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Thermally Stimulated Luminescence Studies of Silicate Minerals

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ABSTRACT

The present paper reports the thermally stimulated luminescence studies of silicate minerals. The trapping parameters and order of kinetics have been investigated. The thermoluminescence (TL) studies of chert and the TL peak of quartz at various heat treatments and irradiations were recorded. 110^0C, 325^0C, 375^0C peak of quartz under different experimental conditions have been investigated. Trapping parameters involved in the process of TL glow peaks were examined within the framework of kinetics formalism. Natural TL results show that the glow peaks of quartz relevant to dating follow a non first order kinetics.

Keywords: thermally stimulated luminescence; trapping parameters; chert; quartz; first order kinetics.

INTRODUCTION

A large number of research works on thermoluminescence (TL) of natural quartzes reported in the literature are often of empirical nature. However, this has resulted in establishing their general descriptive features and typomorphic potentialities. The glow curves of quartz are of quite intricate in nature and display variability in passing from one genetic type of the sample to another.Among the TL glow curves the three main emission bands at about 360 – 440 nm (near UV to violet), 460 – 500 nm (blue) and 610-650 nm (orange, mostly reported as red TL) have been used for dating. Generally, quartz exhibits TL glow peaks at about 85,110,180,220,325, 370 and 480^oC 9for a heating rate of $5^oC/s$). The preferred peaks for dating are the ones at 110,325 and 375^oC, because 110^oC peak has a high “pre-dose” (sensitization) effect, 325^oC peak bleaches rapidly (RBP) while 375^oC peak, which bleaches slowly (SBP) has sufficient long trap electron lifetimes.The problem of the exact mathematical description of glow curves of material used for any practical applications, dosimetry and dating, still remains unsolved. This limitation in no way has limited the use of the technique because of the well-developed phenomenological description of the process with the introduction of the order of kinetics [1-4]. The TL dosimetry community has given a new dimension to the technical success of the technique for determination of radiation does in terms of reliability and sensitivity by resorting to computerized glow curve deconvolution[5].Incidentally, the order of kinetics is of supreme importance not only from theoretical point of view but also for its practical applications[6]. With this background, the order of kinetics of natural / synthetic TL peaks of quartz has

MATERIALS AND METHODS

Experimental

In the present study a number of variety of quartzes from different origin are chosen:

(I). Chert (a form α–quartz) from near the limestone deposit is heated upto 400°C. The heating rate in most of the cases is from 300°C/min, and recorded using a commercial recording system modelTL-1404.The sample size is ~100μ (after grounded). It was irradiated with γ–rays from a ^{60}Co source with dose rate of 0.5Gy/Sec.

(ii). Synthetic single crystals of α-quartz (Premium Q) with or without firing / air swept were irradiated with γ-radiation from a ^{60}Co source with dose rate of 0.041Gy/sec at room temperature. TL was recorded at a heating rate of 2°C/sec in nitrogen atmosphere [8].

(iii). Isolated 380°C peaks of quartz (flint) at various doses of irradiation (60, 120, 240, 480, 960 and 1920 Gy) at a heating rate of 10°C/sec [9].

(iv).Two natural TL curves with and without standard preheat (for 5 min. at 200°C) at a heating rate of 5°C/s [10] are used in the present investigation

RESULTS AND DISCUSSION

i). Case of 110°c peak of chert / quartz:

The natural silicate mineral is subjected to various heat treatments and irradiations. Then the mineral Thermoluminescence (TL) is recorded and is shown in Fig.-1. The important parameters of the glow peaks are presented in Table-1.The 110°C peaks of chert under different experimental conditions are within the framework of kinetics formalism. The trapping parameters are also presented in Table-1.The values of the trapping parameters (E, s and b) agree well with the earlier results [11,12-13].Even the value of 'E' used in the complex model [14] is in agreement with the present finding though the value of 's' differs.Thus results of the 100°C peak of chert / quartz shows that the order of kinetics is non-consistent following non-first-order kinetics, whereas its values of E and s are consistent with E=0.814 ± 0.03 eV and $s \approx 10^{10}\ S^{-1}$ .In the more generalized models where one considers the presence of competing traps to account sensitization and super-linearity in quartz, first-order kinetics in the case of 100°C peak can not be true for all situations. Incidentally, high concentrations of thermally disconnected traps lead to apparent first-order looking glow curves irrespective of the presence of re-trapping.

ii). Case of 325 and 375°c peaks of quartz.

Isolated 375°C glow peak of flint presented by Valladas [15] under various doses (60, 120, 240, 480, 960 and 1920 Gy) of irradiation with a uniform heating rate β = 10°C/s is shown elsewhere while the peak parameters are presented in Table-2.The results show that with the increase of dose the peak temperature (T_m) shifts to lower temperature indicating non-first-order kinetics[16].Moreover, the shape factor (μ_g) is more or less constant with a value ≈ 0.52 indicating that it follows second-order kinetics[17].

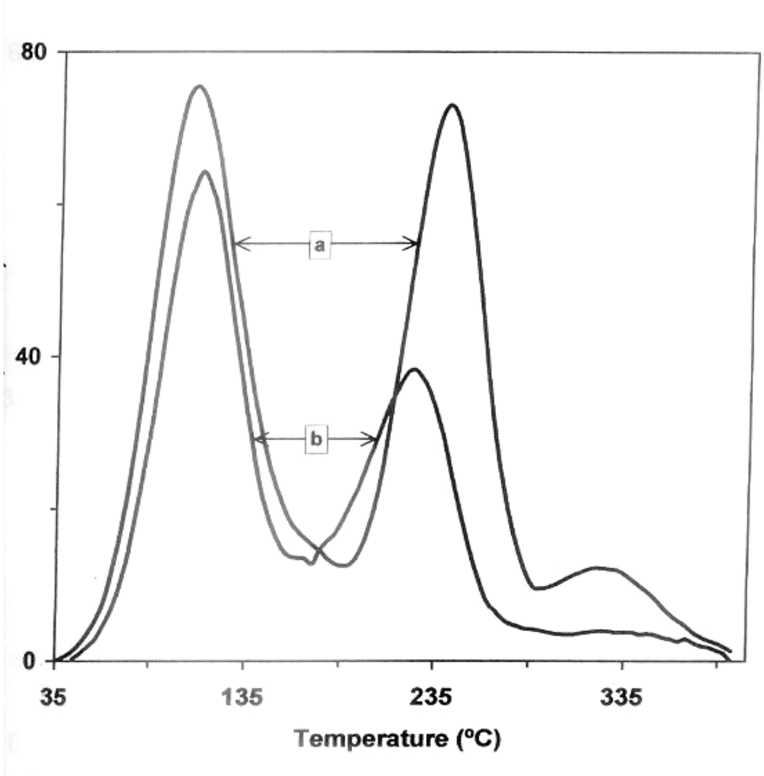

Fig.1: Glow curve of chert (a) NTL + 15 min γ-radiation (b) 6- min laser bleached and 2-hrs γ-radiation.

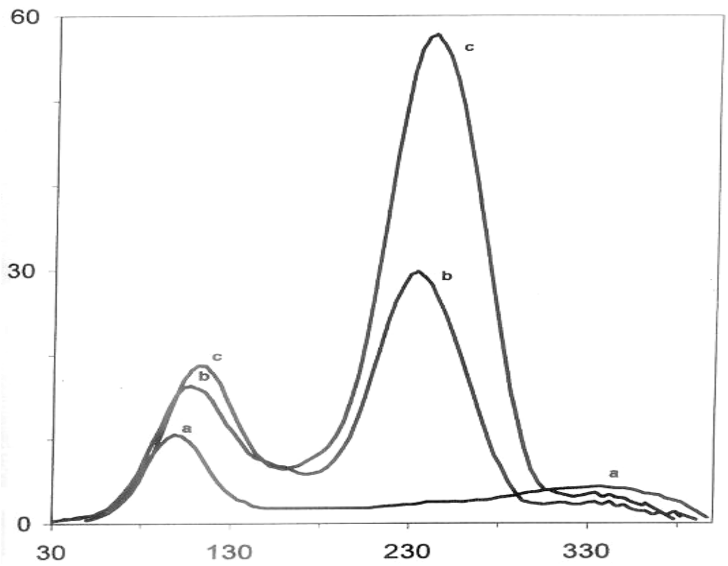

Fig.2: TL Glow curve of γ-irradiated (15min) chert with preheat treated at different Temperatures. Curves a, b and c are for preheating at 300, 500 and 700°C (β= 3.38 °C /Sec)

In order to substantiate these data we have subjected the glow curve obtained using a preheat of 220°C for 5 min [7], to deconvolution. Comparison of the values of activation energy (E) and frequency factor (s) of the two peaks thus obtained are in total disagreement with those obtained from the deconvolution of the natural TL curve of the same sample. Therefore, we have fitted the preheat treated TL curve with three peaks as was done for the natural TL. Thus, it is unlikely that preheat at 220°C for 5 min., erases the 280°C completely. This fact is supported by the fact that figure of merit (FOM) [18], with three peaks systems are similar for both the natural TL as well as the heat treated natural TL, while that for the two peaks systems is much higher. Incidentally, the figure of merit (FOM) has become not only popular but also acceptable by the community of workers, where FOM values of a few percent are taken as an acceptable fit [19].In agreement with the conclusion of Prokein and Wagner, the present rigorous curve fitting demonstrates non-first order kinetics for all the natural TL peaks of quartz relevant to dating. However, unlike Prokein and Wagner, who infer the predominance of second order kinetics, our data show the order of kinetics to be ≈ 1.5.The values of E ands, obtained by Prokein and Wagner, for the 280^{o}C peak by initial rise (IR) and peak shift with heating rate (PS), are in very good agreement with the present data, which confirms the theoretical prediction of Gartia et al [20] and Singh et al [21].

Table.1: Peak temperature T_m , heating rate (β), shape factor (μ_g) and trapping parameters of the 100^{o}C peak of Chert and Quartz.

Material	Specification	Expt. Parameters			Trapping parameters		
		T_m (^{o}C)	β (^{o}C/s	μ_g	E (eV)	s (sec^{-1})	b
Chert	NTL +15 min.γ-irradiated	105.0	3.37	0.57	0.814	1.5x10^{10}	2.00
	6min.Laser +2 hr γ–irradiated	109.7	3.37	0.52	0.817	1.2x10^{10}	2.00
	Preheat at 300oC +15 min.γ-irradiated	98.6	3.38	0.52	0.821	2.9x10^{10}	2.00
	Preheat at 500^{o}C +15min. γ–irradiated	106.6	3.38	0.59	0.827	2.0x10^{10}	2.00
	Preheat at 700^{o}C +15min.γ–irradiated	112.4	3.38	0.54	0.841	2.1x10^{10}	2.00
Quartz	Unfired	120	2.00	0.38	0.796	2.1x10^9	1.03
	Fired	96	2.00	0.42	0.846	3.0x10^{11}	1.13
	Unswept	97	2.00	0.39	0.822	2.3x10^{10}	1.01
	Swept	89	2.00	0.37	0.743	2.5x10^9	1.18

Table.2: Peak parameters of the 370^{o}C glow peak of flint.

Dose	I_m	T_m(^{o}C)	ω(^{o}C)	μ_g
60	5	367.69	67.07	0.527
120	12	364.31	70.77	0.526
240	28	361.54	70.46	0.519
480	51	356.92	70.46	0.511
960	82	352.31	73.23	0.516
1920	110	347.69	75.38	0.518

The values of the trap-depth (E) and frequency factor (S) obtained in the present study for 110,325 and 375^{o}C peaks are similar to the results of earlier workers.The present results has shown that the frequency factor (s) which is equally important in estimation of the lifetimes (τ) of the electrons in the trap is in the range $10^{11} \le s \le 10^{13}$, as expected to be an order of magnitude less than the vibrational frequency of the crystal namely ~10^{12} s^{-1}. The lifetime (τ)at 15 ^{o}C for 325^{o}C peak of the quartz is ≈10^7 years, while that for 370^{o}C peak is ≈10^8 years.

CONCLUSION

I). The natural TL peaks of quartz confirms the values of trapping parameters and electron lifetime obtained by earlier workers using conventional simple methods like initial-rise (IR), isothermal decay (ID) and peak shifting with heating rates (PS). However, a major conclusive finding is the fact that all the three natural TL peaks relevant to dating follow anon-first order kinetics with b≈1.5.

ii).The information provided in this paper in terms of the values of E, s and b are expected to help in deconvolution complex natural TL curves which will enable one to evaluate the areas of the individual glow peaks, a vital information for practical application of quartz in dating.

iii) The value of trapping parameters as established does not question to the utility of these peaks in TL dating. However, it has other implications especially in palaeodose plateau test for the two high temperature peaks since they follow non first-order kinetics.

iv) A 5 min pre-heat at 220^{o}C, a protocol followed to erase the 280^{o}C peak may not be a reality, which has significant implications in luminescence dating based on TL and OSL techniques.

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