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Glow curve analysis of the 110°C glow peak of quartz in the simplified GOT equation

L. Lovedy
RIST, Manipur University, Imphal – 795003, India
Corresponding author: lovedyo1@yahoo.co.in:

Abstract

A recently developed glow curve deconvolution technique which employs the Hybrid Genetic Algorithm has been applied to the 110°C glow peak of quartz. This technique enables one to determine the key intrinsic trapping parameters, namely, the activation energy (E), the frequency factor (s) and the ratio of retrapping probability to recombination probability (α). The analysis shows that the 110°C glow peak of quartz has trap depth of 0.974±0.002 eV, frequency factor of (5.27 ± 0.06) x 10^{12} sec^{-1} and retrapping to recombination ratio in the range of $3x10^{-4} \leq \alpha \leq 3x10^{-2}$. It is proved that retrapping is not negligible; rather it is ≈1/10000 to 1/100 times the recombination probability. The value of N suggests that the trap created by beta irradiation is not linear.

Keywords: Thermoluminescence; OTOR; 110; Quartz

1. Introduction

The highly luminescent properties of quartz such as sensitivity, reliability, versatility and compatibility with the detection system make it extremely useful in luminescence studies. Typical glow peaks of quartz above room temperature are centered around 110, 230, 325 and 375°C [1] though glow curves vary from sample to sample. Thermoluminescence (TL) properties are quite diverse in nature and depend strongly on its origin and the region from which it is derived [2-4]. The effective utilization of the 110° C glow peak of quartz has attracted considerable interest since several unwanted effects due to heating at higher temperatures, such as black-body background radiation, thermal quenching and heat-induced chemical reaction in the specimen may be avoided. Monitoring the increase in the sensitization rather than the conventional accumulation of TL is a mode of measurement which has been utilized to great effect in archaeological dating [2,5-6], retrospective dosimetery [7-9], authenticity testing [10], firing temperature measurements [11] etc. This method also possesses great potential in the estimation of the low equivalent dose (paleo-dose) in the range of ~mGy to a few Gy. Correlation of the 110° C glow peak with Optically Stimulated Luminescence (OSL) has enhanced the importance of this peak [12-14], as a result of which it has been routinely employed in monitoring the sensitivity change occurring in the OSL measurements due to pre-heat, involved in the protocol. It is now widely accepted that the two luminescence phenomena share the same centers [15].

Prior to Lovedy and Gartia [16] the analysis of TL curves was performed using the Order of Kinetics model. The Order of Kinetics model possesses the following drawbacks:

a. It is purely empirical

b. The concept of b = 1 denotes that recombination dominates retrapping, but does not indicate by how much

c. The order of kinetics cannot provide information about the trap created during irradiation.

Recently Lovedy and Gartia [16] developed a simplified form of the OTOR eqn and applied it to the analysis of NaCl [17]. In this paper an attempt has been made to analyse the 110°C TL peak of quartz using the simplified OTOR model of Lovedy and Gartia [16] in order to have first-hand knowledge of the trapping parameters, namely, activation energy, frequency factor (s^{-1}), ratio of retrapping to recombination probability (α), defects created during irradiation (N cm^{-1}) and the ratio of the number of traps created to the number of traps filled (γ). Again, in most analyses related to deconvolution of TL curves, the statistical

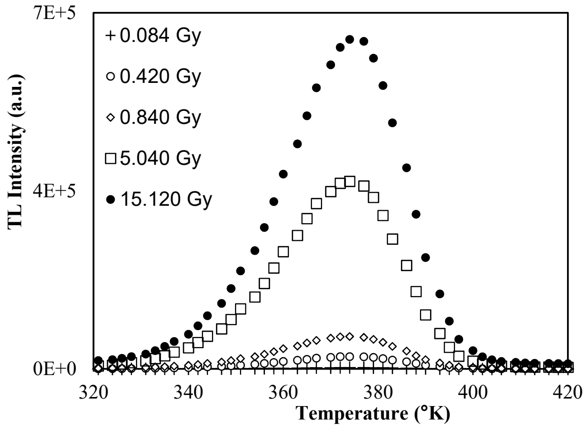

Fig. 1 The 110°C peak of quartz at different beta dose. Inset shows the amount of beta dose given.

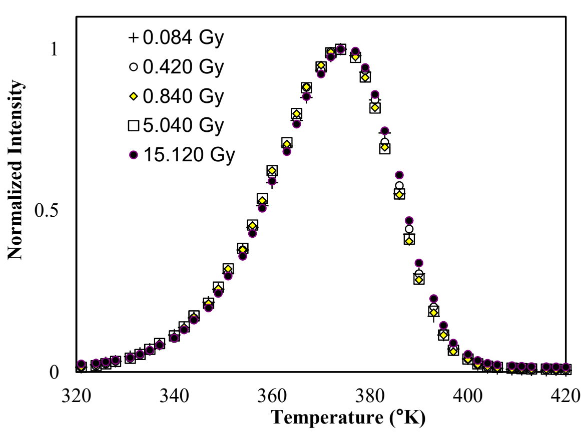

Fig. 2 The normalized plot of the 110°C peak of quartz at different beta dose. Inset shows the amount of beta dose given.

analysis is seldom left behind; in this paper a simple test available in the commercially found Microsoft Excel 2003 has been attempted.

2. Experimental and theoretical techniques

2.1 Experimental

The TL measurement of quartz minerals was performed using the commercial Risø TL/OSL (model TL DA-15A) [18-19]. The samples were irradiated at room temperature with an inbuilt beta irradiation (90Sr/90Y) source at a dose rate of 0.084Gy/sec. The irradiated samples were recorded in a flowing nitrogen atmosphere. Standard clean glass filters (combination of Schott UG-11 and BG-39) were installed in the reader between the sample and the photomultiplier tube (EMI 9635). These filters permit the passage of light whose wavelength is between ~300 and 400nm, and attenuate blackbody radiations emitted by the heating plate at high temperature. The time duration between irradiation and TL readout was always kept constant at about 10 min. A heating rate of 5°C/sec was used for the TL measurement. No preheat was given to the samples. Each experimental measurement consisted of two readouts. The second readout, considered as the background of the reader plus sample was subtracted from the first one.

2.2. Theoretical

Lovedy and Gartia in a recent paper [16] have theoretically derived a simplified form of the OTOR model. This is given by:

$$I_{TL} = \frac{N\alpha s e^{-E/kT}}{\beta(1-\alpha)^2\omega(x(T))(1+\omega(x(T)))} \tag{1}$$

where N(cm^{-3}) is the concentration of traps of the kind responsible for the peak being considered, α is the ratio of the retrapping probabilities to the recombination probabilities, s (s^{-1}) is the frequency factor, E(eV) is the activation energy, β is the heating rate, ω is the Wright omega function (see the annexure of Lovedy and Gartia [16]) and *x(T)* given by:

$$x(T) = \frac{s}{\beta(1-\alpha)}\int_{T_o}^{T} exp\left(\frac{-E}{kT}\right)dT + \left(\frac{N\alpha}{1-\alpha}\right)\frac{1}{n_o} + log\left(\left(\frac{N\alpha}{1-\alpha}\right)\frac{1}{n_o}\right) \tag{2}$$

If we take $\gamma = N/n_o$ then the eqn (2) can be written as:

$$x(T) = \frac{s}{\beta(1-\alpha)}\int_{T_o}^{T} exp\left(\frac{-E}{kT}\right)dT + \left(\frac{\gamma\alpha}{1-\alpha}\right) + log\left(\left(\frac{\gamma\alpha}{1-\alpha}\right)\right) \tag{3}$$

3. Results and Discussion

The TL glow curves of β-irradiated (0.084, 0.42, 0.84, 5.04 and 15.12 Gy) quartz minerals are shown in Fig 1. It may be seen from Fig 2 that T_m does not shift with the increase in dose. This clearly indicates a case of first order kinetics which the OTOR formalism developed by Lovedy and Gartia [16] implies. In order to ascertain these observations, all the glow curves of Fig. 1 are analyzed using CGCD by employing eqn (1), eqn (3) and Hybrid Genetic Algorithm [17]. Prior to the curve fitting using CGCD, the effective heating rate (β) was calculated according to Kitis and Tuyn [20]. The effective heating rate was found to be 4.813°C/sec (c=2.88539), which has been employed in CGCD analysis.

In order to visually check the fitting, a comparison of the experimental (β-irradiated 0.084 and 15.12 Gy) and the CGCD fitted curves is shown in Fig 3 and Fig 4. The goodness of fit may also be judged from the figures. The goodness of fit of the measured glow curve is tested using standard chi tests.

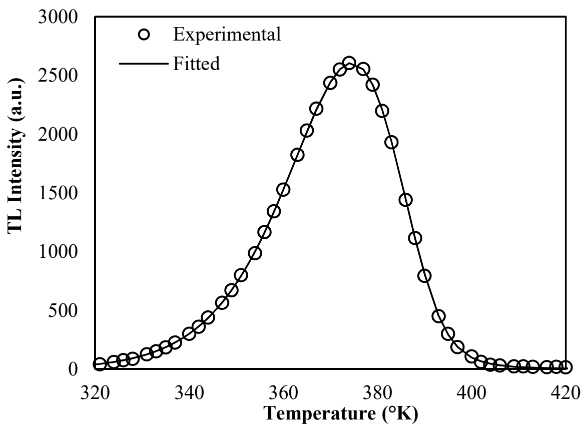

Fig 3. The experimental and simulated curve for the 0.084 Gy irradiated 110°C peak of quartz

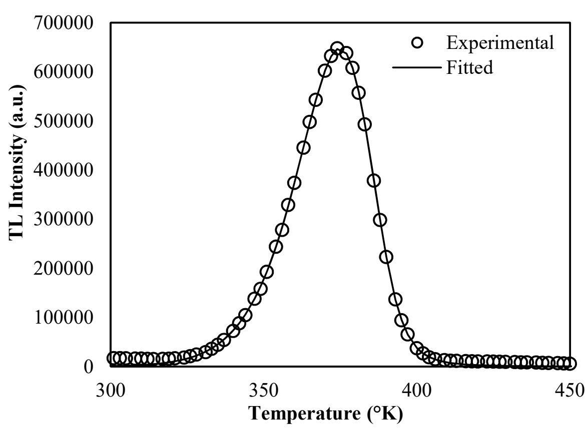

Fig 4. The experimental and simulated curve for the 15.12 Gy irradiated 110°C peak of quartz

The $\chi 2$ test is performed using CHITEST present in Excel 2003. The actual frequency and expected frequency for the CHITEST are calculated using the popular statistical software Statistica. Figure of merit (FOM) [21-22] is also calculated as a cross check. The trap parameters determined through the simplified OTOR eqn and Hybrid Genetic Algorithm are tabulated and shown in Table 1.

Table 1: The TL parameter of 110 °C peak of quartz extracted using the Hybrid Genetic Algorithm in the simplified OTOR eqn (effective heating rate =4.813°C/sec (c=2.88539))

Dose(Gy)	Tm(°K)	E(eV)	s(sec^{-1})	N(cm^{-3})	α	γ	FOM	χ^2 (DOF)
0.084	375	0.971	4.81x10^{12}	4.46 x10^{6}	3.13x10^{-4}	52.984	0.740%	0.939(5)
0.420	375	0.976	2.82x10^{12}	6.49 x10^{6}	1.73 x10^{-3}	8.166	0.530%	0.834(5)
0.840	375	0.979	6.28x10^{12}	7.88 x10^{6}	6.21 x10^{-3}	3.826	0.254%	0.962(6)
5.040	375	0.973	5.01x10^{12}	1.77 x10^{7}	1.52 x10^{-2}	1.479	0.828%	0.998(5)
15.120	375	0.973	4.68x10^{12}	2.35 x10^{7}	3.06 x10^{-2}	1.105	0.762%	0.936(3)

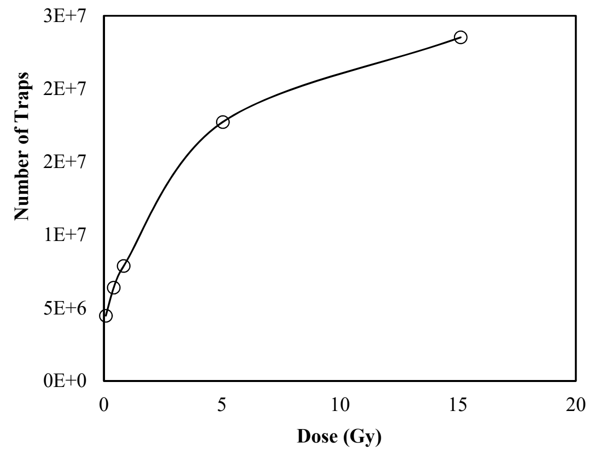

Fig 5. N(cm^{-2}) vs dose(Gy) for 110°C peak of quartz

The FOM values along with the standard $\chi 2$ test values are also presented in Table 1. It is seen from the analysis that the 110°C peak of quartz is characterized by trap depth of 0.974±0.002 eV, frequency factor of (5.27 ± 0.06) x 10^{12} sec^{-1} and retrapping to recombination ratio in the range of 3 x $10^{-4} \leq \alpha \leq 3$ x 10^{-2} . The value of s is in accord with that suggested by Furetta [23] and Brockhouse [24], i.e., 10^{12} sec^{-1}. The value of α suggests that the recombination probability is ≈10000 to 100 times the retrapping probability in the case of 110°C peak of quartz, i.e., it shows first order as well as non-first order behavior. From Table 1 it is seen that the number of traps (N) goes on increasing with dose; this is expected since irradiation creates trap.

Fig 5 shows N vs dose of the 110°C peak. This suggests that the traps created by the beta rays are not linear. According to Tandon et al [25] and Reft et al [26] $^{90}Sr/^{90}Y$ emits electrons with a broad range of energies. This means that the penetration of the electrons is a jumble of different electrons of varying energies, suggests that the electron penetration will not be linear and hence the dose response of $^{90}Sr/^{90}Y$ will not be linear, which is reflected by N vs dose in Fig 5. From Table 1 we find that the value of γ decreases with increase in dose. This suggests that with increase in dose the trap filling increases and equals trap creations.

3. Conclusion

The CGCD analysis of the experimental curves shows that the 110°C peak of quartz is characterized by trap depth 0.974±0.002 eV, frequency factor of (5.27 ± 0.06) x 10^{12} sec^{-1} and retrapping to recombination ratio in the range of 3 x $10^{-4} \leq \alpha \leq 3$ x 10^{-2}. It is proved that retrapping is not negligible, i.e., not of the first order; rather it is ≈1/10000 to 1/100 times the recombination probability. The value of N suggests that the trap created by beta irradiation is not linear.

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