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THERMAL QUENCHING OF THERMOLUMINESCENCE IN QUARTZ SAMPLES OF VARIOUS ORIGINS

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ABSTRACT

The effect of thermal quenching plays an important role in the thermoluminescence (TL) of quartz on which many applications of TL are based. In present work it is investigated that the thermal quenching parameters i.e. the activation energy W and the dimensionless parameter C, are more or less the same for every kind of quartzes or sample dependent on strong external treatment like a high temperature annealing. This preliminary investigation of seven quartz samples of different origin showed that the thermal quenching parameters W and C are common (universal) for most of the quartz samples.

KEYWORDS: Quartz, thermal quenching, efficiency, quenching parameters

INTRODUCTION

Quartz is one among the most widely deliberate naturally occurring minerals giving rise to luminescence, under both thermal and optical stimulation. The luminescence properties of natural quartz have attracted considerable experimental interest due to its extensive use in retrospective dosimetry and in archaeometry for dating ceramics and sediments.

These properties are almost exclusively investigated in specific quartz samples and then extrapolated them to any quartz sample. The reason is that the quartz included in the samples during the practical dating applications of ceramics, sediments etc, is completely unknown.

The validity of this crucial extrapolation has to be investigated experimentally. Towards this direction, an experimental way is to focus on thermal quenching parameters and to examine their properties if they are common or not to quartz samples of different origins. It is therefore a subject of great interest.

An attempt of such an investigation was reported by Pagonis et al. (2002), who had found that the "110^{0}C" TL peak exhibits certain properties such as the location of the maximum TL intensity, the activation energy E, the kinetic order b and the sensitization factor to small doses, which were proved to be universal, i.e. independent of the origin of the quartz samples.

intle (1975) measured the thermal quenching parameters of annealed natural quartz using radioluminescence as $C = 2.8 \times 107$ and $W = 0.64$ eV indicating that the quenching properties are independent of the wavelength of the observed luminescence, except at 495 nm. It was followed by McKeever et al. (1997) with $C = 7.9 \times 10^6$ and $W = 0.60$ eV by measuring the OSL intensity as a function of the sample temperature. Petrov and Bailiff (1997) evaluated these parameters for synthetic quartz and obtained $W = 0.78$ eV and $C = 3.1 \times 10^{10}$.

The values of these parameters were further studied by Kitis (2002). The aim of the present work is to investigate a universality of the thermal quenching parameters (W, C) seven unannealed and annealed at high temperatures.

EXPERIMENTAL

Seven sedimentary quartz samples of different origins were studied. Among them, three were collected from Northern Greece (Europe); these were given the laboratory code names Koupa Quartz, Kilkis (Kilk) quartz and Chalkidiki (Chalk) quartz. Quartz samples collected from USA and Norway (Europe) are named as pinky and grey quartz respectively. The remaining two samples were from Nepal (Asia). Laboratory code names reflect the respective origin, namely USA, Europe and Asia. All these quartz samples of various origins were smashed after scarping incrustation of dirt and finally sieved in order to get grains of size between 80-140 μm for TL measurements. All quartz samples were annealed at 900^{0}C for one hour and were followed by immediately fast cooled at RT. The reason of annealing was twofold; to simulate the conditions of pottery firing as well as to increase sensitivity of the samples studied (Liritzis, 1982). Reproducibility of ±5% was accomplished for each aliquot using mass weighting, while mass was selected as 6mg. Thermoluminescence signals of all quartz samples were obtained using the RISØ TL/OSL reader (model TL/OSL–DA–15) equipped with a 0.085Gy/s ^{90}Sr/^{90}Y β ray source (Bøtter-Jensen et.al, 2000). The reader was fitted with a 9635QA PM Tube. The detection optics consisted of a 7.5 mm Hoya U-340 filter (270-380nm, FWHM 80 nm). Several different heating rates were applied, ranging between 0.25 to16 ^{o}C/s.

RESULTS AND DISCUSSION

The experimental method to evaluate the thermal quenching parameters using TL measurements is to study the TL efficiency as a function of the readout heating rate. In the ideal case of absence of thermal quenching, the integrated area of the TL glow-peak has to be stable and independent of heating rate although the temperature corresponding to maximum intensity shifts. Therefore, any reduction of the TL output in terms of integrated signal due to heating rate is directly correlated with the thermal quenching effect. The thermal quenching efficiency versus temperature, η(T), is given by the following equation (McKeever1985, Petrov and Bailiff 1996,1997):

$$\eta(T) = \frac{1}{1 + C \cdot \exp(-\frac{W}{kT})} \quad (1)$$

The usual method to evaluate the (W, C) pairs is to fit the experimental TL peak integral as a function of peak maximum temperature, T_m according to the Equation (1). In order to obtain precise results, a heating rate is required which can be free from thermal quenching. In most cases, however, this requirement is not fulfilled. Furthermore, the shift of T_m as a function of the experimentally available heating rate covers no more than 50^0C in any case. So, the experimental values of η(T) in this short temperature region are not enough in order to perform a reliable fit using the previous equation for obtaining W and C. For these reasons in the present paper another procedure is followed to study the universality of the thermal quenching parameters W and C. This procedure has the following steps:

Table 1: Results showing the thermal quenching parameters followed by individual TL glow-peaks of various quartz samples. Where PB are the thermal quenching parameters of Petrov and Bailif, 1996 and W that of Wintle, 1975.

Name of quartz	T_m (K)	(W,C)	Figure
Koupa	325 402, 455	PB W W	1B
Pinky	336, 402 455	PB W W	2B
Grey	344 410 467	PB W W	3B
Nepalian B-2	306 339 368 432	PB W W W	4B
Nepalian A-2	334 442 571	PB W W	5B
Kilk	338 406 466	W W W	6B
Sedimentary	337 404 452	W W W	7B

Step 1: We consider that the thermal quenching parameters given in literature are quite universal. Among these, the most widely used parameters are suggested by Wintle (1975), W =0.64eV and C=2.8x10^7 as well as those reported by Petrov and Bailiff, (1996), W =0.74eV and C=3.1x10^7. These are also adopted in the present work.

Step 2: Using the above values of the thermal quenching parameters, we plot the thermal quenching efficiency versus T.

Step 3: Evaluation of the peak maximum temperature, T_m corresponding to the lower heating rate, 0.25 ^{o}C/s for each TL peak.

Step 4: Estimation of the theoretical value of n(Tm) corresponding to the T_m yielded by the previous step.

Step 5: The evaluated η(T_m) value is attributed to the integral of the corresponding TL peak.

Step 6: Based on the normalization of step 5 the values of η(T_m) corresponding to the resulted integrals for the higher heating rates are evaluated.

Step 7: The values of η(T_m) obtained in step 6 are placed on the η(T) plot derived by the selected W, C pairs at the temperatures T corresponding to the T_m at each heating rate.

Step 8: In case the values of η(T_m) coincide with the values of the η(T) plot then the thermal quenching parameters of the under study quartz sample are those by which the η(T) plot is derived.

The application of the above discussed method to the experimental data gave very interesting results concerning thermal quenching parameters and their universality.

Figure 1 shows the results of Koupa quartz. The Figure 1(A) shows a selection of TL glow-curves after different heating rates gave the following results. The first TL peak at 336K indicated by the solid circles seems to follow the behavior of thermal quenching parameters of Petrov and Bailif represented by the solid line (a). On the other hand the TL peaks at 405K and 459K represented by the solid triangle and boxes correspondingly, seem to follow very well the thermal quenching parameters by Wintle, solid line (b).

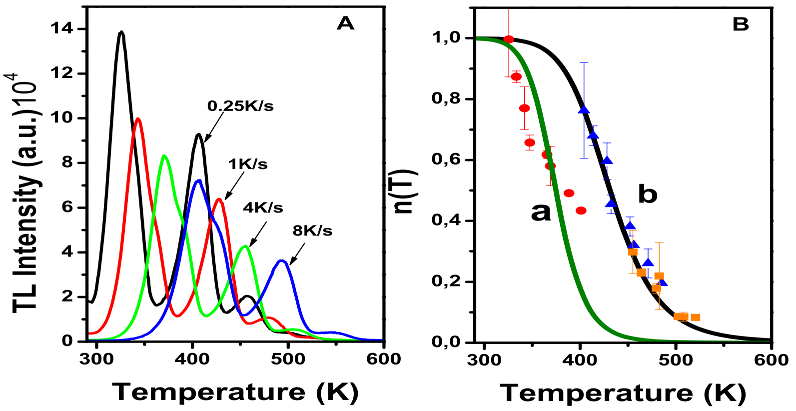

Figure 1: (A) TL glow-curves of the Koupa quartz for four different heating rates. (B) The solid line (a) is the theoretical thermal quenching efficiency curve evaluated using the thermal quenching parameters by Petrov and Bailiff (1996) and (b) the similar curve using the parameters by Wintle (1975). The symbols refer to experimental data of different peaks of different temperature.

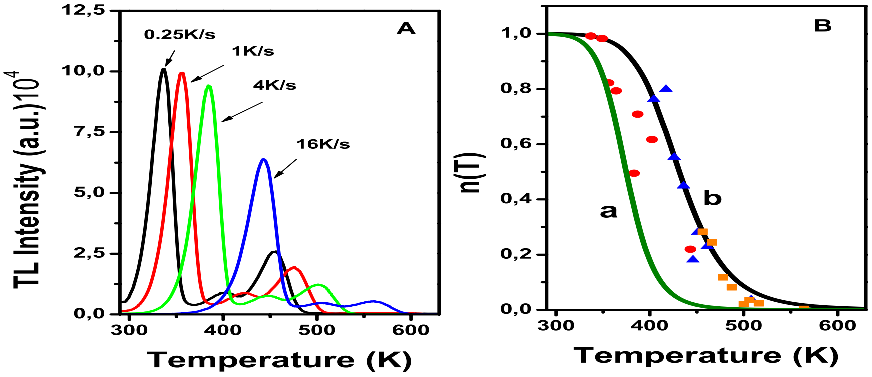

Figure 2: (A) TL glow-curves of the Pinky quartz for four different heating rates. (B) The solid line (a) is the theoretical thermal quenching efficiency curve evaluated using the thermal quenching parameters by Petrov and Bailiff (1996) and (b) the similar curve using the parameters by Wintle (1975). The solid circles are referred to the TL peak at 336K the solid triangles to TL peak at 405K and solid boxes to TL peak at 459K.

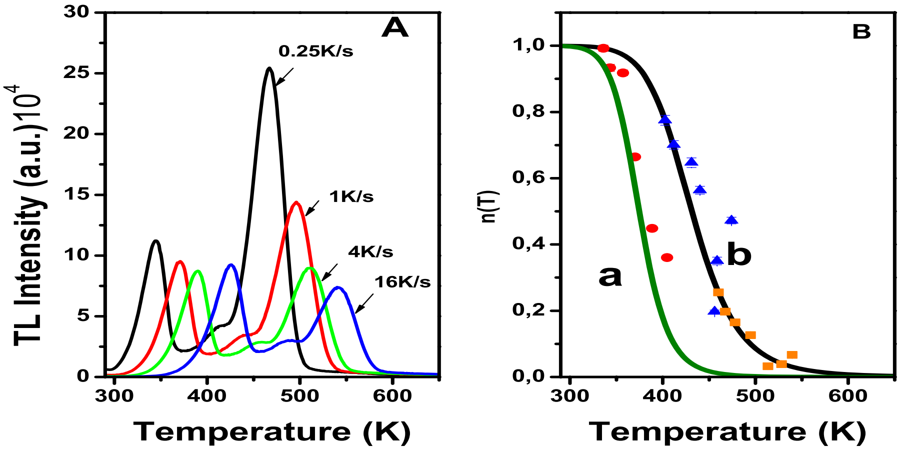

Figure 3: (A) TL glow-curves of the grey quartz for four different heating rates. (B) The solid line (a) is the theoretical thermal quenching efficiency curve evaluated using the thermal quenching parameters by Petrov and Bailiff (1996) and (b) the similar curve using the parameters by Wintle (1975). The solid circles are referred to the TL peak at 336K the solid triangles to TL peak at 405K and solid boxes to TL peak at 459K.

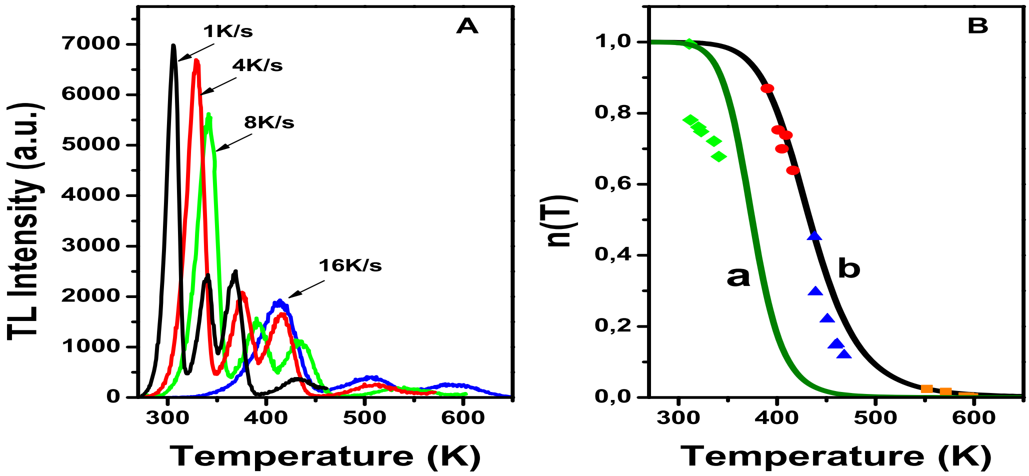

Figure 4: (A) TL glow-curves of the Nepalian B-2 quartz for four different heating rates. (B) The solid line (a) is the theoretical thermal quenching efficiency curve evaluated using the thermal quenching parameters by Petrov and Bailiff (1996) and (b) the similar curve using the parameters by Wintle (1975). The solid boxes are referred to the TL peak at 306K the solid circle, triangles and rectangle correspond to TL peak at 339K, 368K and 432K respectively.

In an exactly similar way Figures 2, 3, 4, 5, 6, and 7 describes the results for other quartz samples indicated on the figure captions. All the results are summarized in Table 1. It is indeed a fortunate surprise the way that the thermal quenching effect in individual TL glow-peaks of various quartz samples can be described by the two sets of values already existing in literature. TL glow peaks of all other quartzes except 110 °C seem to present a thermal quenching behavior very similar to the respective suggested by Wintle (1975). This is a quite interesting feature, especially due to the fact that this set of thermal quenching values was derived after studying natural TL signals. However, in the present study, only signals after artificial irradiations were used.

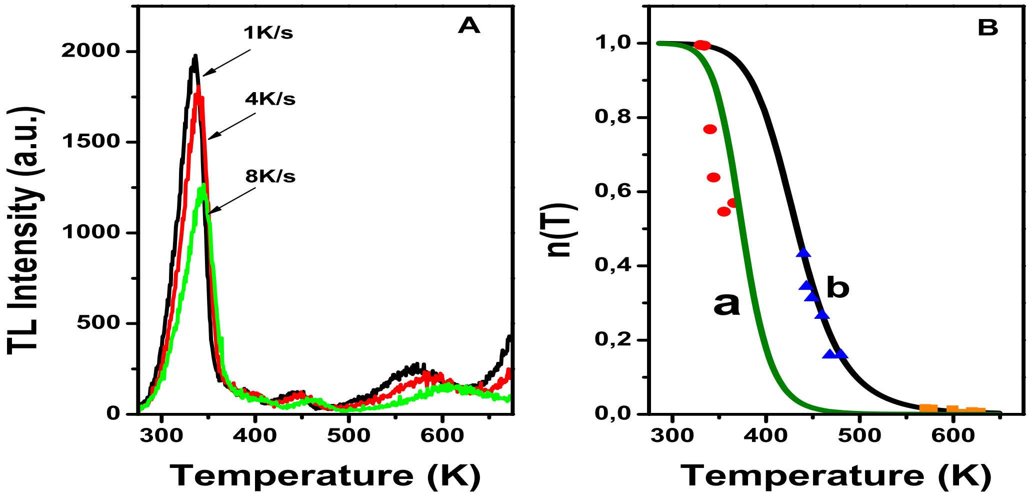

Figure 5: (A) TL glow-curves of the Nepalian quartz A-2 for four different heating rates. (B) The solid line (a) is the theoretical thermal quenching efficiency curve evaluated using the thermal quenching parameters by Petrov and Bailiff (1996) and (b) the similar curve using the parameters by Wintle (1975). The solid circles are referred to the TL peak at 336K, the solid triangles and boxes to TL peak 442K and 571K respectively.

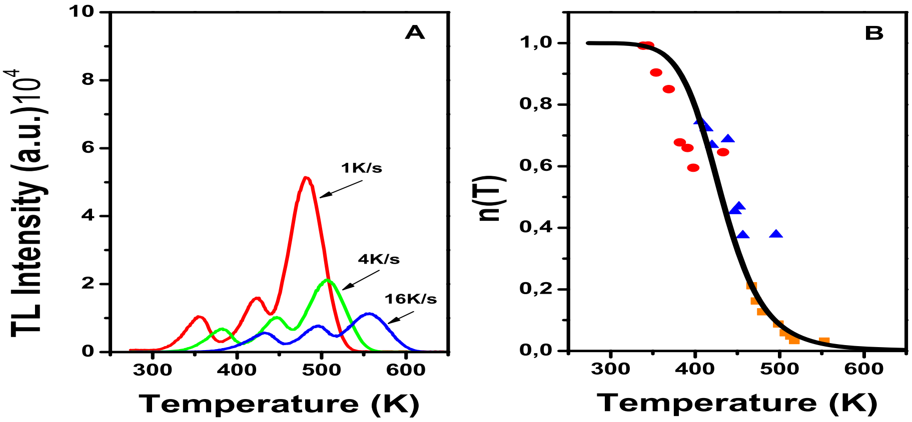

Figure 6: (A) TL glow-curves of the Kilk quartz for four different heating rates. (B) The solid line is the theoretical thermal quenching efficiency curve evaluated using the thermal quenching parameters by Wintle (1975). The solid circles, solid triangles, solid boxes are referred to the TL peak at 336K, 405K and 459K respectively.

In the case of the "110°C" TL peak in quartz, things are more complicated. For most quartz samples, the behavior of the latter peak does not seems to be in accordance with the behavior suggested by Wintle (1975), except the case of the two quartz samples termed as Kilk and Chalk. However, in the case of the other five samples, this behavior presents a tendency towards the result suggested by Petrov and Bailiff (1997), without being identical. Therefore, there is no indication of existence of one universal thermal quenching efficiency function in the case of "110 °C". Of course there is a need for further investigation of more quartz samples in order to have a clear vision concept. However, although the present results are limited to seven different quartz samples, they strongly support the existence of a kind of universality to the thermal quenching parameters in quartz annealed at 900°C.

CONCLUSIONS

The results of the present work show that the thermal quenching parameters for TL glow-peak at "110°C" are for the most of the samples those of Wintle (1975), except the synthetic quartz for which the thermal quenching parameters are that of Petrov and Bailiff (1996). The thermal quenching parameters of the TL glow-peaks beyond the "110°C" are all quartz samples those of Wintle (1975). Therefore, it is possible to argue a clear universality holds only for TL glow-peaks having their peak maximum temperatures beyond the TL glow-peak at "110°C".

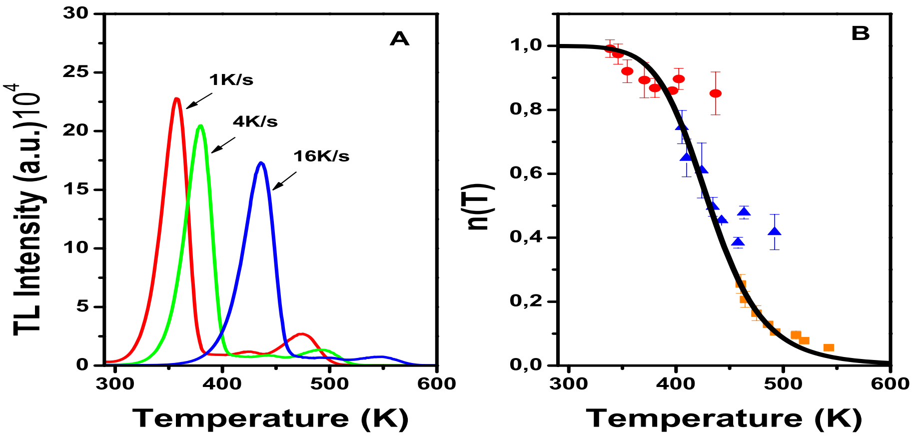

Figure 7: (A) TL glow-curves of the Chalk quartz for four different heating rates. (B) The solid line is the theoretical thermal quenching efficiency curve evaluated using the thermal quenching parameters by Wintel. The solid circles, solid triangles, solid boxes are referred to the TL peak at 336K 405K and 459K respectively.

REFERENCES

Akselrod M., Larsen N. Agersnap, Whitley V., and McKeever S.W.S., (1998). Thermal quenching of F-center luminescence in Al2O3:C, *J. Appl. Phys. 84, 3364*

Chen R.and Kirsh Y., *(1981).* Analysis of Thermally Stimulate Processes *(Pergamon Press. Oxford,).*

Curie D., *(1963)Luminescence in crystals* (Methuen, London).

Kitis G. (2002). Confirmation of the Influence of Thermal Quenching on the Initial Rise Method in a-Al_2O_3 :C, *phys. stat. sol.* (a) 191, No. 2, 621–627.

Liritzis, Y. (1982). Non-linear response of quartz grains: some annealing experiments PACT 6 209-213.

McKeever S.W.S., (1985). Thermolummescence of Solids *(Cambridge University Press, Cambridge).*

McKeever S.W.S., Bøtter-Jensen L., Larsen, N.A., Duller, G.A.T. 1997. Temperature dependence of OSL de cay curves: Experimental and theoretical aspects, *Radiation Measurement*, Vol 27, Issue 2, 161-170.

Nanjundaswamy R., Lepper K. and. McKeever S.W.S (2002). Thermal quenching of thermoluminescence in natural quartz, *Radiation Protection Dosimetry,* Vol. 100, No 1-4, 305–308.

Petrov S.A., Bailiff I.K., (1996). Thermal quenching and the Initial Rise technique of trap depth evaluation, *Journal of Luminescence* 65, 289-291.

Petrov S.A. and Baliff I.K., (1997). Determination of trap depths associated with TL peaks in synthetic quartz (350-550 K), *Radiation Measurements*, Vol. 27, No. 2, 185-191.

Pagonis V.,Tatsis E.,Kitis G., Drupieki C., (2002). Search for common characteristics in the glow curves of quartz of various origins, *Radiation Protection Dosimetry* Vol.100, No1-4, 377-376

Rinkevicius V. and Kavaliauskiene G.,(1994). Thermal quenching of thermally stimulated conductivity, *J. Phys. D: Appl. Phys*. 27, p1267-1271.

Wintle A.G.(1975) Thermal Quenching of Thermoluminescence in Quartz, *Geophys. J. Roy. Astronom. Sot*. 41, 107-113.

Wintle A.G. and Murray A.S., (1998). Towards the development of a preheat procedure for OSL dating of quartz, *Radiation Measurement* 29, 81-94.