

QUARTZ TL DECAY DUE TO OPTICAL BLEACHING

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Abstract: In spite of many years of research our knowledge of the traps responsible for the OSL in quartz is not complete. Many papers report the complexity of this signal, determine its components and discuss their usefulness for dating procedures. However the experiments carried out so far do not clearly show the source of carriers which recombine on the luminescence centres emitting the OSL. The trap connected with TL peak around 325°C commonly accepted as easily bleachable, can not be the only one responsible for the many observed OSL components. There should exist TL peaks corresponding to these various OSL components, although it is possible that they are related to very deep traps (that is having large energy of thermal ionisation) which are not detected in the conventional TL measurements carried out up to 500°C. With this in mind detailed analysis was made, using sedimentary quartz, of TL curves measured after successively increased time of bleaching performed during the OSL measurement. TL components were determined using fitting procedures and the effect of bleaching on each of these components is demonstrated.

1. INTRODUCTION

Considerable investigation of optically stimulated luminescence (OSL) from quartz has been made since the OSL dating method was proposed for the first time in 1985 (Huntley *et al.*, 1985). Linearly modulated – OSL (LM-OSL; Bulur, 1996) measurements allowed the distinguishing of many OSL components which can then be examined in detail. These components are usually fitted using a first order kinetics model (Jain *et al.*, 2003; Singarayer and Bailey, 2003) and so the traps responsible for them should be clearly characterised. However no simple correlation between the traps emptied during optical and thermal stimulation has been proposed until now. Investigation of TL bleaching processes appears to be the natural way of investigating such correlation.

The aim of this work was to establish a convenient method for investigation TL bleaching. However the measurement of TL bleaching characteristics is not a simple task. TL sensitivity changes with use and the differences between the TL signals from individual quartz samples require careful evaluation. Two different approaches are presented and compared, and some aspects of TL bleaching in sedimentary quartz samples are shown.

2. EXPERIMENTAL DETAILS

TL measurements were carried out using a Risø OSL-TL-DA-12 system (Bøtter-Jensen *et al.*, 1992) equipped with a halogen lamp (15 mW/cm²) and GG-420 excitation filter or the Risø TL/OSL-DA-15 system (50 mW/cm²) (Bøtter-Jensen and Murray, 1999) with the blue diodes (470±50nm) as the stimulation light source. Optical stimulation was conducted at room temperature. In both systems U-340 detection filter was employed.

Two different methods of measuring TL bleaching were used. In the first approach, a separate quartz aliquot was used for each TL bleaching curve point. This avoids the problems of TL sensitivity changes from one TL measurement to another. In this case, however, any differences in TL curve shape from aliquot to aliquot must be taken into consideration. The second approach uses only one aliquot, but then any TL sensitivity changes must be monitored, and allowed for.

Two different sediment quartz samples were used in these investigations: QM and QB. They were chosen because of their high luminescence intensity. For the QM sample both bleaching measurement methods were carried out; in the case of the QB sample the single aliquot method was used.

The multi-aliquot measurements were undertaken as follows: Five aliquots were prepared, heated to 500°C and each given a dose of 450 Gy. After 24 hours four aliquots were bleached using blue diodes for different periods of time, and then the TL of all the aliquots was measured. This procedure was repeated three times. Finally inter-aliquot calibration was undertaken by measurement of TL after a dose of 25 Gy. For each temperature point in the TL calibration curves of all aliquots, the relative deviation from the averaged calibration curve was calculated, and this deviation was used to normalise the response of each aliquot. **Fig. 1** presents the results of such calibration procedure; the bleached TL curves before (A) and after (B) correction are shown. The bleaching results can be then presented in form of the three-dimensional graph.

In the case of the single aliquot approach all the measured TL curves were decomposed into first order TL peaks. It should be stressed that the complexity of the TL curve suggests that the assumption of the trap independence is incorrect for quartz. However, this problem is not considered here, and the assumption of first order peaks is treated as a tool for analysis rather than as a de-

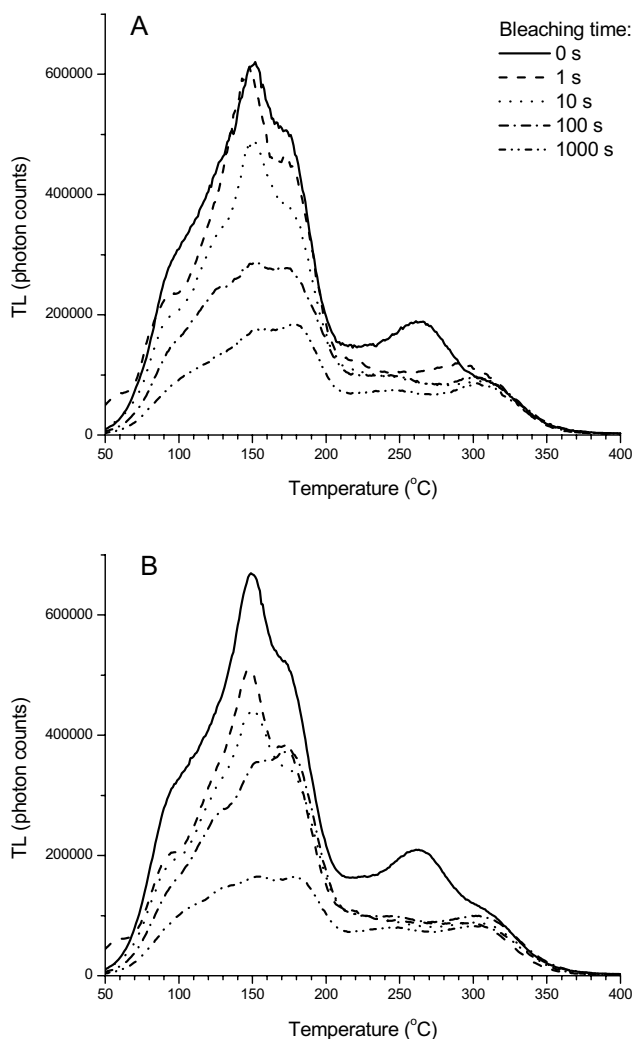


Fig. 1. The TL curves measured for 5 aliquots of sample QM after different times of blue light bleaching (heating rate 0.1K/s). The curve sequence before (a) and after (b) calibration eliminating the differences in the TL signal of particular aliquots.

scription of the general kinetics model in quartz. The usefulness of this assumption was checked by the fitting performed for the glow curves measured for different trap occupation (different doses and bleaching times) and different heating rates. The trap parameters obtained from the initial fitting gave good fitting results (residue lower than 5% of TL intensity) in all cases.

The first use of the decomposition procedure was for the glow curves measured without bleaching. The initial parameters for this fitting were trap depths, E_k (for the k -th trap). The frequency factors, s_k , and trap occupations, n_{0k} , were treated as unknown. The set of trap depth values was established on the basis of the former investigation carried out for quartz by means of the fractional glow technique (Chruścińska *et al.*, 1996). The trap parameters E_k and s_k , obtained from this initial fitting, were then kept constant during the next stage and the only fitting parameter was the trap occupation, n_{0k} . As can be seen in **Table 1** the trap parameters found in result of this analysis for quartz samples QM and QB are in good accordance.

In the single aliquot approach each bleach was preceded by a non-bleached TL measurement, to allow a check on TL sensitivity changes. The initial trapped charge concentration in the trap responsible for particular component, n_{0k} , obtained from fitting was taken as the measure of the component intensity. Two non-bleached TL curves, one determined before and the second after the bleached TL measurement, were used for the interpolation of the trap occupation immediately before each optical bleaching. The results of this analysis are presented in the form of the trapped charge concentration dependence on the bleaching time for each TL component separately. The dose administered before each step of the procedure was 100 Gy. After each dose, a 20 s preheat at 80°C was applied.

3. RESULTS AND DISCUSSION

Both multiple and single aliquot approaches were applied to the QM sample. **Figs 2 - 3** present the results of the multiple approach – the TL signal decay due to bleaching by blue light. Three measurement sequences were performed on the same aliquot set. The heating rate during the TL measurement (0.3, 1, 0.1°C/s) and the total dose applied from the beginning of experiments (450, 900, 1350 Gy) define the differences between these three measurement sequences. Comparing the three non-bleached TL curves obtained for different heating rates the luminescence quenching effect can be observed. Although the TL decay in all cases looks similar, when the decay curves are derived for a particular curve maximum (in **Fig. 4**) the small differences between the subsequent experiments can be seen. The majority of light sensitive TL originates from the traps responsible for the TL maximum below 250°C. One can find another effectively bleached region of the TL curve around 280°C (this peak is observed around 325°C for a heating rate of 10°C/s). Here the bleaching is rapid in the first stage (1 s) but then it is very slow and seems to reach a residual level not much lower

than 30% of the initial value. The high temperature edge of the glow curve is clearly not light sensitive.

Fig. 5 shows the bleaching result for the same sample obtained by the single aliquot method using the green light for stimulation. Before the experiment started the sample was dosed and heated many times. At the beginning the TL sensitivity of the sample increased, next it decreased and finally it became stable. It then remained constant during the bleaching experiments and so the TL intensity versus bleaching time can be presented in the same manner as in the multi-aliquot method. The TL versus bleaching time for the particular curve maxima is also presented in **Fig. 4**, for comparison with the results for the stimulation by blue light. The differences between the results for blue and green light are remarkable. The different stimu-

lation light power is not a sufficient explanation. One can suppose here that the wave length plays a role, but other effects should also be considered. In the multi-aliquot method a dose of 450 Gy was employed whereas in the single-aliquot method the dose was 100 Gy. Bleaching of peaks below 200°C at higher doses is more efficient than for low doses (**Fig. 4 b,c**). The opposite is observed for the peak at 325°C (here marked as 280°C) which is bleached more efficiently at low doses (**Fig. 4a**). One more reason for the various bleaching characteristics observed in both measurement sequences could be the different concentration of charge trapped in deep level which are not emptied during the TL measurement to ~500°C. This could also cause the small differences observed earlier in the successive multi-aliquot measurements.

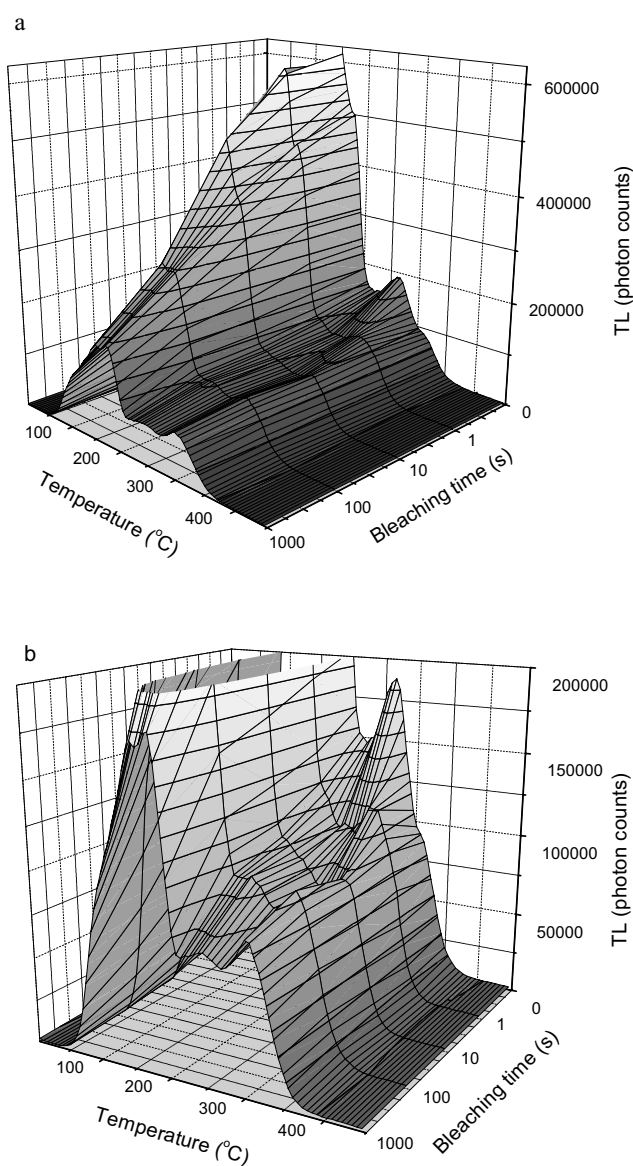


Fig. 2. The TL signal of QM sample decay due to bleaching by the blue light. The multi-aliquot method of registration. The heating rate during the TL registration was 0.1°C/s and the excitation dose for each aliquot was 450 Gy; (a) – full temperature range; (b)- the glow curve detail demonstrated the quick decay of the maximum around 280°C.

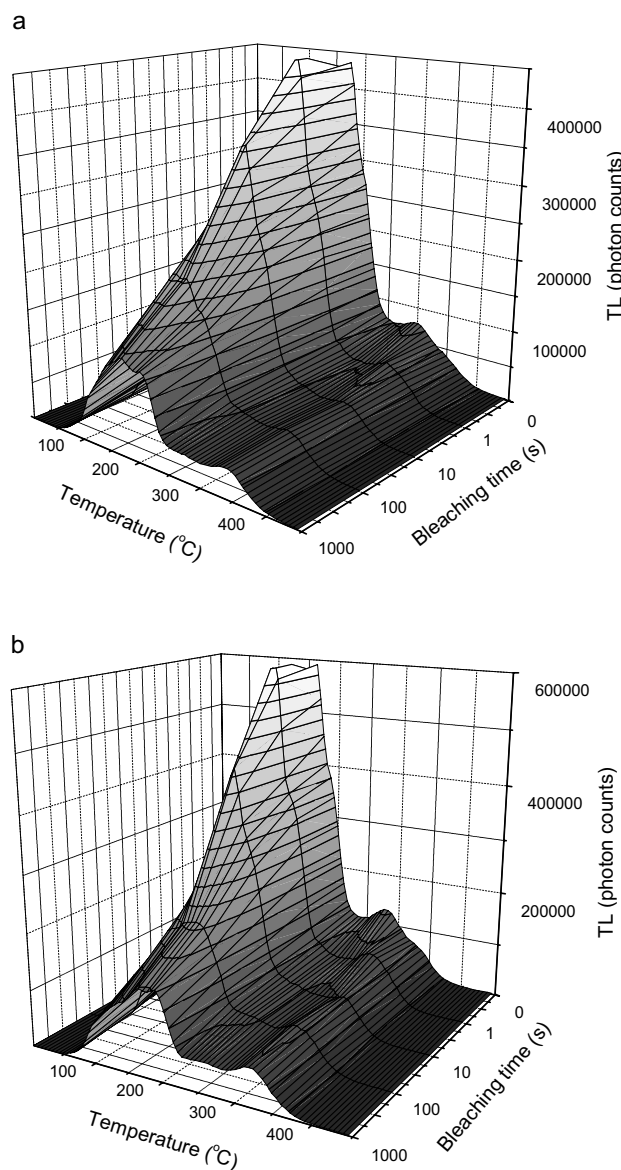


Fig. 3. The TL signal of QM sample decay due to bleaching by the blue light. The multi-aliquot method of registration. The heating rate during the TL registration was 1°C/s (a) and 0.3°C/s (b). The excitation dose for each aliquot in both sequences was 450 Gy.

TL curves of the QM sample obtained in single aliquot approach were decomposed into first order peaks as described above. Over 10 peaks were necessary for good fitting (Fig. 6). Ten of them could be treated as uninfluenced by the preheat and pause between excitation and heating. The bleaching of the components related to particular TL peaks (maximum 160°C – component 5, maximum 180°C – component 1, maximum 280°C – component 4) are also presented in Fig. 4. The results obtained

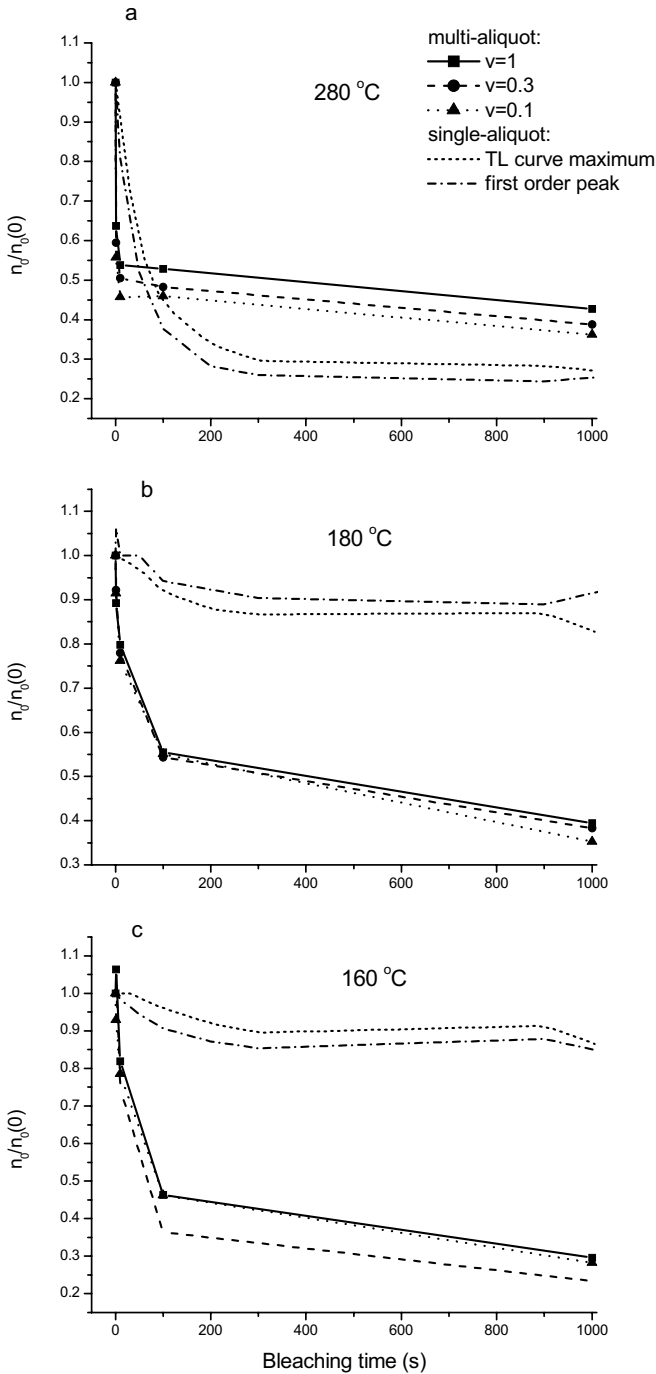


Fig. 4. The TL versus bleaching time for the particular maxima of QM sample glow curve – 280°C (a), 180°C (b) and 160°C (c). The decay curves obtained by the multi-aliquot method (blue light stimulation) in three measurement sequences with different heating rate – 0.3, 1 and 0.1°C/s (symbol plots) and the decay curves obtained by the single-aliquot method (green light stimulation) (line plots).

for curve maximum and results for related TL component are similar. However the glow curve, as one can see in Fig. 6, is made up of a lot of overlying peaks. The dependencies of trapped charge, n_0 , on the bleaching time for all TL components are shown in Fig. 8a. They behave in different ways during the optical stimulation. Some of them do not decay but increase. Only the detailed knowledge of the traps active in TL and OSL can permit the explanation of such behaviour.

Fig. 7 presents the TL sensitivity changes for sample QB during the bleaching experiment performed by the single-aliquot method and the Fig. 8b shows the TL components decay due to optical bleaching. The comparison of the results in Fig. 8a-b shows that the majority of TL peak in both samples behaves alike, although discrep-

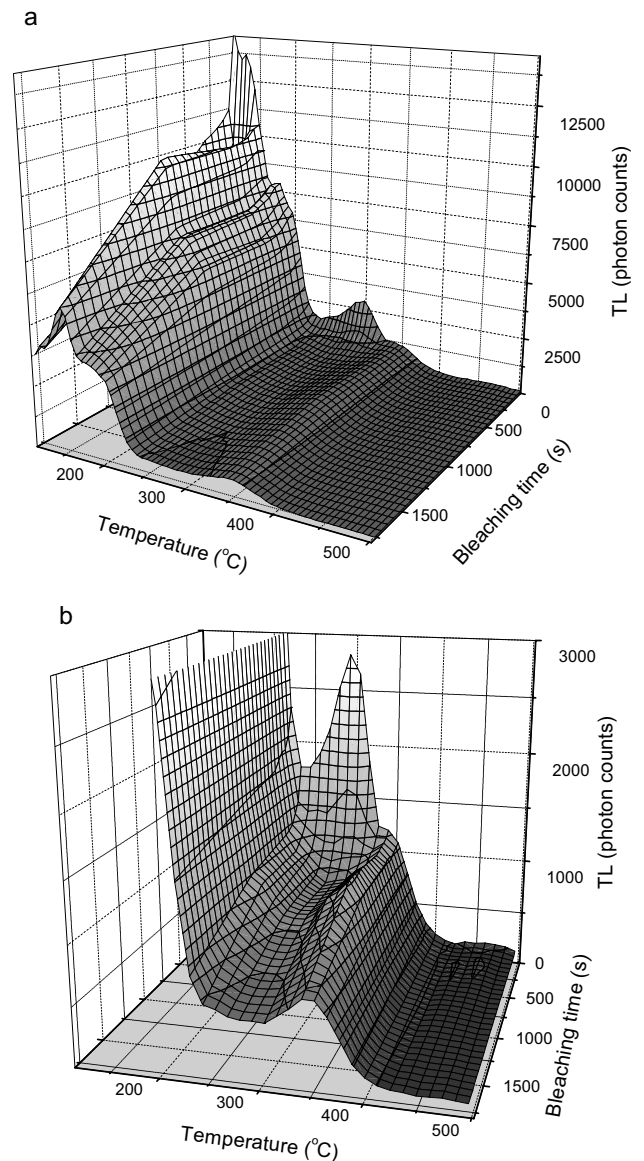


Fig. 5. The TL signal of QM sample decay due to bleaching by the green light. The single-aliquot method of registration. The heating rate during the TL registration was 1°C/s and the excitation dose for each aliquot was 100 Gy; (a) – full temperature range; (b) – the glow curve detail demonstrated the quick decay of the maximum around 280°C.

ancies are observed. For the QM sample the components 9, 10, 11 increase significantly at the beginning of the bleaching before beginning to decay, but even after 300s their occupation is higher or equal to the initial value before bleaching. In sample QB these components decay from the beginning of experiment and one of them (11) decay very quickly.

Another difference between the samples QM and QB concerns the behaviour of components 5 and 12. The peak 12 in sample QM is very effectively bleached. In sample QB the initial population is low and any changes in height are difficult to determine on the background of peak 1 and 5. So in the case of sample QB, peak 12 was added to peak 5 and the decay of the sum of the two peaks is presented as peak 5 in Fig. 8b.

As one can see, component 4 is well bleached in both samples. This component is responsible for the so called 325°C peak (here at the heating rate of 1°C/s – about 280°C) but only partially. This TL curve maximum is composed also of the peaks 3 and 10 and their decay is slower and, in the case of peak 10, can depend on the sample. Moreover, the decay of peak 4 is not a simple first order decay. The level of 25% of the initial trapped charge concentration seems to be a residual level for this component

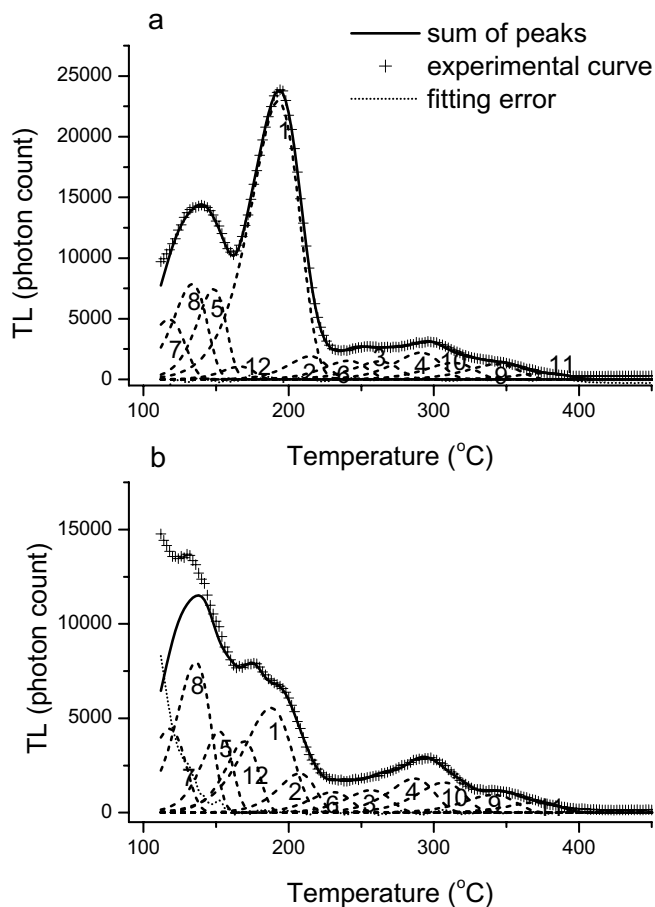


Fig. 6. The non-bleached TL curve for sample QB after the 100 Gy excitation dose (a) and for sample QM after the excitation dose 450 Gy (b). TL curves were decomposed into first order peaks by means of the fitting procedure. The fitting parameters are presented in Table 1. The heating rate in both cases was 1°C/s.

Table 1. The trap parameters obtained as a result of TL curve decomposition into first order peaks for samples QM and QB. Experimental and fitted curves are presented in Fig. 6.

Peak number	QM		QB	
	E_k (mev)	s_k (1/s)	E_k (mev)	s_k (1/s)
1 (180°C)	1100	$6.42 \cdot 10^{10}$	1100	$4.66 \cdot 10^{10}$
2	1550	$1.41 \cdot 10^{15}$	1550	$7.72 \cdot 10^{14}$
3	1591	$8.87 \cdot 10^{13}$	1591	$6.01 \cdot 10^{13}$
4 (280°C)	1722	$2.00 \cdot 10^{14}$	1722	$1.51 \cdot 10^{14}$
5 (160°C)	1502	$6.15 \cdot 10^{16}$	1502	$9.21 \cdot 10^{16}$
6	1540	$1.83 \cdot 10^{14}$	1540	$8.48 \cdot 10^{13}$
9	1856	$9.77 \cdot 10^{13}$	1856	$8.51 \cdot 10^{13}$
10	1773	$1.54 \cdot 10^{14}$	1773	$1.02 \cdot 10^{14}$
11	2058	$7.18 \cdot 10^{14}$	2058	$2.33 \cdot 10^{14}$
12	1500	$1.08 \cdot 10^{16}$	1500	$1.42 \cdot 10^{16}$

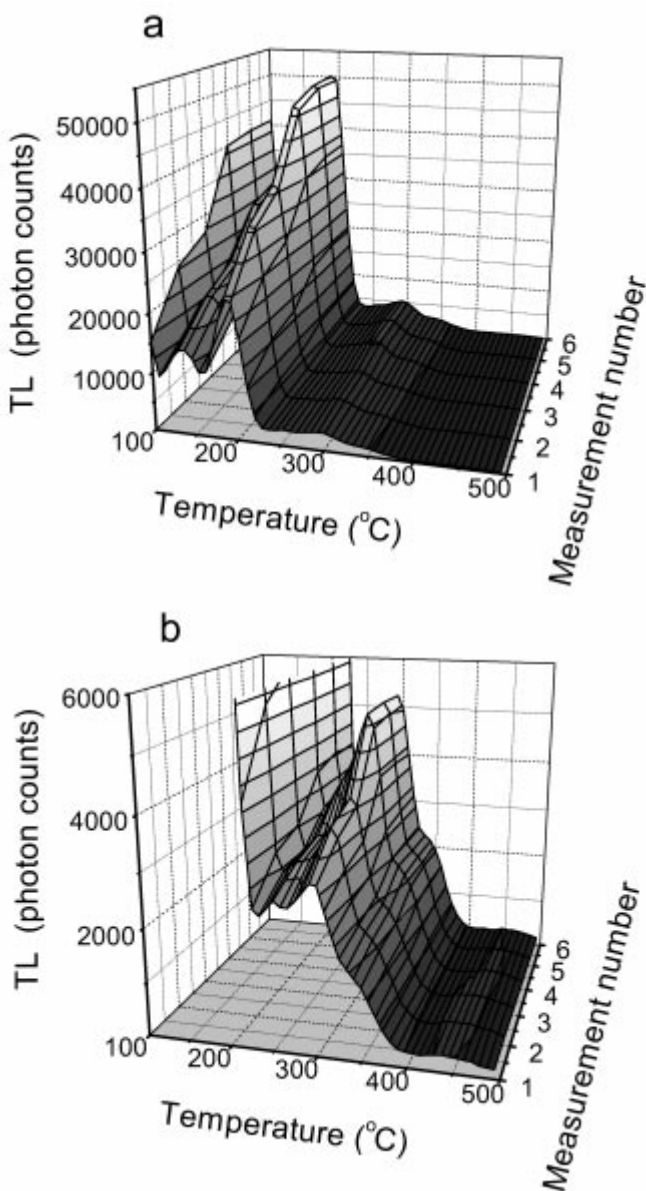


Fig. 7. The TL sensitivity changes appearing for the sample QB during the bleaching experiment performed by the single-aliquot method. (a) – full temperature range, (b) – the detail.

in the case of lower excitation dose and 40% in the case of higher excitation dose (Fig. 4a). Another candidate for the well bleached component is peak 12. As one can see from present investigation the concentration of traps responsible for this component can vary from sample to sample.

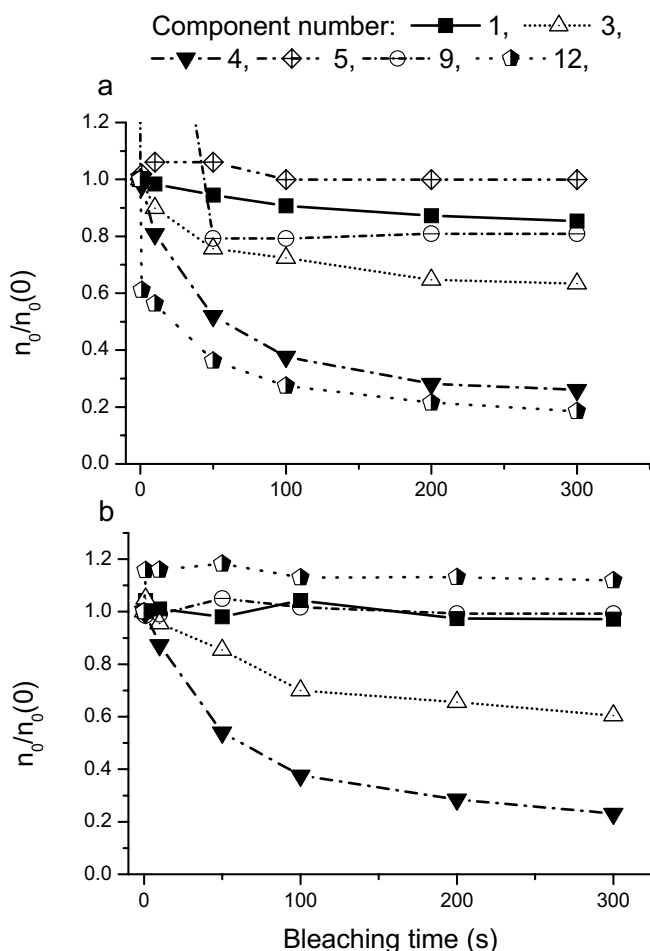


Fig. 8. The optical bleaching of some chosen TL components obtained in result of single aliquot method (green light stimulation). Results for sample QM (a) and sample QB (b). The decomposition of glow curves into particular TL components are presented in Fig.6 and the kinetics parameters are listed in Table1.

4. CONCLUSION

Two experimental approaches to examining the TL bleaching processes were applied: the single-aliquot and multi-aliquot. In the case when big differences appear between the aliquots of the same grain sample the single-aliquot method can be applied but special attention should be paid to any sensitivity changes in the sample. TL curve decomposition into first order peaks can be used to take the sensitivity changes into consideration.

The examples presented here of TL bleaching indicate that the bleaching processes in quartz require the application of a more complex model of stimulated luminescence kinetics than the model of independent traps. More complex investigation, including LM-OSL and TL bleaching measurements, should be conducted in order to allow the correlation between the traps active in TL and in OSL process.

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