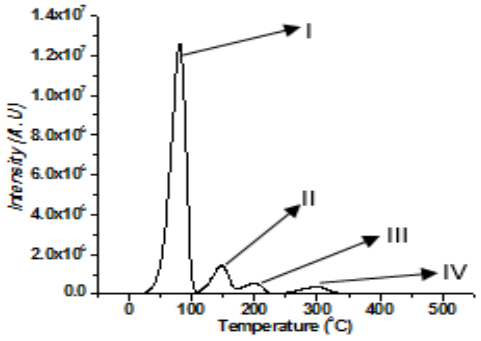


Fig. 2(b) *Simulation result (present study). The peaks occur at 81, 148, 200 and 289°C*

Table 1. Details of numerical parameters of each center

Levels	N (cm^{-3})	E (eV)	s (s^{-1})	A (cm^3s^{-1})	B (cm^3s^{-1})
$i=1$	1.5×10^{10}	0.96	2.1×10^{13}	1.0×10^{-8}	0.0
$i=2$	2.8×10^{9}	1.27	6.0×10^{14}	1.0×10^{-8}	0.0
$i=3$	2.4×10^{10}	1.43	4.8×10^{14}	1.0×10^{-9}	0.0
$i=4$	1.0×10^{11}	1.70	3.5×10^{14}	5.0×10^{-10}	0.0
$i=5$	1.0×10^{11}	1.73	3.0×10^{14}	5.0×10^{-10}	0.0
$i=6$	1.0×10^{11}	1.75	2.5×10^{14}	5.0×10^{-10}	0.0
$i=7$	5.0×10^{13}	2.00	1.0×10^{10}	1.0×10^{-10}	0.0
$i=8$	3.0×10^{11}	1.43	5.0×10^{13}	5.0×10^{-7}	5.0×10^{-9}
$i=9$	1.0×10^{13}	1.75	5.0×10^{14}	1.0×10^{-9}	5.0×10^{-10}
$i=10$	1.0×10^{14}	5.00	1.0×10^{13}	1.0×10^{-9}	1.0×10^{-10}
$i=11$	5.0×10^{12}	5.00	1.0×10^{13}	1.0×10^{-10}	1.0×10^{-10}

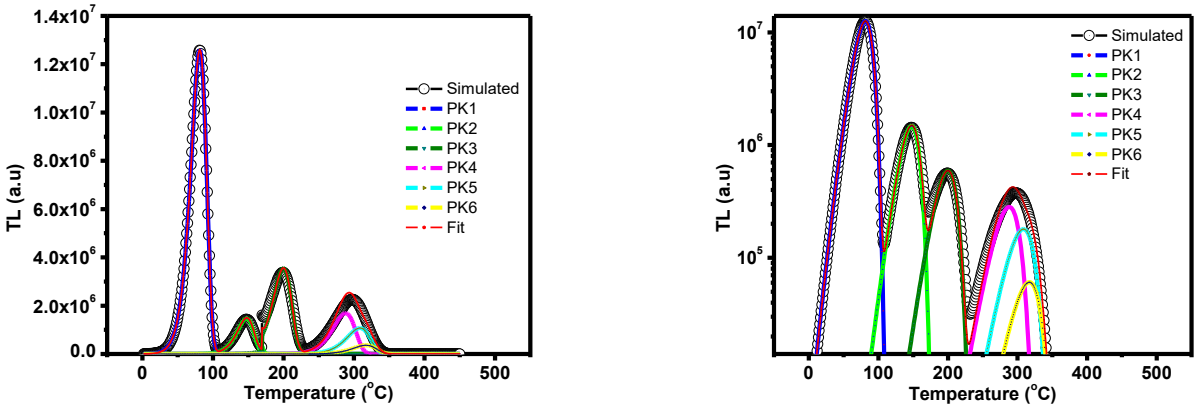


Fig. 3 *CGDA Analysis of glow curve. Irradiation dose was 50 Gy. The figure on the right is for clearer view.*

While this figure shows that the peaks I-IV occur at 81, 148, 200 and 289°C respectively, two additional peaks are expected in peak IV according to the predetermined trapping parameters in Table 1 that were used for the simulation of this glow curve. The result of the deconvolution of this numerically generated glow curve, as described in section 3 is shown in Fig. 3. This was undertaken in order to recover some of the trapping parameters used in the simulation (namely; E and s) and to determine the peak positions of the composite peak IV. The quality of the fit tested with figure of merit (FOM) was 1.71% The glow curve consists of six peaks which suggests that peak IV is composite in nature consisting of three overlapping glow peaks (IVa, IVb and IVc in table 2) at 288, 300 and 317°C with respective activation energies of 1.70, 1.73 and 1.75eV. The revealed six clear peaks from the CGDA show a considerably close result to [20] in which six peaks were obtained at an irradiation dose of 52.7 Gy.

The temperature range of the six peaks also compares favourably with their average temperatures over the range of doses considered. Comparing the present results with theirs is pertinent because in the literature, this kind of anomalous behavior of peak IV with change in irradiation dose which they accorded to multiple overlapping peaks, was first reported by them. Although the present simulation has not given particular attention to dose dependence of the peaks of the glow curve, the observation of six peaks at irradiation dose of 50 Gy may be a reasonable argument to further substantiate their claim that the number of peaks required to fit a glow curve increases with irradiation dose. Another notable observation from the present study is the small variation in the activation energies of peaks IVa, IVb and IVc. This may have in some way, contributed to their appearance as one single peak at low doses. The glow curve obtained using the two traps introduced into the Bailey model used in this study as agreed with the experimentally obtained glow peak of [21] suggests the possibility of composite nature the thermally stable trap.

Table 2. The t_m and E values obtained from numerical CGDA analysis of glow curve obtained from irradiation dose of 50 Gy

TL Peak	t_m (^oC) [21]	t_m (^oC) [20]	t_m (^oC) Present	E (eV)	$s(s^{-1})$	*b*
I	82	80	81	0.96	$4.03x10^{12}$	1.00026
II	148	146	148	1.27	$1.36x10^{14}$	1.00026
III	200	199	200	1.43	$1.29x10^{14}$	1.00026
IVa	300	295	288	1.70	$1.18x10^{14}$	1.00026
IVb		350	300	1.73	$5.90x10^{13}$	1.00026
IVc			317	1.75	$5.25x10^{13}$	1.00026

5. Conclusion

In the present work, an energy level model for a thermoluminescent material that includes seven trapping states (two states added to existing Bailey's model) and four recombination centers has been described. The stages of excitation, relaxation and heating have been followed. It has also been shown that by solving the appropriate sets of coupled differential equations in sequence for appropriate choices of the sets of trapping parameters, four TL peaks occur related to four recombination centers. The situation of applying a given dose at a particular dose rate was then simulated. A numerical deconvolution procedure was then employed to retrieve the preset parameters employed for the simulation of the glow curve. The result of the present study shows six peaks alluding to previously stated experimental observations by some authors that the fourth peak's anomalous behaviour with respect to dose is actually due to it being a composite of at least two peaks. Part of future work is to employ the present model on the dependence of the glow curve behaviour on dose.

Conflict of Interest

The authors have no conflict of interest.

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