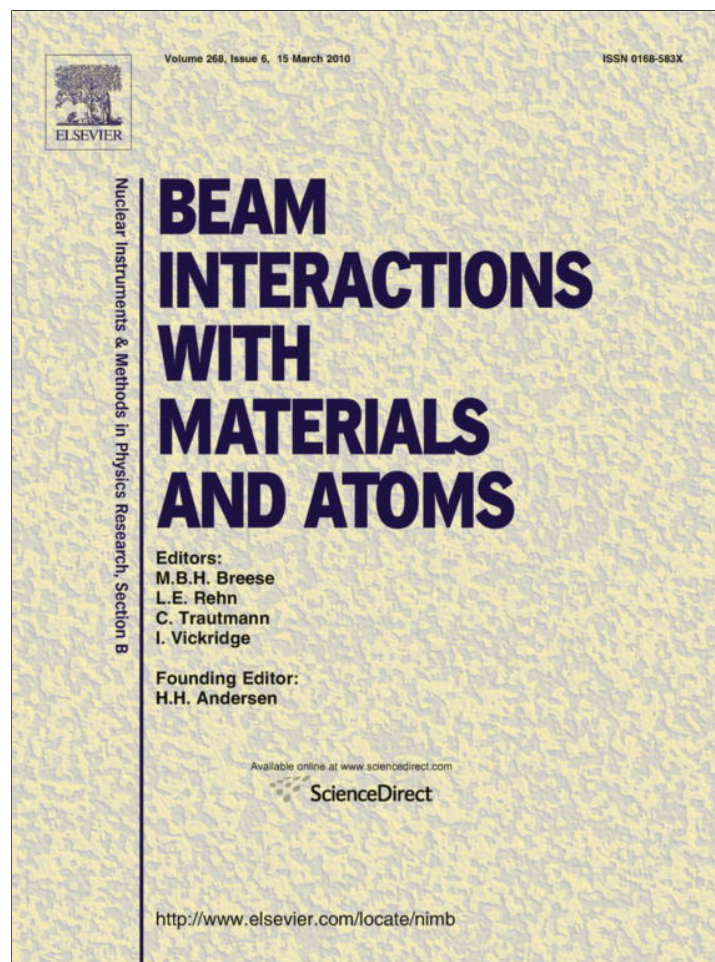


Provided for non-commercial research and education use.
Not for reproduction, distribution or commercial use.



This article appeared in a journal published by Elsevier. The attached copy is furnished to the author for internal non-commercial research and education use, including for instruction at the authors institution and sharing with colleagues.

Other uses, including reproduction and distribution, or selling or licensing copies, or posting to personal, institutional or third party websites are prohibited.

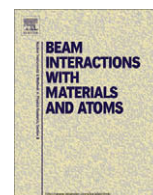
In most cases authors are permitted to post their version of the article (e.g. in Word or Tex form) to their personal website or institutional repository. Authors requiring further information regarding Elsevier's archiving and manuscript policies are encouraged to visit:

<http://www.elsevier.com/copyright>



Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Investigation of OSL signals from very deep traps in unfired and fired quartz samples

G. Kitis^a, N.G. Kiyak^b, G.S. Polymeris^{b,c}, V. Pagonis^{d,*}^a Aristotle University of Thessaloniki, Nuclear Physics Laboratory, Thessaloniki, Greece^b ISIK University, Physics Department, Faculty of Science and Arts, 34980 Sile, Istanbul, Turkey^c Cultural and Educational Technology Institute – R.C. Athena/Archaeometry Laboratory, Xanthi, Greece^d McDaniel College, Physics Department, Westminster, MD 21157, USA

ARTICLE INFO

Article history:

Received 19 November 2009

Received in revised form 4 December 2009

Available online 16 December 2009

Keywords:

Luminescence

Thermoluminescence (TL)

Optically stimulated luminescence (OSL)

Quartz

Dosimetry

Dating

ABSTRACT

This paper presents an attempt to isolate experimentally optically stimulated luminescence (OSL) signals which may originate from very deep traps (VDT) in quartz samples. As VDT we consider those traps which are responsible for TL glow peaks with a peak maximum temperature above a TL readout temperature of 500 °C. The basic experimental procedure used to isolate OSL signals from VDT is heating the quartz samples to 500 °C immediately before measuring the OSL signal. The study was carried out on eight quartz samples of very different origins; it is found that all eight samples exhibit OSL signals from VDT, and for a wide region of OSL stimulation temperatures. The OSL signal from VDT depends strongly on the type of quartz sample studied and on whether the sample was fired at high temperatures or not. The behavior of the OSL signal from VDT as a function of the stimulation temperature is found to be very different in fired and unfired samples. The thermal activation energy E for the OSL signals from VDT is obtained in both fired and unfired samples. The OSL signal from VDT in quartz samples fired at 800 °C for 1 h is very high, and the OSL curves consist of three well-defined components and a fourth slow component which is rather poorly resolved. The dose response of these components is obtained using a computerized deconvolution procedure for the dose region 0.5–300 Gy. The results are of importance for dating of ancient fired ceramics, since OSL signals from VDT could potentially extend appreciably the equivalent dose region toward both lower and higher values.

© 2009 Elsevier B.V. All rights reserved.

1. Introduction

Experimentally observed luminescence signals from quartz samples in both thermoluminescence (TL) and optically stimulated luminescence (OSL) studies are of a complex nature. These luminescence signals have been known to originate from several electron and hole traps, as well as from several recombination centers [1–4]. During OSL dating of quartz the OSL signal is typically measured at an elevated temperature of 125 °C, in order to avoid problems associated with the 110 °C TL trap. It is commonly assumed that the OSL signal measured at 125 °C consists of several light sensitive components usually termed the fast, medium and slow OSL components.

This paper presents an attempt to isolate experimentally OSL signals which may originate from very deep traps (VDT) in quartz samples. As VDT we consider those traps which are deeper than those associated with peaks occurring up to a TL readout temperature of 500 °C. The basic experimental procedure used to isolate OSL signals from VDT is to heat the quartz samples to 500 °C,

and to measure the OSL signal immediately after; it is assumed that OSL signals measured in this manner will be due to VDT. There are no other studies in the literature which specifically target OSL signals from very deep traps in quartz. In the rest of this paper we will refer to these signals as OSL from VDT. These VDT traps are postulated here for the first time in order to explain the OSL results of our experiments.

In previous work Singarayer and Bailey [5] and Jain et al. [6] separated the linearly-modulated optically stimulated luminescence (LM-OSL) signals from quartz into several components, and obtained their dose response after the samples underwent optical and thermal treatments. These authors did not attempt to isolate the OSL signals from VDT, which is the goal of this work. The aim of the present work is twofold. Firstly, to investigate the magnitude of the OSL signals originating from VDT of unfired quartz samples of various origins, in which the VDT are occupied by electrons trapped during geological irradiation. The presence or absence of such OSL signals is of great practical importance, because they may lead to incorrect estimates of the equivalent dose (ED) during dating protocols. Secondly, to investigate the properties of VDT in samples fired at very high temperatures in the laboratory, in order to empty all the VDT.

* Corresponding author. Tel.: +1 (410) 857 2481; fax: +1 (410) 386 4624.
E-mail address: vpagonis@mcDaniel.edu (V. Pagonis).

2. Materials and methods

The samples were quartz of various origins and all from diverse locations of Turkey. The samples with laboratory codes SLE and PDK were from very young coastal-dunes and were collected from the surrounding area of Istanbul, with equivalent doses of 0.011 and 0.008 Gy Kiyak and Canel [7]. INK sample was also from young dunes from Inkumu-Bartın, a coastal area in the middle part of the Black Sea Region with a natural dose 0.022 Gy. The sample PTR is a deposit from Patara in the Mediterranean coast and contains a natural dose about 2.2 Gy. The samples SRZ1 and SRZ2 were collected from different layers of a marine terrace in Saroz on the Aegean coastal area. These samples are old and the natural dose accumulated in quartz grains were estimated as 181 and 195 Gy correspondingly by using a SAR protocol. Samples ME1 and ME2 are fluvial terrace sediments collected from an area close to the Marmara Ereglisi in the European part of the Marmara Region, having natural doses of 127 and 145 Gy correspondingly. The original samples were initially wet sieved, and quartz grains with dimensions between 90 and 180 μm were selected and deposited on stainless steel disks of 1 cm^2 area.

Blue light emitting diodes (LEDs) (470 nm, 40 mW/cm^2) were used for optical stimulation. All measurements were performed using a RISO TL/OSL reader (model TL/OSL-DA-15) equipped with a $^{90}\text{Sr}/^{90}\text{Y}$ β -ray source delivering a dose rate of about 0.1 Gy/s. The reader is fitted with an EMI 9635QA PM Tube, and luminescence signals were detected through U-340 filters. The TL readout heating rate used was 1 K/s for all measurements.

3. Results and discussion

3.1. Unfired samples

The experimental multi-aliquot protocol used to study the magnitude of the OSL signals from VDT was as follows:

- Step 1: Irradiation of the quartz sample possessing its natural TL signal (NTL) with an added dose of 300 Gy.
- Step 2: TL readout up to 500 °C.
- Step 3: Test dose TD = 10 Gy and measurement of TL by heating up to 500 °C.
- Step 4: Blue CW-OSL at a variable stimulation temperature T_i for 2000 s. T_i is varied in the temperature region from 125 up to 375 °C in steps of 25 °C.
- Step 5: Measurement of the residual TL signal by heating up to 500 °C.
- Step 6: Test dose TD = 10 Gy and measurement of TL by heating up to 500 °C.
- Step 7: Repeat steps 1–6 for a new aliquot and for new stimulation temperature T_i .

At step 1 an additional irradiation is given to the natural sample to ensure the filling of the VDT, since their irradiation history is unknown. In step 2 any trapping level responsible for TL glow peaks up to 500 °C is erased. The test dose measurement in step 3 is used to evaluate the sensitivity of the sample to a low test dose, and to verify that heating up to 500 °C erases the TL signal down to the background level. Step 4 aims to study the magnitude of the OSL signal from VDT as a function of the OSL stimulation temperature. Step 5 is applied to find whether some TL signal reappears between 20 and 500 °C after the stimulation at high temperatures, by some other mechanism, for example phototransfer. Finally step 6 evaluates the sensitivity to the low test dose as in step 3, and allows an evaluation of how the protocol may affect the sensitivity of the quartz sample.

The protocol was applied to the eight different quartz samples described above. In six of the samples only three OSL measuring temperatures were used (190, 250 and 375 °C). However, two samples were selected for a more detailed study, one exhibiting a high OSL signal from VDT, and the second one exhibiting a low OSL signal from these traps. In these two samples (INK and PDK) the optical stimulation was studied in the whole temperature region from 125 to 375 °C in steps of 25 °C. The purpose of this experiment is to study the dependence of the OSL signals from VDT on the stimulation temperature, and to attempt an evaluation of the activation energy for the processes involved.

One of the main results of this experiment is that all quartz samples exhibit OSL signals from VDT as shown in Figs. 1 and 2. Fig. 1 shows the samples having a high OSL signal from VDT, whereas Fig. 2 shows the samples having correspondingly low OSL signals. In all cases the background level is also shown. Not surprisingly, the intensity of the OSL signals from VDT was found to be strongly dependent on the type of quartz sample used.

It is useful to present in Fig. 3 typical results of steps 1–6 of the protocol. The TL glow-curve (a) of Fig. 3 results after the irradiation with the laboratory dose of 300 Gy. The TL glow-curve (b) measured in step 3 of the protocol gives (i) the sensitivity to a low dose after the heavy irradiation in step 1 and (ii) ensures that the TL readout in step 2 erases the TL signal down to the usual background level. The absence of any signal in curve (c) of step 5, shows that the optical stimulation at the high temperatures of step 4 does not cause any retrapping at the traps responsible for the low temperature TL glow peaks. Finally, the TL glow-curve (d), which almost coincides with glow-curve (b), shows that the sample treatment in steps 2–5 does not change significantly the sensitivity of the samples.

The basic characteristic of the OSL curves shown in Figs. 1 and 2 is that they increase from zero to a somewhat higher value, and then remain more or less constant with time. However, in some samples a fast component appears at very short times. These cases are shown in Fig. 4, where only the first 300 s of the stimulation curves are included in the plot. Furthermore, this fast OSL component appears only for OSL stimulation temperatures lower than 175 °C, and is not present at higher stimulation temperatures. Obviously this component can not be related to the known fast OSL component originating from the electron trap responsible for the 325 °C TL glow peak of quartz, which was emptied by the TL readout up to 500 °C.

Fig. 5a shows the behavior of the total OSL from VDT as a function of the stimulation temperature. It is rather surprising that the OSL signals in Fig. 5a increase continuously with the stimulation temperature, even at the highest stimulation temperature of 375 °C. The exponential behavior in Fig. 5a allows the evaluation of the activation energy E of this process for the two samples studied. The natural logarithm of the OSL values is drawn against $1/kT$, where T represents the stimulation temperature, and the slopes of these graphs represent the activation energy E . The respective Arrhenius plots are shown in Fig. 5b. In the case of INK quartz which exhibits a strong OSL signal from VDT, the activation energy was found to be $E = 0.439 \pm 0.03$ eV, whereas for the case of PDK quartz, which shows a weak OSL signal from VDT, the activation energy was found to be very similar, namely $E = 0.423 \pm 0.04$ eV.

However, the above analysis does not take into account the possible presence of thermal quenching in the quartz samples. It is well known that luminescence signals from quartz exhibit a reduced intensity at elevated stimulation temperatures due to the phenomenon of thermal quenching. Even though the presence of thermal quenching effects is commonly assumed during TL/OSL studies, the universality of this phenomenon for all types of quartz samples has never been proved. We now present an analysis of the data by assuming the presence of thermal quenching, and by cor-

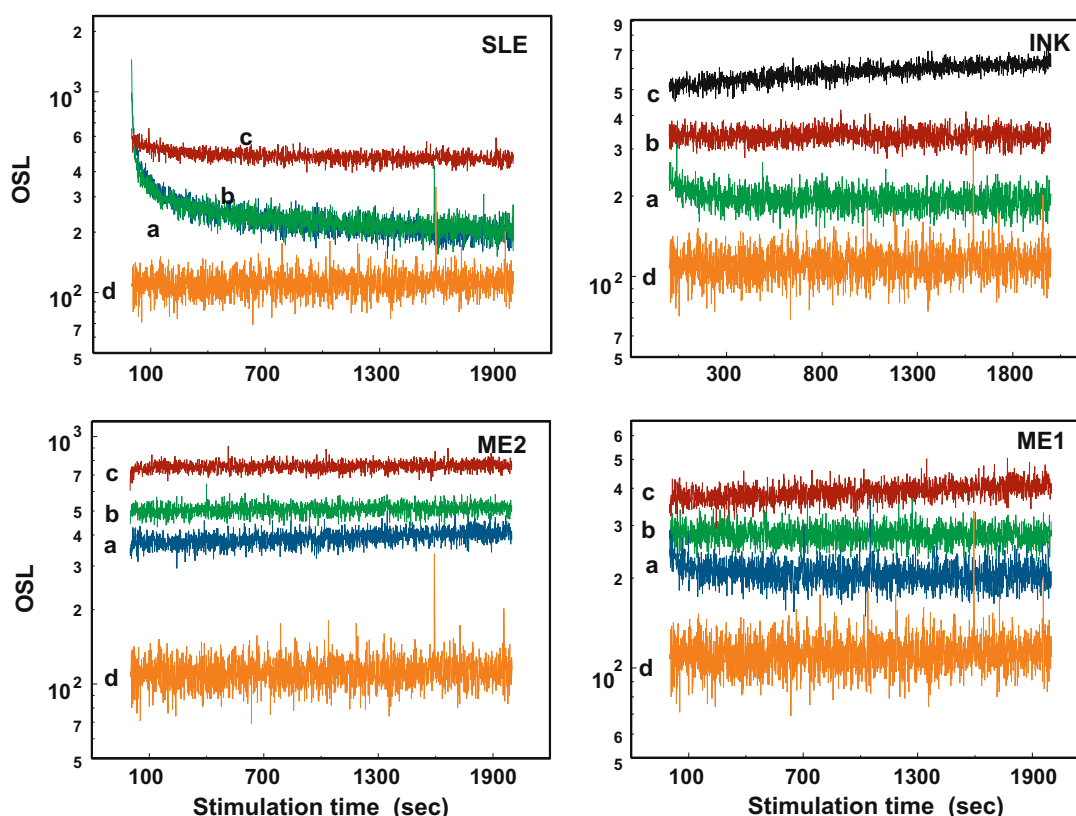


Fig. 1. The measured OSL signals for four quartz samples exhibiting high OSL from VDT, and for three different stimulation temperatures. Curve (a) 190 °C, curve (b) 250 °C and curve (c) at 375 °C. Curve (d) corresponds to the background OSL signal measured at room temperature.

recting the OSL signals for the presence of this effect. By using typical values of the thermal quenching parameters $C = 2.8 \times 10^7$ and $W = 0.64$ eV given by Wintle [8], it is possible to correct the OSL intensities due to thermal quenching as follows. Initially, the values of the thermal quenching efficiency $\eta(T)$ are evaluated for each one of the stimulation temperatures T using the well known expression for the luminescence efficiency:

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

where k is the Boltzmann constant and T is the stimulation temperature. Next the OSL intensity at each temperature T is corrected for the effect of thermal quenching by dividing with the value of $\eta(T)$. Finally Arrhenius plots of the logarithm of the corrected OSL values against $1/kT$ are drawn, with the slopes of these graphs representing the thermal activation energy E for this process. The new results are shown in Fig. 6, and the new values of activation energies assuming thermal quenching effects are $E = 1.02 \pm 0.03$ eV for INK quartz and $E = 0.93 \pm 0.03$ eV for PDK quartz. These results are further discussed in a later section along with similar results obtained for fired samples.

In summary, if one analyzes the data neglecting the effect of thermal quenching, the OSL signals from VDT can be described by a thermally assisted process with an activation energy $E \sim 0.4$ eV. However, if one corrects the data for thermal quenching effects, the OSL signals from VDT can be described by a thermally assisted process with a much higher activation energy $E \sim 1.0$ eV. In order to decide which method of data analysis is correct, additional measurements are required to ascertain the presence of thermal quenching for OSL signals from VDT, and also to measure the corresponding thermal quenching parameters C , W for this process.

3.2. Fired samples

The irradiation history of the VDT of unfired quartz samples is unknown. It is very probable that during the natural irradiation of the sample these traps were filled with electrons to a high degree, and that the additional artificial irradiation causes their complete saturation. The experimental results in Figs. 1 and 2 showed clearly the presence of OSL signals from VDT in eight quartz samples of very different origin. However, the most important property of the VDT traps from both a physical and a practical point of view, is the behavior of these signals as a function of VDT occupancy, i.e. as a function of the irradiation dose. This requires the zeroing of the VDT occupancy, which is possible by firing the samples at very high temperatures.

The behavior of the OSL signals from VDT as a function of trap occupancy was studied for quartz samples which were fired at 800 °C for 1 h, and were subsequently cooled quickly to room temperature by placing them on a ceramic block immediately after their extraction from the furnace. The firing temperature of 800 °C was selected firstly as a representative firing temperature of ancient ceramics, and secondly because no irreversible phase transitions occur up to this temperature in quartz samples. The samples studied were two samples which exhibited strong OSL signals from VDT (samples SLE and INK in Fig. 1), and two samples which exhibited weak OSL signals from VDT (samples PTR and SRZ1 in Fig. 2).

3.2.1. OSL signals from VDT as a function of stimulation temperature

In the first part of the study the OSL signal from VDT was studied as a function of the stimulation temperature, in order to find the optimal stimulation temperature for performing the dose response study. A multiple aliquot protocol was used as follows:

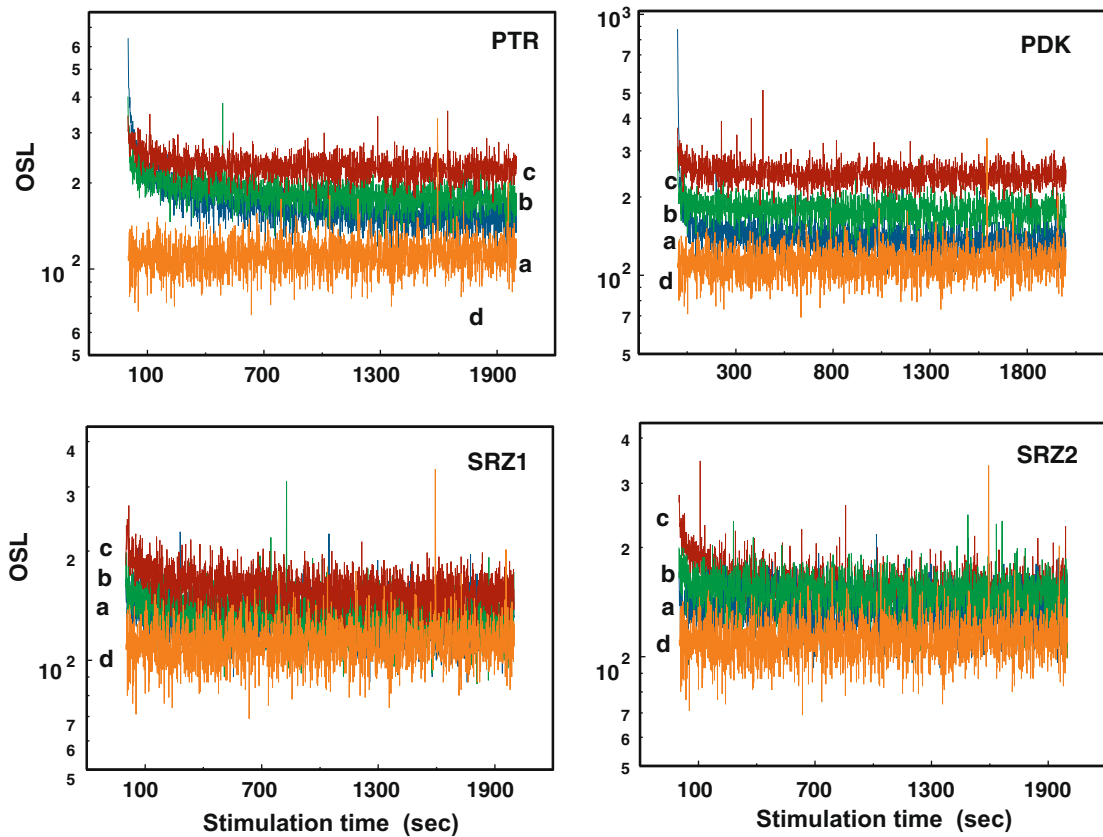


Fig. 2. The measured OSL signals from VDT for four quartz samples exhibiting low OSL from VDT, and for three different stimulation temperatures. Curve (a) 190 °C, curve (b) 250 °C and curve (c) at 375 °C. Curve (d) corresponds to the background OSL signal measured at room temperature.

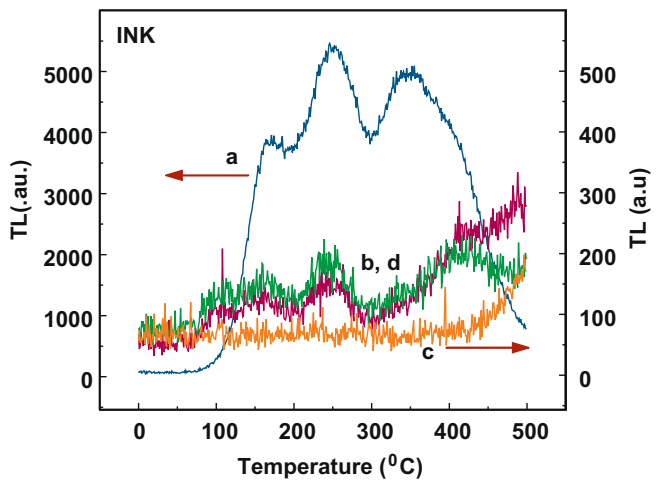


Fig. 3. Typical examples of TL glow curves measured during the experimental protocol described in the text. (a) NTL plus 500 Gy beta dose. (b) TL glow curve from step 3. (c) Residual TL from step 5 and (d) TL from step 6 of the protocol.

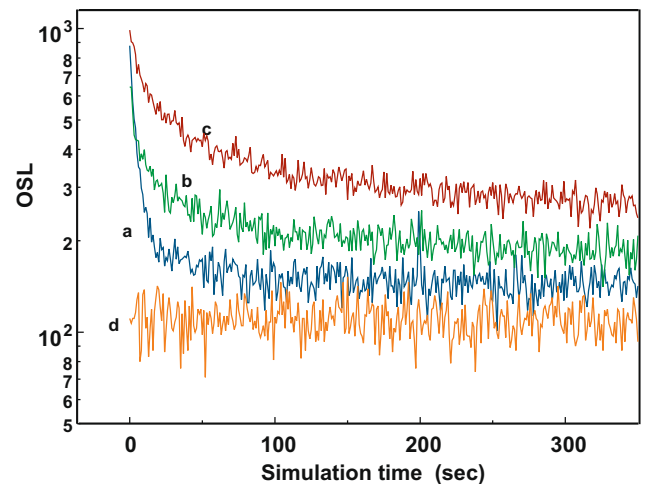


Fig. 4. Examples of OSL curves from VDT measured at 125 °C showing a fast component at very short stimulation times. Curve (a) PDK quartz, curve (b) PTR quartz, curve (c) SLE quartz and curve (d) is the OSL background.

- Step 1: Test dose TD = 1 Gy and TL readout up to 250 °C.
- Step 2: Irradiation of the sample with added dose of 300 Gy, and TL readout up to 500 °C.
- Step 3: TD = 1 Gy and TL readout up to 500 °C.
- Step 4: Blue CW-OSL at T_i for 2000 s, where the stimulation temperature T_i varies from 125 to 375 °C in steps of 25 °C.
- Step 5: Residual TL readout up to 500 °C.
- Step 6: TD = 1 Gy and TL readout up to 500 °C.
- Step 7: Repeat steps 1–6 for a new sample and new temperature.

The steps of interest in this experiment are steps 4 and 5. Steps 3 and 6 are included as a test for variations occurring in the sensitivities of the samples during the protocol. It was found that the sensitivity was increased from steps 1 to 3; this change corresponds to the usual pre-dose sensitization, whereas the sensitivity remains the same between steps 3 and 6. The first important observation is the very high intensity of the OSL signal from VDT shown in Fig. 7. The signal consists of two very distinct parts;

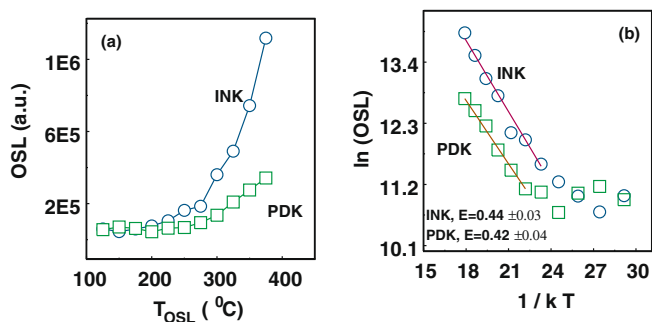


Fig. 5. (a) The integrated OSL signal from VDT for the case of two unfired samples INK and PDK. (b) Corresponding Arrhenius plot assuming that no thermal quenching is present in these samples. The activation energy for INK quartz is $E = 0.439 \pm 0.03$ eV, and for PDK quartz $E = 0.423 \pm 0.04$ eV.

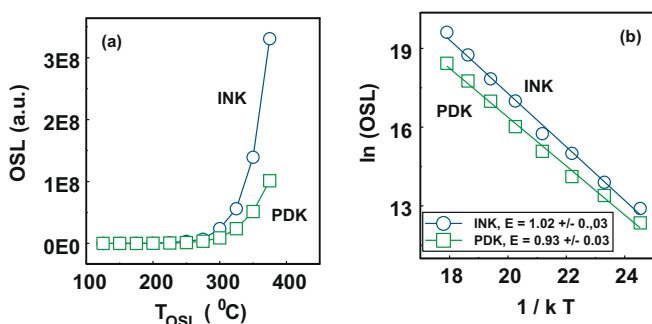


Fig. 6. (a) The integrated OSL signal from VDT in Fig. 5a, after correction for thermal quenching. (b) The corresponding Arrhenius plots yield new E values for INK quartz $E = 1.02 \pm 0.03$ eV, and $E = 0.93 \pm 0.03$ eV for PDK quartz.

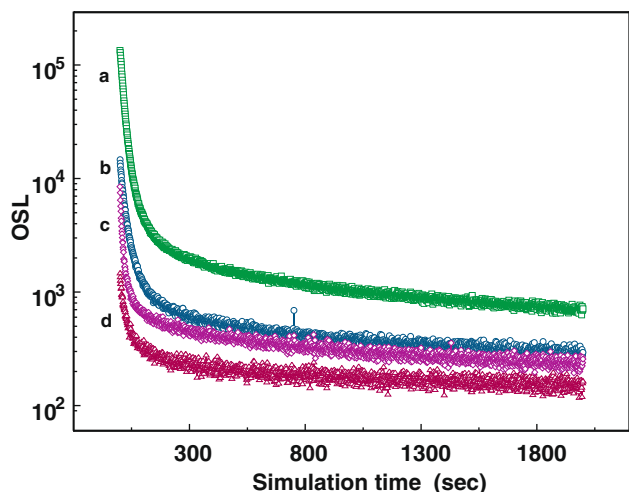


Fig. 7. Examples of OSL curves from VDT measured at 125 °C for four quartz samples. (a) SLE, (b) INK, (c) SRZ1 and (d) PTR.

the first part of the signal appears for stimulation times much less than 50 s, and the second part appears at longer stimulation times up to 2000 s. This dual nature of the OSL signal was also verified by transforming the CW-OSL curves into peak shaped curves, as is discussed in the next section. As a measure of the OSL signal we use the integral of each of these two parts, with the results shown in Fig. 8.

The qualitative behavior shown in Fig. 8 is the same for the four quartz samples. The OSL signal from VDT increases with the stim-

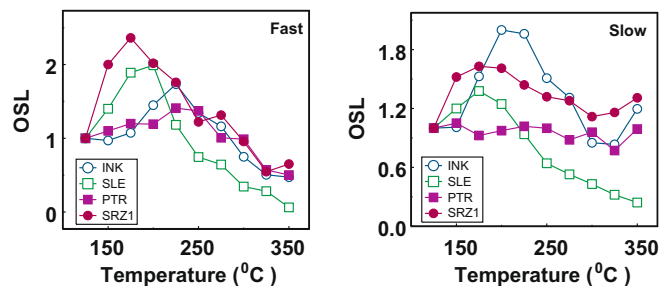


Fig. 8. Integrated OSL intensity from VDT as a function of the OSL stimulation temperature. (a) Data for the fast integrated part of the signal (0–50 s), and for the slow integrated part of the signal (50–2000 s).

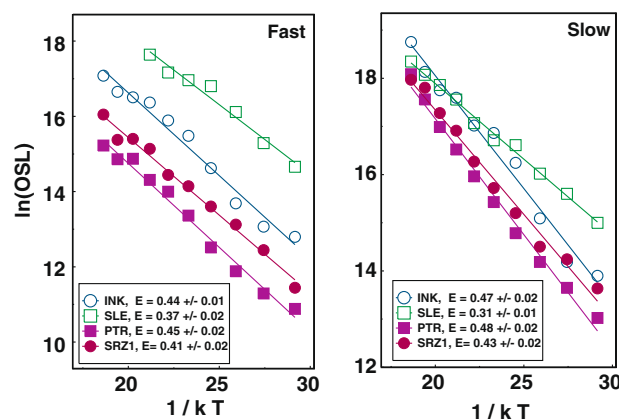


Fig. 9. Arrhenius plot resulting from the data of Fig. 8, which were corrected for thermal quenching effects at the respective OSL measuring temperatures.

ulation temperature and reaches a maximum at some temperature depending on the sample; subsequently the integrated OSL signal decreases continuously up to the highest stimulation temperature of 375 °C. The decrease of the OSL signal at higher stimulation temperatures is a strong indication for the presence of thermal quenching [9,10]. Therefore a correction procedure of the OSL intensity similar to the one applied above for unfired quartz is necessary. By applying this correction to the data of Fig. 8, new plots of the logarithm of corrected OSL values against $1/kT$ are obtained and are shown in Fig. 9 for both integrated regions. It is rather impressive how the thermal quenching efficiency correction transforms the data into exponential functions at all stimulation temperatures, as shown in Fig. 9a and b. The resulting values of the activation energies are given in Fig. 9, and are very similar at $E \sim 0.4$ eV for all four types of quartz sample studied.

It is interesting to compare the results of Fig. 9 with the results concerning the unfired samples of Figs. 5 and 6. One observes that the thermal activation energy of all fired quartz samples after the thermal quenching correction is very similar $E \sim 0.4$ eV. This E value is very similar to the activation energy for the unfired quartz samples before applying the thermal quenching correction (Fig. 5b). On the other hand, when the thermal quenching correction is applied to the unfired samples, the result is an increase of the energy values to $E \sim 1$ eV (Fig. 6). Obviously different assumptions during the analysis lead to very different conclusions. Fig. 8 seems to clearly support the presence of thermal quenching for fired samples, while the presence of thermal quenching for unfired samples is not supported by the data in Fig. 5, in which the OSL signal from VDT does not decrease at high stimulation temperatures.

The fundamental questions concerning the OSL signals from VDT which remain unanswered are:

- (a) Is thermal quenching present in both fired and unfired samples?
- (b) If thermal quenching is indeed present in both fired and unfired samples, are the thermal quenching parameters C , W the same?
- (c) More generally, is the thermal quenching effect universal in quartz?
- (d) Does firing of the samples affect the thermal quenching properties and parameters of the samples?

Further experimental work is necessary to establish the C , W parameters for the OSL signals from VDT.

It is well known that firing of quartz samples at high temperatures causes changes to both their sensitivity and their superlinearity properties [11–14]; and references therein. Our study suggests that firing of quartz samples to 800 °C may also cause alterations of their thermal quenching properties.

3.2.2. OSL from VDT as a function of irradiation dose

Based on the results of Fig. 8, two of the four samples were selected in order to study the dose response of the OSL signals from VDT. The SLE and INK samples showed maximum OSL response at 180 and 200 °C correspondingly. The dose dependence of the OSL signals originating from VDT was studied using the following multiple aliquot protocol.

- Step 1: Test dose $T_D = 1$ Gy and TL readout up to 250 °C.
- Step 2: Given dose $D_i = 0.5, 1, 2, 3.5, 5, 10, 25, 50, 80, 130, 180, 230$ and 300 Gy.
- Step 3: TL readout up to 500 °C.
- Step 4: T_D and measurement of TL up to 500 °C, to test sensitivity changes from step 1.
- Step 5: Blue CW-OSL at a stimulation temperature $T = 200$ °C for sample INK and $T = 180$ °C for sample SLE, for 2000 s.
- Step 6: Residual TL readout up to 500 °C.
- Step 7: T_D and TL up to 500 °C, to test sensitivity changes from step 4.
- Step 8: Repeat steps 1–7 for a new aliquot and a new dose D_i .

Step 1 is used as a normalization procedure for this multiple aliquot protocol, in addition to the equal masses being used for all aliquots. The OSL curve shapes are those shown in Fig. 7. In the previous section these OSL curves were treated as two separate parts, by considering signals in the intervals 0–300 s, and 300–2000 s. However, for the needs of the OSL dose response study and in order to get the maximum possible information on the OSL curve structure, all the OSL curves were analyzed further using a computerized deconvolution procedure.

Initially the CW-OSL curves were transformed into pseudo-linearly-modulated OSL (LM-OSL) peak shaped curves, by using the transformation method of Bulur [15]. This is achieved by transforming the real time variable t into a pseudo time variable $u(t)$ given by

$$u(t) = \sqrt{2 \cdot t \cdot T} \quad (2)$$

Furthermore, the real intensity CW-OSL intensity $OSL(t)$ is transformed into a pseudo intensity $OSL(u)$ according to:

$$OSL(u) = \frac{u(t) \cdot OSL(t)}{T} \quad (3)$$

with T being the total stimulation time. The CW-OSL curves transformed into pseudo LM-OSL ones are shown in Fig. 10 as a function of dose for the case of SLE quartz. The results for INK quartz are exactly similar. All resulting pseudo LM-OSL curves were analyzed in order to find the number of components comprising the signal. This

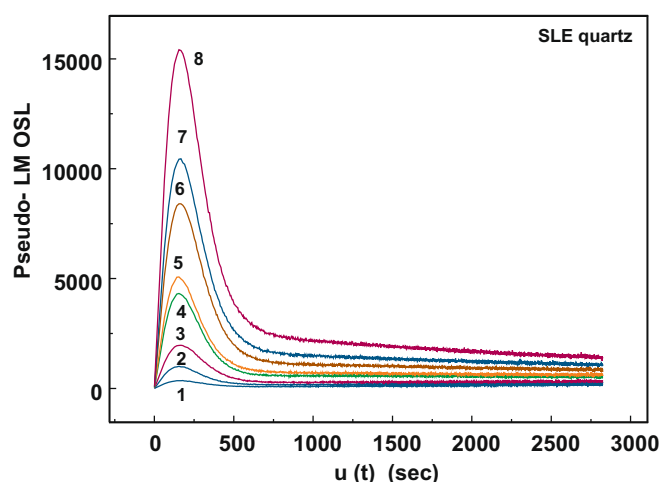


Fig. 10. Pseudo-LM-OSL curves of SLE quartz for various doses. Curves 1–7 correspond to 5, 10, 25, 50, 130 230 and 300 Gy correspondingly.

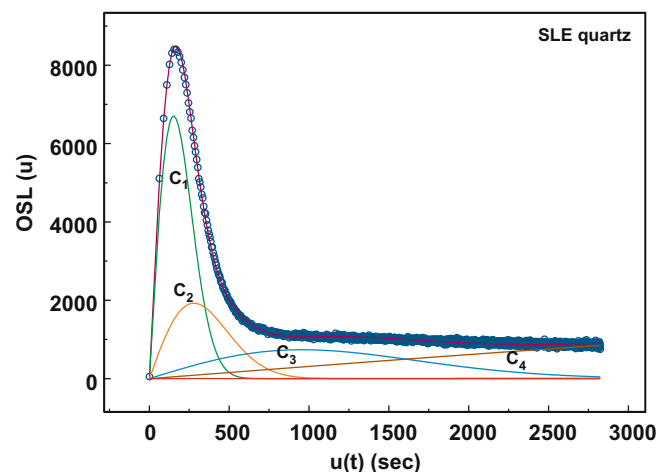


Fig. 11. Example of a pseudo LM-OSL curve analyzed into its individual components.

was achieved by a Computerized Curve Deconvolution Analysis (CCDA) Kitis and Pagonis [16].

A characteristic example of an analyzed pseudo LM-OSL curve is shown in Fig. 11. As seen in this figure, four first order components are needed for the analysis. Three of them labeled as C_1 , C_2 and C_3 are resolved completely in Fig. 11, whereas the partially resolved component C_4 is part of a component with a very low bleaching cross-section. The mean values of the pseudo-LM-OSL maximum, u_m , of the three components derived from the CCDA analysis of the experimental curves from all doses used, are as follows (in seconds): SLE quartz, $C_1(148 \pm 5.5)$, $C_2(271 \pm 18)$ and $C_3(940 \pm 42)$. INK quartz $C_1(143.8 \pm 4)$, $C_2(307.3 \pm 3.3)$ and $C_3(940.6 \pm 48)$. According to routine OSL terminology, component C_1 can be termed fast, the C_2 medium and the C_3 slow. From Eq. (3) the real integral $OSL(t)$ of each component is evaluated. This integral is then plotted as a function of irradiation dose, resulting in the OSL dose response curves given in Fig. 12.

In the case of SLE quartz there is an excellent linear OSL dose behavior in the whole dose region, which is the same for all three components C_1 , C_2 and C_3 . In the case of INK quartz all components have the same behavior, but the OSL signal is sublinear at low doses, then it becomes linear and finally saturation starts above

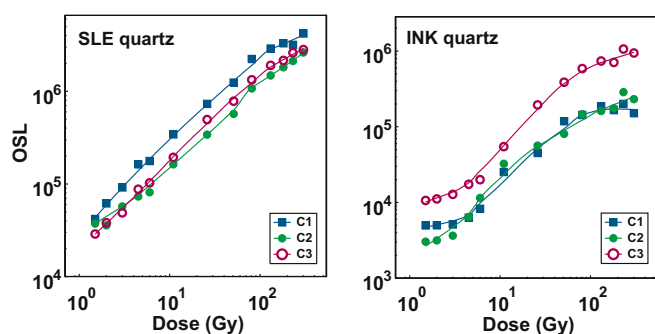


Fig. 12. Dose response of the OSL components for SLE and INK quartz samples.

100 Gy. Concerning the fourth not fully resolved component C_4 , it exhibits also a relatively well-defined dose response behavior with a dose threshold between 25 and 50 Gy. To the best of our knowledge, the results of Fig. 12 are the only ones in literature reporting such a clearly resolved behavior of the dose response of VDT in quartz.

The present work has important implications for dosimetry and dating. The lowest dose used in the protocol was 0.5 Gy and a test dose of 1 Gy is used before the OSL measurement, so that the lowest dose delivered to the VDT traps is 1.5 Gy. It was found, however, that even at this low dose the intensity of component C_1 (and to a less amount of C_2) is quite high. This means that the real Lowest Dose Detection Limit (LDDL) of OSL signals from VDT is lower than 1.5 Gy. This is a very low dose detection threshold, and deserves further investigation in quartz from fired ceramics. The reason is that the conventional TL/OSL dating methods usually cannot evaluate accurately doses from fired ceramic less than 3 Gy. Assuming a mean annual dose rate of 4×10^{-3} Gy/year, the lower age limit is about 700 years. However, if the doses of the order of 0.5 Gy can be measured using the OSL signals from VDT studied here, the age limit could be reduced perhaps to about 100 years.

4. Conclusions

Under the combined action of thermal activation and optical stimulation electrons trapped at VDT are liberated into the conduction band and recombine emitting photons. Our study showed that all eight unfired quartz samples of various origins exhibit OSL signals from VDT, and for a wide region of OSL stimulation temperatures. It was found that the OSL signal from VDT depends strongly on the type of quartz sample studied, and on whether the sample was previously fired at high temperatures or not.

In the case of unfired samples the OSL from VDT increases continuously with the stimulation temperature up to 375 °C, and seems to be unaffected by thermal quenching effects. When the data is analyzed assuming no thermal quenching effects, the process is described by a thermal activation energy of $E \sim 0.4$ eV. However, analysis of the data assuming the presence of thermal

quenching leads to much higher apparent activation energy $E \sim 1.0$ eV.

In the case of fired quartz samples the OSL from VDT initially increases with the stimulation temperature, and subsequently decreases at high stimulation temperatures, supporting the presence of thermal quenching effects. Data analysis assuming the presence of thermal quenching effects yields a value of the activation energy $E \sim 0.4$ eV for all eight fired quartz samples.

The OSL signal from VDT in quartz samples fired at 800 °C for 1 h is very high, and consists of three well-defined components and a fourth slow component which is rather poorly resolved. The three well-defined components C_1 , C_2 and C_3 which were termed fast, medium and slow, exhibit the same behavior as a function of beta dose between 0.5 and 300 Gy. The dose response behavior of these components deserves further investigation in ancient fired ceramics, since potentially their study could extend appreciably the equivalent dose region toward both lower and higher values.

References

- [1] R.M. Bailey, Towards a general kinetic model for optically and thermally stimulated luminescence of quartz, *Radiat. Meas.* 33 (2001) 17–45.
- [2] R.M. Bailey, Paper I – simulation of dose absorption in quartz over geological timescales and its implications for the precision and accuracy of optical dating, *Radiat. Meas.* 38 (2004) 299–310.
- [3] G. Adamiec, A. Bluszcz, R. Bailey, M. Garcia-Talavera, Finding model parameters: genetic algorithms and the numerical modelling of quartz luminescence, *Radiat. Meas.* 41 (2006) 897–902.
- [4] V. Pagonis, A.G. Wintle, R. Chen, X.L. Wang, A theoretical model for a new dating protocol for quartz based on thermally transferred OSL (TT-OSL), *Radiat. Meas.* 43 (2008) 704–708.
- [5] J.S. Singarayer, R.M. Bailey, Further investigations of the quartz optically stimulated luminescence components using linear modulation, *Radiat. Meas.* 37 (2003) 451–458.
- [6] M. Jain, A.S. Murray, L. Bøtter-Jensen, Characterization of blue-light stimulated luminescence components in different quartz samples: implications for dose measurement, *Radiat. Meas.* 37 (2003) 441–449.
- [7] N.G. Kiyak, T. Canel, Equivalent dose in quartz from young samples using the SAR protocol and the effect of preheat temperature, *Radiat. Meas.* 41 (2006) 917–922.
- [8] A.G. Wintle, Thermal quenching of thermoluminescence in quartz, *Geophys. J. R. Astron. Soc.* 41 (1975) 107–113.
- [9] S.W.S. McKeever, L. Bøtter-Jensen, N. Agersnap Larsen, G.A.T. Duller, Temperature dependence of OSL decay curves: experimental and theoretical aspects, *Radiat. Meas.* 27 (1997) 161–170.
- [10] L. Bøtter-Jensen, S.W.S. McKeever, A.G. Wintle, *Optically Stimulated Luminescence Dosimetry*, Elsevier, 2003, p. 77.
- [11] R. Chen, X.H. Yang, S.W.S. McKeever, The strongly superlinear dose dependence of TL in synthetic quartz, *J. Phys. D: Appl. Phys.* 21 (1988) 1452–1457.
- [12] C. Charitidis, G. Kitis, C. Furetta, S. Charalambous, Superlinearity in synthetic quartz: dependence on the firing temperature, *Nucl. Instr. Meth. Phys. Res. B* 168 (2000) 404–410.
- [13] G. Kitis, E. Kaldoudi, S. Charalambous, Thermoluminescence dose response of quartz as a function of irradiation temperature, *J. Phys. D: Appl. Phys.* 23 (1990) 945–949.
- [14] G. Polymeris, G. Kitis, V. Pagonis, The effects of annealing and irradiation on the sensitivity and superlinearity properties of the 110 °C thermoluminescence peak of quartz, *Radiat. Meas.* 41 (2006) 554–564.
- [15] E. Bulur, A simple transformation for converting CW-OSL curves to LM-OSL curves, *Radiat. Meas.* 32 (2000) 141–145.
- [16] G. Kitis, V. Pagonis, Computerized curve deconvolution analysis for LM-OSL, *Radiat. Meas.* 43 (2008) 737–741.