



Reconstruction of thermally quenched glow curves in quartz

Bhagawan Subedi^{a,*}, George S. Polymeris^{b,c}, Nestor C. Tsirliganis^c, Vasilis Pagonis^d, George Kitis^a

^a Nuclear Physics Laboratory, Physics Department, Aristotle University of Thessaloniki, GR-54124 Thessaloniki, Greece

^b İŞIK University, Physics Department, Faculty of Science and Arts, 34980 Sile, Istanbul, Turkey

^c Laboratory of Radiation Applications and Archaeological Dating, Department of Archaeometry and Physicochemical Measurements, Cultural and Educational Technology Institute (C.E.T.I.), R.C. "Athena", Tsimiski 58, GR-67100 Xanthi, Greece

^d McDaniel College, Physics Department, Westminster, MD 21157, USA

ARTICLE INFO

Article history:

Received 26 June 2011

Received in revised form

29 November 2011

Accepted 29 January 2012

Keywords:

Quartz

Thermoluminescence

Thermal quenching

Dating

ABSTRACT

The experimentally measured thermoluminescence (TL) glow curves of quartz samples are influenced by the presence of the thermal quenching effect, which involves a variation of the luminescence efficiency as a function of temperature. The real shape of the thermally unquenched TL glow curves is completely unknown. In the present work an attempt is made to reconstruct these unquenched glow curves from the quenched experimental data, and for two different types of quartz samples. The reconstruction is based on the values of the thermal quenching parameter W (activation energy) and C (a dimensionless constant), which are known from recent experimental work on these two samples. A computerized glow-curve deconvolution (CGCD) analysis was performed twice for both the reconstructed and the experimental TL glow curves. Special attention was paid to check for consistency between the results of these two independent CGCD analyses. The investigation showed that the reconstruction attempt was successful, and it is concluded that the analysis of reconstructed TL glow curves can provide improved values of the kinetic parameters E , s for the glow peaks of quartz. This also leads to a better evaluation of the half-lives of electron trapping levels used for dosimetry and luminescence dating.

© 2012 Published by Elsevier Ltd.

1. Introduction

It is widely accepted in literature that the luminescence efficiency of most quartz samples is strongly influenced by the effect of thermal quenching (Wintle, 1975; McKeever, 1985). This effect causes a drastic decrease of the luminescence efficiency as a function of temperature, following the equation (Akselrod et al., 1998):

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

where W (eV) is the activation energy for the thermal quenching process, C is a dimensionless constant, k (eV/K) is the Boltzmann constant, and T (K) represents the temperature of the sample.

The presence of thermal quenching has a major impact not only on the intensity of the thermoluminescence (TL) signal, but also on the structure of the entire glow curve (Chen and McKeever, 1997; Kitis, 2002). These effects also influence the methods for

evaluating the kinetic parameters of the TL process, namely the activation energy E and the frequency factor s (Chen, 1969; Kitis and Pagonis, 2007; Chen and McKeever, 1997; Nanjundaswamy et al., 2002).

Recently Subedi et al. (2010) performed a detailed simulation of the influence of thermal quenching on the shape and on the evaluation of kinetic parameters E , s . These authors verified the drastic impact of this effect on the TL glow curves. In the case of quartz this effect influences strongly the high temperature TL glow peaks which are used in TL dating. This sheds some ambiguity about the validity of the presently published values of the kinetic parameters. One of the goals of this paper is to obtain an improved set of kinetic parameters for TL peaks in quartz, and to compare these with reported values in the literature.

Due to the importance of this effect on TL studies of quartz, there have been extensive efforts for evaluating the values of the thermal quenching parameters W and C (Wintle, 1975; Petrov and Bailiff, 1997; Murray and Wintle, 1997; Schilles et al., 2001; Chithambo, 2003). Recently, Subedi et al. (2011) showed that the values of the thermal quenching parameters yielded are common to different types of quartz samples subsequent annealing at 900 °C for samples of different origin. This is a very interesting and useful result, because it shows that quartz can be a reliable natural

* Corresponding author. Tel./fax: +30 2310998175.

E-mail addresses: bhsubedi@hotmail.com, bsubedi@physics.auth.gr (B. Subedi).

dosimeter, and that one can use the values of the thermal quenching parameters C , W universally with confidence, especially after heating as in the case of pottery.

All experimental TL glow curves of quartz samples are highly perturbed by the presence of thermal quenching. The quenched experimental TL intensity is given empirically by an equation of the form

$$I_{\text{quenched}} = I_{\text{unquenched}} \cdot \eta(T) \quad (2)$$

where $\eta(T)$ is the temperature-dependent luminescence efficiency in Eq. (1). Therefore, the *real* or *unquenched* TL luminescence intensity $I_{\text{unquenched}}$ of the quartz sample is unknown. This intensity can be evaluated by reversing Eq. (2) i.e.

$$I_{\text{unquenched}} = \frac{I_{\text{quenched}}}{\eta(T)} \quad (3)$$

In this paper, the procedure described by Eq. (3) is termed as *reconstruction of the quenched TL glow curve*. Starting with the experimentally determined intensity I_{quenched} and the experimentally determined parameters W , C we reconstruct the unquenched intensity $I_{\text{unquenched}}$ using Eq. (3). Once the reconstructed TL glow curve is obtained, it is possible to apply a computerized glow-curve deconvolution (CGCD) analysis and to evaluate the kinetic parameters. This reconstruction procedure was applied successfully by Dallas et al. (2008) to the experimentally obtained thermally quenched TL glow curve of $\text{Al}_2\text{O}_3\text{:C}$. Mandowski et al. (2010) also presented a method to reconstruct the unquenched glow curve from the data of quenched glow curves. Their data was obtained from heating rate measurements, and by evaluating the pair of thermal quenching parameters W , C using a Monte-Carlo algorithm.

The aim of the present work is three-fold:

- To perform a reconstruction to the experimental, quenched TL glow curves of quartz, and to evaluate the kinetic parameters using a CGCD analysis of this reconstructed TL glow curve.
- To perform a separate and independent CGCD analysis of the experimental (thermally quenched) TL glow curve and,
- To carry out a comparison of the results obtained in (a) and (b) with each other, and also with several reported values of the kinetic parameters found in the literature.

2. Experimental

The present study is mainly based on two quartz samples: firstly, a Nepalese quartz sample (laboratory code B2), and secondly

a quartz sample from northern Greece (laboratory code Kilkis quartz). Both of these quartz samples were gently crushed and sieved in order to obtain grains of size 80–140 μm . The quartz samples were annealed at 900 °C (1173 K) for 1 h prior to the TL measurements. The Kilkis sample was slow cooled to room temperature, while the B2 quartz sample was cooled to room temperature quickly, after taking out from furnace. However, the cooling rate affects just the sensitivity of the samples but not the number of peaks.

The criteria for selecting these two samples were the different behavior of their thermal quenching parameters C , W . Subedi et al. (2011) showed that all glow peaks in the TL glow curves of the Kilkis sample can be described by the same C and W values. However, the B2 quartz was shown to be described by two different set of parameters C , W . The first set of parameters described thermal quenching effect for the TL glow peak at “110 °C”, while a different second set of C , W values was found to describe thermal quenching for the high temperature glow peaks in this sample.

The experimental protocol used for B2 quartz was:

- Step 1 Deliver test dose D_i
- Step 2 Measure the TL glow curve up to a temperature T_i , to remove the low temperature peaks. The temperature T_i will depend on the heating rate, with the position of the peaks shifting as a function of the heating rate.
- Step 3 Measure the TL glow curve up to 500 °C at 0.5 °C/s, in order to obtain the TL glow curve.
- Step 4 Measure the TL glow curve up to 500 °C at 0.5 °C/s, to obtain the respective background.
- Step 5 Repeat steps 1–3 for a new sample and a new test dose D_i .
- Step 6 Repeat steps 1–4 for heating rates of 1 and 5 °C/s.

For the case of Kilkis quartz, all glow peaks of its glow curve obey to the same thermal quenching parameters, so that step 2 was not included.

The measurements were performed with dose 75 Gy. The reason for measuring the TL glow curves at such low heating rates was in order to avoid temperature lag effects, and to better measure the high temperature peaks for which the thermal quenching is expected to have the most effect (Kitis and Tuyn, 1998). Furthermore, three different heating rates were applied in order to check the reproducibility of each glow curve integral subsequent reconstruction. The sample mass was selected as 4 mg, deposited on stainless steel disks of 1 cm^2 area. TL signals of both quartz samples were obtained using the RISØ TL/OSL reader (model TL/OSL-DA-15) equipped with a 0.085 Gy/s $^{90}\text{Sr}/^{90}\text{Y}$ β ray source (Bøtter-Jensen

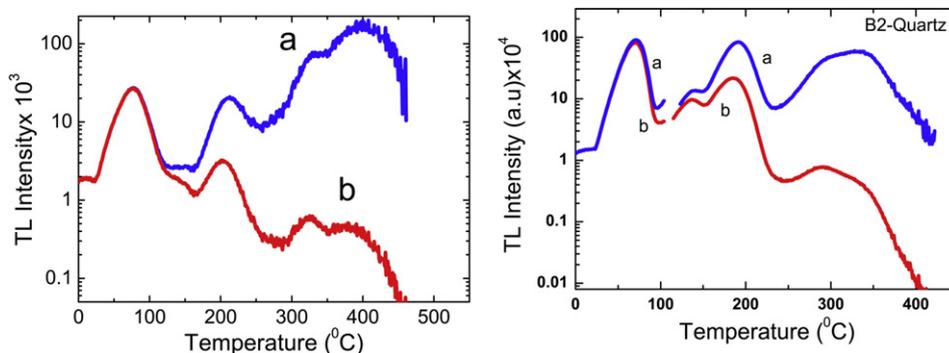


Fig. 1. Examples of experimental (quenched) and reconstructed (unquenched) glow curves obtained at a heating rate of 0.5 K/s. Curve (a) is the reconstructed glow curve, and curve (b) is the experimental glow curve. Left plot: Kilkis quartz, the glow curve (a) is reconstructed by using thermal quenching parameters reported by Wintle (1975). Right plot: B2 quartz, the curve (a) is reconstructed using two different pairs of thermal quenching parameters. The 110 °C peak is reconstructed by using parameters reported by Petrov and Bailiff (1997). The remaining part of the glow curve is reconstructed by using the parameters of Wintle (1975). The break in the glow curve is due to separate measurements of the 110 °C peak and the high temperature peaks.

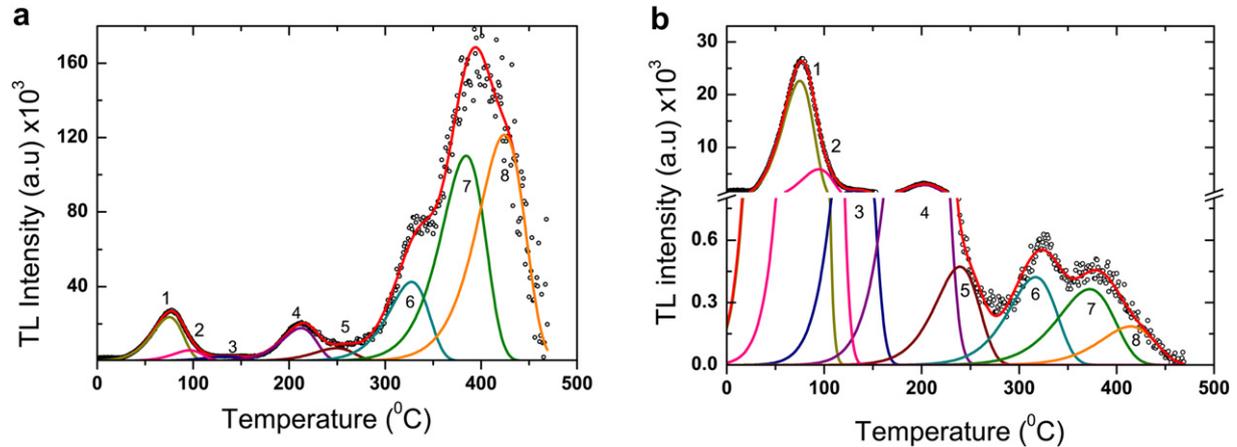


Fig. 2. Left plot (a): reconstructed (unquenched) glow curve of Kilikis quartz, showing all component peaks obtained from CGCD analysis. Among them 1, 2, 4, 6 and 7 are prominent peaks. Right plot (b): CGCD of experimentally obtained glow curve of Kilikis quartz. The vertical scale contains a break in order to show the detailed positions of the quenched high temperature glow peaks in the glow curve. The peak labeling in both plots is the same.

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 (FWHM 80 nm).

3. Methods of analysis

The method of analysis consists of the following steps:

3.1. Reconstruction of the TL glow curves

For reconstruction of the TL glow curves, Eq. (3) was used. The thermal quenching efficiency $\eta(T)$ was evaluated using the following W and C pairs. In the case of Kilikis quartz $W = 0.64$ eV and $C = 2.7 \times 10^7$ (Wintle, 1975) were used for the entire TL glow curve. In case of B2 quartz we used the values $W = 0.78$ eV and $C = 3.1 \times 10^{10}$ for the “110 °C” TL glow peak (Petrov and Bailiff, 1997), while for the higher temperature glow peaks we used the values $W = 0.64$ eV and $C = 2.7 \times 10^7$ (Wintle, 1975). For a complete discussion of the evaluation of these kinetic

parameters, the reader is referred to the recent experimental work of Subedi et al. (2011).

3.2. Computerized glow-curve deconvolution (CGCD)

The application of the CGCD analysis on thermally quenched glow curves will most likely produce incorrect values of the kinetic parameters E , s . The reason is that the usual TL expressions hold only when the luminescence efficiency is constant throughout the TL glow curve, and not temperature dependent as in Eq. (1). In practice, the thermal quenching effect can alter the shape of a TL glow peak, so one has to investigate whether the thermally quenched TL glow peak can be described anymore by the usual TL expressions. This topic was examined, discussed and tested in detail in the simulation study of Subedi et al. (2010). The present work represents an extension of this previous simulation work, to actual experimental TL glow curves.

Both the reconstructed and experimental TL glow curves were analyzed using a CGCD analysis procedure using the TL equation of general order kinetics as proposed by Kitis et al. (1998):

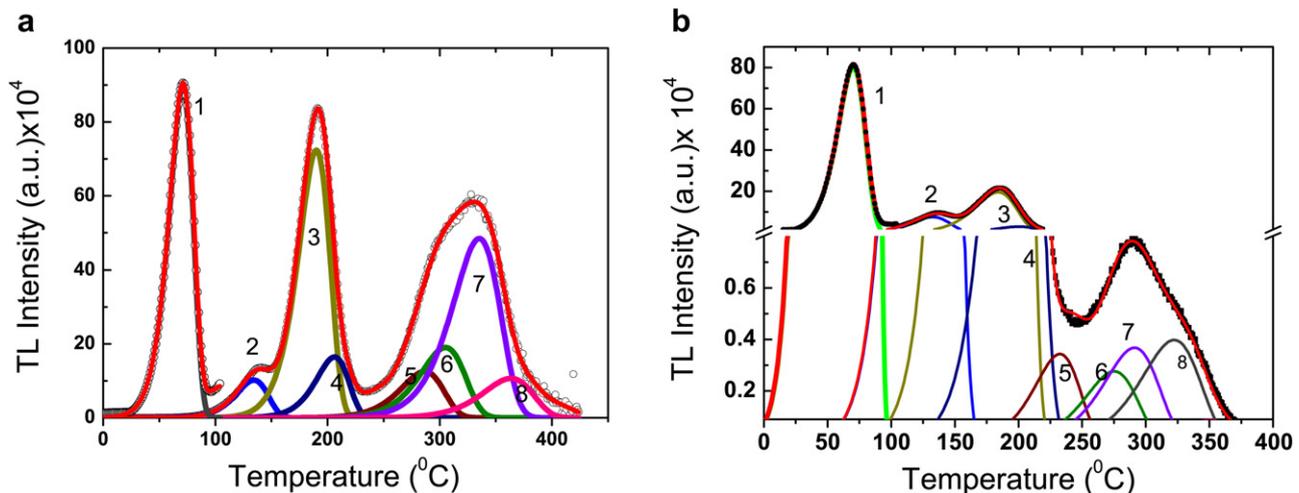


Fig. 3. Left plot (a): reconstructed glow curve of B2 quartz showing all component peaks obtained from CGCD analysis. Among eight different peaks, 1, 2, 3, 4, 6 and 7 are prominent peaks. The 110 °C peak (1) was measured separately and is combined with high temperature peaks, to represent the real shape of the complete glow curve. Right plot (b): CGCD of experimentally obtained glow curve of B2 quartz. The peaks with numbers 1–8 are eight different component peaks. The vertical scale contains a break, in order to show the higher temperature peaks with very low intensity. The labeling scheme of the peaks appearing in both panels is the same.

Table 1

Position of peak maxima and estimations of activation energies, frequency factors (s^{-1}) and half lives $T_{1/2}$ of TL peaks at room temperature (RT) for both Kilkis and B2 quartz samples in the case of experimental and reconstructed (unquenched) glow curves using the heating rate of 0.5 °C/s.

Quartzes	Experimental (quenched) glow curves				Reconstructed (unquenched) glow curves			
	T_m (°C)	E (eV)	Frequency factor s	$T_{1/2}$ (in yrs.)	T_m (°C)	E (eV)	Frequency factor s	$T_{1/2}$ (in yrs.)
Kilkis quartz	75	0.64	5.6×10^8	1.7×10^{-4}	75	0.64	7.3×10^8	2.1×10^{-4}
	203	0.91	1.0×10^8	0.61	212	1.15	2.5×10^{10}	26.17
	317	1.18	2.4×10^8	8940	327	1.46	4.3×10^{10}	2.4×10^6
	372	1.20	3.9×10^7	115,004	384	1.57	2.33×10^{10}	3.2×10^8
	414	1.41	3.8×10^8	4.0×10^7	424	1.67	2.29×10^{10}	1.5×10^{10}
B2 quartz	71	0.91	8.3×10^8	3.9×10^{-4}	71	0.91	8.7×10^8	4.1×10^{-4}
	134	0.96	6.3×10^9	1.2	134	0.97	6.9×10^9	1.8
	200	1.19	6.2×10^{10}	21	206	1.33	1.12×10^{13}	210
	275	1.24	5.4×10^9	3542	286	1.39	8.9×10^{10}	81,087
	295	1.25	2.8×10^9	11,422	305	1.42	5.7×10^{10}	382,894
	326	1.27	9.0×10^8	68,986	346	1.47	1.8×10^{10}	7.5×10^6

$$I(T) = I_m \cdot b^{\frac{b}{b-1}} \cdot \exp\left(\frac{E}{kT} \cdot \frac{T - T_m}{T_m}\right) \times \left[(b-1) \cdot (1-\Delta) \cdot \frac{T^2}{T_m^2} \cdot \exp\left(\frac{E}{kT} \cdot \frac{T - T_m}{T_m}\right) + Z_m \right]^{-\frac{b}{b-1}} \quad (4)$$

with $\Delta = 2kT/E$, $\Delta_m = 2kT_m/E$, $Z_m = 1 + (b-1) \cdot \Delta_m$, where I the TL intensity, E (eV) the activation energy, I_m the peak maximum intensity, T_m (K) the peak maximum temperature, k the Boltzmann constant and T the absolute temperature.

All glow peaks were considered to follow first order kinetics and this was achieved by using $b = 1.0001$. The goodness of fit was tested by the Figure of Merit (FOM) by Balian and Eddy (1977) which can be expressed as

$$FOM = \sum \frac{|Y_{Expt} - Y_{Fit}|}{A} \quad (5)$$

where Y_{Expt} is the experimental glow curve, Y_{Fit} is the fitted glow curve, and A is the area of the fitted glow curve. All deconvoluted glow curves for all heating rates used were accepted if their FOM values were lower than 3%.

In the present work the background signal obtained in Step 4 of the experimental protocol was subtracted from the experimental TL glow curve. However, even after the subtraction, a positive residual background characterized by a strong scatter of the experimental points remained. Obviously during the reconstruction procedure, this highly scattered portion of the glow curves is reconstructed as well (see Fig. 1). This “background effect” was taken into consideration by including in the CGCD analysis a separate background function of the form:

$$Bg(T) = \frac{P}{\eta(T)} \quad (6)$$

where P is a constant which was left to vary during CGCD analysis.

3.3. Consistency of the CGCD analysis

The consistency of the CGCD analysis results between unquenched and quenched glow curves was tested as follows.

Step 1 Every unquenched TL glow curve resulting from the CGCD analysis is “re-quenched”, by applying Eq. (2).

Step 2 The TL glow peaks resulting by the “re-quenching” procedure in Step 1, must coincide with the individual TL glow peaks resulting from the independent

CGCD analysis of the experimental (quenched) TL glow curve.

4. Experimental results

4.1. Reconstruction of unquenched TL glow curves

Examples of reconstructed TL glow curves along with the experimental data are shown in Fig. 1. The right plot in Fig. 1 presents the results for B2 quartz, and the left plot presents the results for Kilkis quartz. Comparison of the unquenched and quenched TL glow curves in Fig. 1 yields the following observations:

1. In both plots of Fig. 1, the reconstructed glow curve reflects clearly the strong influence of thermal quenching on the shape

Table 2

Some values for trapping parameters of quartz TL peaks as reported by various authors.

Methods	Peaks tem. (°C)	E (eV)	Frequency factor (s^{-1})	References
ITD	237 ± 10	1.55	1.1×10^{14}	Petrov and Bailiff (1997)
	308	1.64	3.6×10^{14}	Prokein and Wagner (1994)
	325	1.70	1×10^{14}	Wintle (1975, 1997)
		1.60	5.7×10^{12}	Spooner and Questiaux (2000)
	326	1.71	1.0×10^{14}	Prokein and Wagner (1994) and references therein
	330	1.85	4.1×10^{14}	Prokein and Wagner (1994) and references therein
	333	1.71	$10^{13} \sim 10^{14}$	Prokein and Wagner (1994)
CF	180	1.19	1.8×10^{11}	Levy (1979)
	320	1.61	5.1×10^{11}	
IR	190	1.42	3.4×10^{14}	Aitken (1985)
	280	1.50	1.5×10^{13}	Spooner and Questiaux (2000)
	280	1.45	5.0×10^{12}	Hütt et al. (1979)
	310	1.68	1.8×10^{13}	Aitken (1985)
PH	220	1.38	7.7×10^{13}	Spooner and Questiaux (2000)
	308	1.44	6.1×10^{11}	Prokein and Wagner (1994)
	325	1.65	3.9×10^{13}	Spooner and Questiaux (2000)
PS	326	1.73	1.0×10^{14}	Prokein and Wagner (1994)
	327	1.74	1.4×10^{14}	Prokein and Wagner (1994)
CGCD	257	1.25	1.6×10^{11}	Kitis et al. (2002)
	288	1.30	9.1×10^{10}	
	322	1.33	3.2×10^{11}	
	360	1.40	2.3×10^{10}	

Note: Peak temperatures refer to the heating rate in the work concerned, usually 1–10 °C/s.

CF= Curve fitting, IR= Initial rise, ITD= Isothermal decay, PH= Peak shift with heating rate, PS= Peak shape, CGCD= Computerized glow curve deconvolution

- of the TL glow curves. This influence is present at all temperatures.
- The general information provided by Fig. 1 is in agreement with the results of the numerical simulation studies performed by Subedi et al. (2010). These simulation studies had focused on the influence of thermal quenching on the shape of the experimental TL glow curves. Consequently, as Subedi et al. (2010) have theoretically predicted for the case of first order of kinetics, both values of T_m and E for each peak of the quenched glow curve are lower than the corresponding values in the case of the unquenched glow curve. These findings were also experimentally verified.
 - However, the most important information given by the reconstructed glow curve is the complete and “real” shape of the unquenched TL glow curve, along with its large difference from the experimental, quenched one, especially in the high temperature region. To the best of our knowledge, this is the first such presentation of the “real” TL glow-curve shape of quartz samples.
 - Furthermore, these results provide the opportunity to evaluate and obtain improved values for the kinetic parameters for each individual TL glow peak of quartz, through a CGCD analysis. The results of such a double CGCD analysis are presented next, for both the experimental data and for the reconstructed “real” TL glow curves.

4.2. CGCD analysis of reconstructed and experimental TL glow curves

Examples of CGCD analysis for both experimental and reconstructed TL glow curves are shown in Fig. 2 for Kilkis quartz. The left plot of Fig. 2 shows an example of a reconstructed TL glow curve of Kilkis quartz, analyzed into its individual TL glow peaks. The right plot of Fig. 2 shows the corresponding experimental TL glow curves, which are of course thermally quenched. The shape of the reconstructed TL glow curve shown is different from that of the corresponding plot of Fig. 1, because in the case of Fig. 2 the reconstructed background signal has been subtracted. As seen in Fig. 2, the high temperature TL glow peaks used for dating are in fact mostly affected by the presence of thermal quenching.

Fig. 3 presents the same results for quartz sample B2. The difference between this quartz sample and Kilkis quartz is that the TL peak at “110 °C”, the TL peak around 200 °C as well as the high temperature TL glow peaks have more or less the same TL sensitivity when the thermal quenching effects is not present.

A selection for the TL glow-peak parameters resulting from the CGCD analysis is listed in Table 1, for both the experimental and the reconstructed TL glow curves, of Kilkis and B2 quartz samples.

Several useful conclusions can be drawn from these results. Firstly, the values of T_m and activation energy E for any individual TL glow peak are in full agreement with the prediction of the

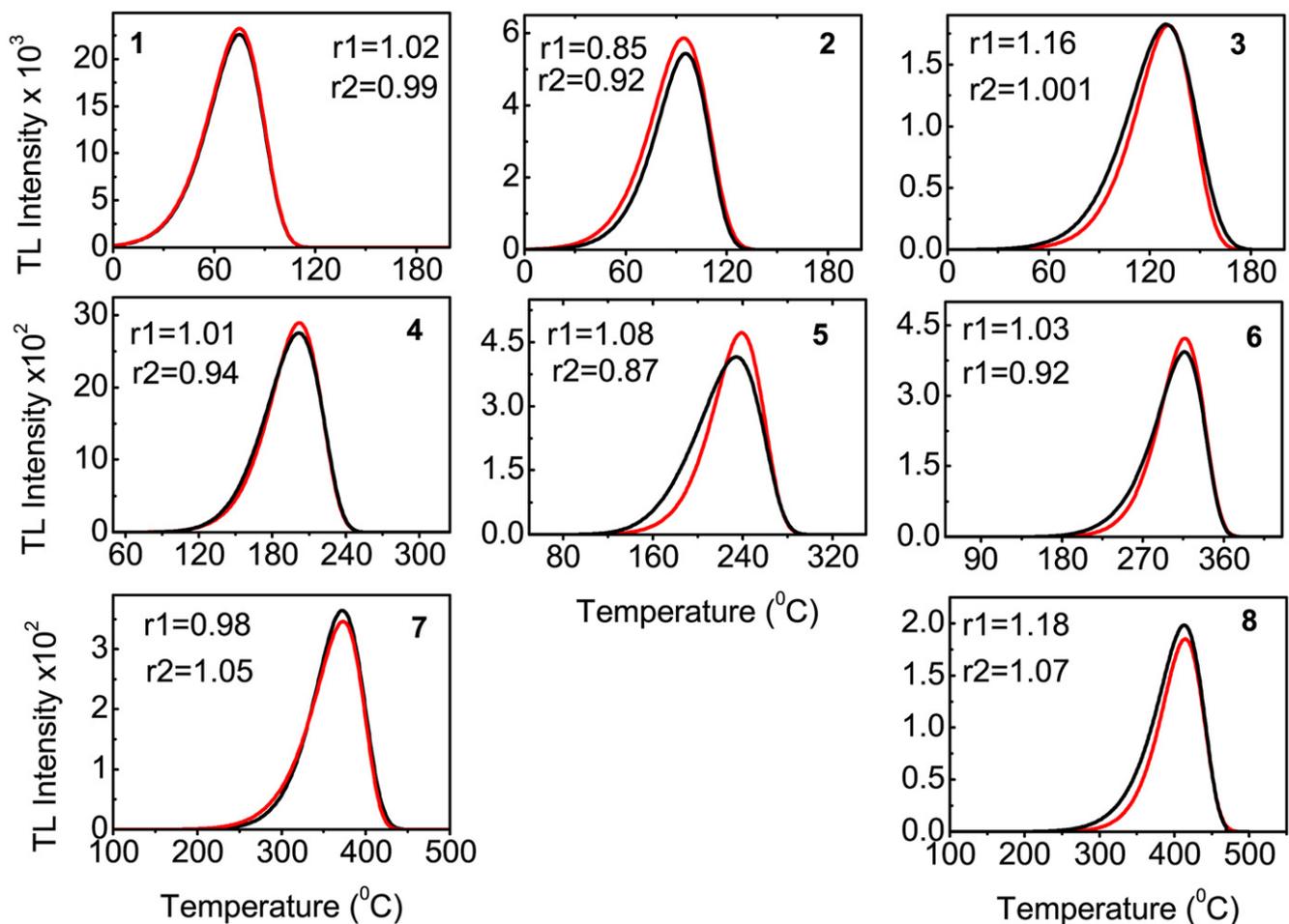


Fig. 4. Results of the double CGCD analysis described in the text, for the Kilkis quartz sample. Here r_1 is the ratio of glow peak areas of the re-quenched glow peak (black lined peak) and the experimental glow peak (red lined peak). r_2 is the ratio of maximum peak height of the re-quenched glow peaks from (black lined peak) and experimentally obtained glow peaks. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

simulation study by Subedi et al. (2010). Specifically, the values of the activation energies become consistently larger after the “real” TL glow curve is reconstructed. These values of E and s obtained from CGCD analysis must be compared with published values in the literature.

However, all methods of evaluating trapping parameters of glow curve are influenced by thermal quenching. The only method which is free from thermal quenching effects is arguably the isothermal decay method; since the temperature of the sample is kept fixed during isothermal measurements, thermal quenching effects should have no effect on the evaluated kinetic parameters.

The trapping parameters evaluated from the CCGD analysis of reconstructed TL glow curves of both quartz samples are listed in Table 1. These results concern the TL glow peaks with peak maximum temperature above 200 °C.

Table 2 presents the trapping parameters evaluated for a variety of quartz samples, by various methods and by several authors (Levy, 1979; Hütt et al., 1979; Prokein and Wagner, 1994; Spooner and Questiaux, 2000; Kitis et al., 2002). Unfortunately, the uncertainties associated with these parameters, and especially with the activation energy and frequency factors, were not quoted in these papers. Additionally, according to Chithambo and Ogundare (2009) the influence of irradiation, preheating, and annealing on the lifetimes can be explained with reference to an energy band scheme to

give luminescence emission which is responsible with the change in concentration of holes at various luminescence centers caused by annealing.

Comparison of Tables 1 and 2 leads to the following conclusions:

1. For each quartz sample, the activation energies of the thermally quenched TL glow curves are enhanced after the reconstruction procedure.
2. The values of activation energies E of quenched glow peaks with T_m larger than 250 °C evaluated in this study, match the activation energies evaluated by Pagonis et al. (2002) and Kitis et al. (2002) which were obtained from CGCD analysis of TL peaks. However, the activation energies obtained from reconstruction process described above are higher than the aforementioned literature values. In fact, the E values obtained from the reconstructed TL glow peaks are very close to the corresponding values listed by Spooner and Questiaux (2000); these authors used the isothermal decay method.
In addition, these E values are also with the acceptable range of values and errors reported by Prokein and Wagner (1994), and the references therein.
3. The values of the activation energies E obtained from the reconstructed glow peaks are consistent with E values obtained from the variable heating rate methods. However, Subedi et al. (2010) argued that the various heating rates method gives

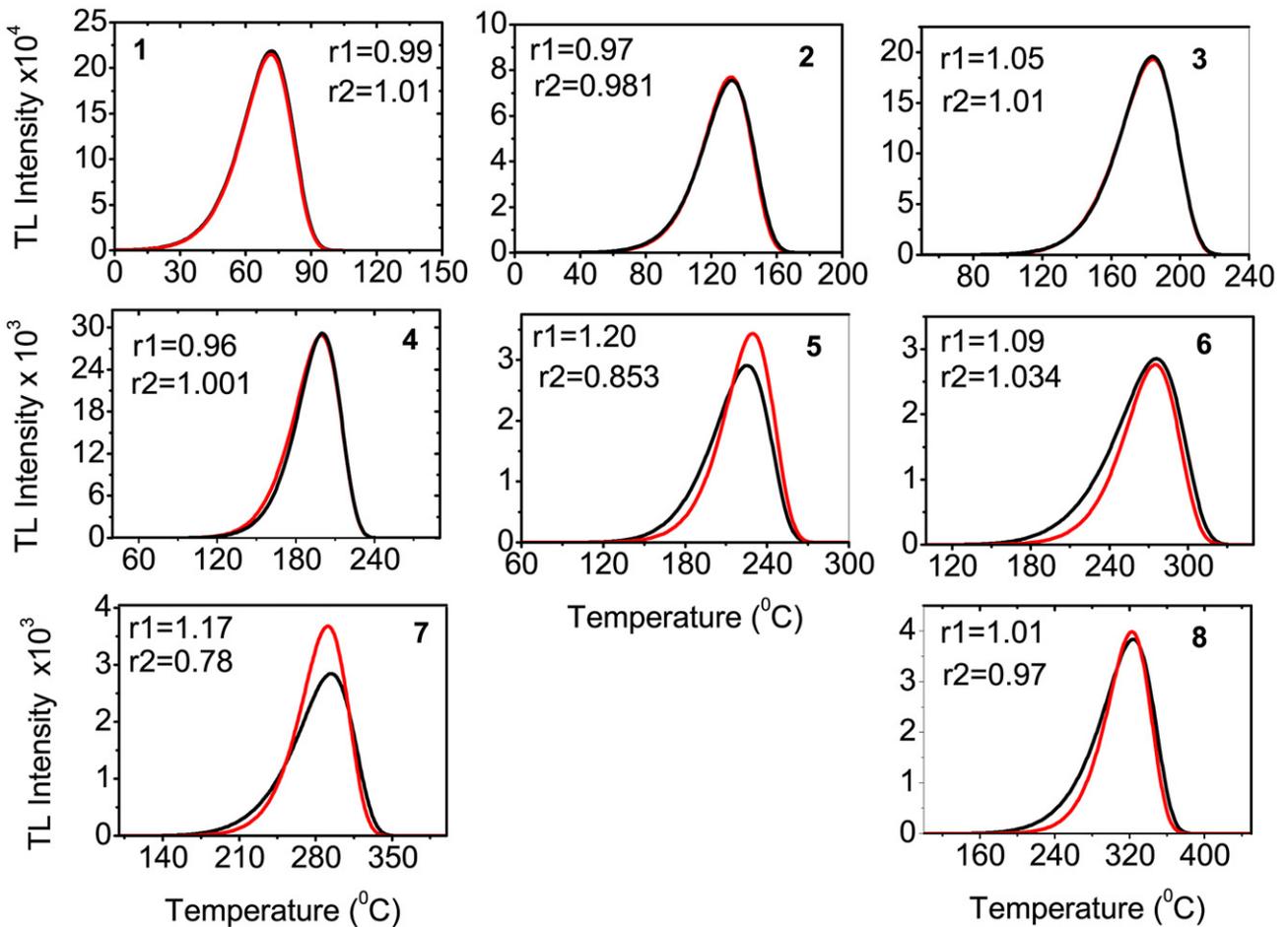


Fig. 5. Results of the double CGCD analysis described in the text, for the B2 quartz sample. Here r_1 is the ratio of glow peak areas of the re-quenched glow peak (black lined peak) and the experimental glow peak (red lined peak). r_2 is the ratio of maximum peak height of the re-quenched glow peaks from (black lined peak) and experimentally obtained glow peaks. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

reliable E values only in the case of narrow TL peaks; in the case of wide TL peaks, this method can overestimate the values of activation energy.

- The values of activation energies reported by Spooner and Questiaux (2000) and Prokein and Wagner (1994) using various heating rates are higher than the E values obtained for the experimental glow peaks with T_m larger than 250 °C, but are very close to those obtained from the reconstructed TL glow peaks. In some cases, the activation energies from the reconstructed TL glow curves are higher than the values quoted in the aforementioned papers in the literature.
- The E values evaluated by Levy (1979) using a curve fitting method are also in agreement with the values obtained in this paper using the reconstructed TL glow curve.
- As in the case of activation energies, the evaluated frequency factors s are enhanced subsequent the reconstruction procedure. Moreover, these evaluated frequency factors are also within the acceptable range of values obtained from theoretical considerations. According to Chen and Kirsh (1981), typical values of frequency factor s (in s^{-1}) can be expected to range from 10^{11} to $10^{15} s^{-1}$.

However, the most important conclusion from this study concerns the half-lives of electron traps obtained from CGCD analysis of the experimental and the reconstructed TL glow curves. These values are listed in Table 1. The clear conclusion from Table 1 is that the stability of the electron level responsible for the TL glow peaks used in dating is much higher than the value predicted from analysis of the experimental TL glow curve alone.

4.3. Consistency between reconstructed and experimental glow curves

The consistency between the experimental TL glow curve and its reconstructed version was checked according to the steps outlined in Section 3.3. An one-to-one comparison was carried out between the individual TL glow peaks obtained from CGCD analysis of the experimental glow curve, and the individual peaks of the reconstructed TL glow curve. The results are shown in Figs. 4 and 5, for Kilkis and B2 quartz correspondingly. In these figures one can see each pair of the various TL peaks plotted against together. The agreement concerning the ratios of the peak heights (r_1), and the ratios of the total areas (r_2) is very satisfactory.

The activation energies of each individual re-quenched TL glow peak were evaluated using the peak shape methods (Chen, 1969;

Kitis and Pagonis, 2007). The resulting E values are listed in Table 3, along with the activation energy values of the individual experimental TL glow peaks. The consistency in the peak position of glow curves is very satisfactory, within a small range of ± 3 °C. Similarly, the evaluated activation energies were also in good agreement with the results from the experimental glow curves. However, some small discrepancy between the activation energies appears, especially at high temperature peaks. This could be the result of very poor statistics for some of the re-quenched glow peaks; such peaks are difficult to resolve.

5. Conclusions

In this study, the unquenched TL glow curves of two different natural quartz samples were successfully reconstructed, by using previously experimentally determined sets of thermal quenching parameters.

The reconstruction of these “real” TL glow curves produced the unquenched shape of the TL glow curve. Furthermore, CGCD analysis of this reconstructed TL glow curve gave improved values of peak maximum positions, trapping parameters E and s , and half-lives of the traps.

By “re-quinching” the reconstructed TL glow curves and carrying out a CGCD analysis, it was found that the reconstructed TL glow curves are consistent with the independently analyzed experimental TL glow curves.

References

- Aitken, M.J., 1985. Thermoluminescence Dating. Academic Press, London.
- Akselrod, M., Agersnap, Larsen N., Whitley, V., McKeever, S.W.S., 1998. Thermal quenching of F-center luminescence in $Al_2O_3:C$. J. Appl. Phys. 84, 3364–3373.
- Balian, H.G., Eddy, N.W., 1977. Figure of merit (FOM), an improved criterion over the normalized chi-squared test for assessing goodness-of-fit of gamma ray spectra peaks. Nucl. Instr. Meth. 145, 389–395.
- Bøtter-Jensen, L., Bulur, E., Duller, G.A.T., Murray, A.S., 2000. Advances in luminescence instrument systems. Radiat. Meas. 32, 523–528.
- Chen, R., 1969. On the calculation of activation energies and frequency factors from glow curves. J. Appl. Phys. 40, 570–587.
- Chen, R., Kirsh, Y., 1981. Analysis of Thermally Stimulated Processes. Pergamon Press, London.
- Chen, R., McKeever, S.W.S., 1997. Theory of Thermoluminescence and Related Materials. World Scientific, Singapore.
- Chithambo, M.L., 2003. Dependence of the thermal influence on luminescence lifetimes from quartz on the duration of optical stimulation. Radiat. Meas. 37, 167–175.
- Chithambo, M.L., Ogundare, F.O., 2009. Luminescence lifetime components in quartz: influence of irradiation and annealing. Radiat. Meas. 44, 453–457.
- Dallas, G.I., Afouxenidis, D., Stefanaki, E.C., Tsagas, N.F., Polymeris, G.S., Tsirliganis, N.C., Kitis, G., 2008. Reconstruction of the thermally quenched glow-curve of $Al_2O_3:C$. Phys. Stat. Sol. (a) 205, 1672–1679.
- Hütt, G., Smimov, A., Tale, I., 1979. On the application of thermoluminescence of natural quartz to the study of geochronology of sedimentary deposits. PACT 3, 362–373.
- Kitis, G., 2002. Confirmation of the influence of thermal quenching on the initial rise method in $\alpha-Al_2O_3:C$. Phys. Stat. Sol. (a) 191, 621–627.
- Kitis, G., Pagonis, V., 2007. Peak shape methods for general order thermoluminescence glow-peaks: a reappraisal. Nucl. Instr. Meth. Phys. Res. B 262, 313–323.
- Kitis, G., Tuyn, J.W.N., 1998. A simple method to correct for the temperature lag in glow-curve measurements. J. Phys. D: Appl. Phys. 31, 2065–2073.
- Kitis, G., Gomez-Ros, J.M., Tuyn, J.W.N., 1998. Thermoluminescence glow-curve deconvolution functions for first, second and general orders of kinetics. J. Phys. D: Appl. Phys. 31, 2636–2641.
- Kitis, G., Pagonis, V., Carty, H., Tatsis, E., 2002. Detailed kinetic study of the thermoluminescence glow-curve of synthetic quartz. Radiat. Prot. Dosim. 100, 225–228.
- Levy, P.W., 1979. Thermoluminescence studies having applications to geology and archaeometry. PACT 3, 466–480.
- Mandowski, A., Bos, A.J.J., Mandowska, E., Orzechowski, J., 2010. Monte-Carlo method for determining the quenching function from variable heating rate measurements. Radiat. Meas. 45, 284–287.
- McKeever, S.W.S., 1985. Thermoluminescence of Solids. Cambridge University Press, Cambridge.
- Murray, A.S., Wintle, A.S., 1997. Factors controlling the shape of the OSL decay curve in quartz. Radiat. Meas. 29, 65–79.

Table 3

Analysis from peak shape method of re-quenched glow peaks (transformed from reconstructed unquenched glow curve). Only the prominent peaks of both Kilkis and B2 are mentioned in table (from heating rate 0.5 °C/s).

Sample	Peak no.	Experimental quenched (CGCD method)		Re-quenched glow peaks (peak shape method)	
		T_m (°C/s)	E (eV)	T_m (°C/s)	E (eV)
Kilkis quartz	1	75	0.64	75	0.60
	2	94	0.67	95	0.69
	4	203	0.91	201	0.89
	6	317	1.18	316	1.11
	7	372	1.20	373	1.18
B2 quartz	1	71	0.91	71	0.85
	2	134	0.96	132	0.97
	3	186	1.12	184	1.11
	4	200	1.19	200	1.13
	6	275	1.24	276	1.18
	7	295	1.25	294	1.20

- Nanjundaswamy, R., Lepper, K., McKeever, S.W.S., 2002. Thermal quenching of thermoluminescence in natural quartz. *Radiat. Prot. Dosim.* 100, 305–308.
- Pagonis, V., Tatsis, E., Kitis, G., Drupieski, C., 2002. Search for common characteristics in the glow-curves of quartz of various origins. *Radiat. Prot. Dosim.* 100, 373–376.
- Petrov, S.A., Bailiff, I.K., 1997. Determination of trap depths associated with TL peaks in synthetic quartz (350–550 K). *Radiat. Meas.* 27, 185–191.
- Prokein, J., Wagner, G.A., 1994. Analysis of thermoluminescent glow peaks in quartz derived from the KTB-drill hole. *Radiat. Meas.* 23, 85–94.
- Schilles, T., Poolton, N.R.J., Bulur, E., Bøtter-Jensen, L., Murray, A.S., Smith, G.M., Riedi, P.C., Wagner, G.A., 2001. A multi-spectroscopic study of luminescence sensitivity changes in natural quartz induced by high-temperature annealing. *J. Phys. D: Appl. Phys.* 34, 722–731.
- Spooner, N.A., Questiaux, D.G., 2000. Kinetics of red, blue and UV thermoluminescence and optically stimulated luminescence from quartz. *Radiat. Meas.* 32, 659–666.
- Subedi, B., Kitis, G., Pagonis, V., 2010. Simulation of the influence of the thermal quenching on the thermoluminescence glow-peaks. *Phys. Stat. Sol. (a)* 207 (5), 1216–1226.
- Subedi, B., Oniya, E., Polymeris, G.S., Afouxenidis, D., Tsirliganis, N.C., Kitis, G., 2011. Thermal quenching of thermoluminescence in quartz samples of various origin. *Nucl. Inst. Meth. Phys. Res. B* 269, 572–581.
- Wintle, A.G., 1975. Thermal quenching of thermoluminescence in quartz. *Geophys. J. Roy. Astronom. Soc.* 41, 107–113.
- Wintle, A.G., 1997. Luminescence dating: laboratory procedures and protocols. *Radiat. Meas.* 27 (5–6), 769–817.