BJRS

BRAZILIAN JOURNAL
OF
RADIATION SCIENCES
10-02A (2022) 01-11

Effect of blue light bleaching in a high TL sensitivity natural quartz crystal

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ABSTRACT

The quartz dating can be performed from optically stimulated luminescence (OSL) or thermoluminescence signals (TL). The relationship between OSL and TL signals is important to ensure adequate measurement of luminescent signals. The bleaching effect on TL signals has been studied by various works. However, there is no consensus. This study investigates the effect of blue light bleaching in a natural quartz crystal whose TL signal can be recorded with test doses lower than 1 Gy. As a result, it was found that blue light is capable to erode almost completely de first TL peak. The bleaching effect for TL peaks above 200 °C is limited. However, TL components that peaked at 320 and 390 °C are more sensitive to blue light than the intermediate TL components peaking at 250 and 280 ˚C.

Keywords: **thermoluminescence, quartz, sensitization, blue light bleaching.**

ISSN: 2319-0612
DOI: https://10.15392/bjrs.v10i2A.2027
Submitted:2021-03-07
Accepted: 2022-05-10

1. INTRODUCTION

Over the last few decades, thermoluminescence (TL) and optically stimulated luminescence (OSL) dosimetry techniques have been widely applied to determine the equivalent dose in quartz grains [1,2]. To ensure an adequate measurement of the luminescent signal, it is important to know some signal characteristics. One of those is the relationship between OSL and TL signals [3]. Quartz OSL signal commonly decays non-exponentially, however it is possible to be fitted as a sum of exponential decays (components) [4]. Quartz dating (e. g., SAR (single-aliquot regenerative-dose) and MAR (multiple-aliquot regenerative-dose) protocols) is usually performed using the fraction of the OSL signal termed as fast component [3], which is more rapidly bleached by light exposure. In a detailed investigation carried out in quartz grains prepared for a sedimentary sample, Spooner [5] concluded that the fast OSL component and the 325 °C TL peak originate from the same set of electron traps.

As known, the models proposed by Bailey [6,7] for quartz luminescence have been widely used to simulate experimental TL and OSL results and to predict the stability of OSL signal over time [2]. One of the main assumptions of these models is the shared electron trap for the fast component of the OSL signal and the TL peak at 325 °C. To make the authenticity of this assumption more general, Kitis et al. [8] investigated the correlation between the fast component of the OSL signal and the TL peak at 325 °C in sedimentary quartz samples from nine different provenances. They did not observe a specific correlation between those signals. Thus, further studies are needed to advance the understanding of the relationship between TL and OSL signals from quartz.

In the previous studies the dose used to obtain TL signals was in the order of several Gy [8,9]. For example, Kitis et al. [8] used a test dose of 100 Gy. Previously, Khoury et al. [10] described a procedure combining high-dose irradiation and moderate thermal treatments to sensitize a peak in the 250-300 ˚C range, which is sensitive for test-dose as low as 1 mGy. To explore this issue, this work aims to investigate the effect of blue light bleaching on the whole TL signal of a natural quartz crystal with a TL sensitivity for test-doses much lower than 1 Gy.

2. MATERIALS AND METHODS

2.1. Sample pellets

The quartz crystal used in this study is originated from a pegmatite named Mina dos Cavalos (MC) located in the municipality of Solonópole (Ceará, Brazil). Fragments of the MC crystal were manually crushed using an agate mortar and pestle. The quartz grains were classified in 75 x 150 µm particle size range using stainless steel sieves. To remove the effect of the previous dose on the quartz grains, the grains were submitted to a thermal treatment (400 °C; 60 min). The quartz grains used to prepare the samples termed as zeroed were separated in this stage. Then, the grains were sensitized by the procedure proposed by Khoury et al. [10]. The procedure comprised irradiation at room temperature with a dose of 30 kGy of gamma radiation (^{60}Co) in a gamma cell irradiator (1.9 $kGy.h^{-1}$) and submitted to three cycles of thermal treatment at 400 °C for 1 hour with 24 h intervals between each cycle. The sensitized samples were prepared with quartz grains submitted to the sensitization procedure previously described.

The quartz pellets were manufactured following the procedure employed by Carvalho Junior et al. [11]. This procedure consisted of pressing a homogeneous mixture of quartz particles and flocculated PTFE (polytetrafluoroethylene) at room temperature using a die manufactured using stainless steel. The ratio of quartz and PTFE in the mixture was 1:1 by mass. The compressive force (~10 kN) was applied during 10 s, using a SPECAC 15T hydraulic press. The pellets are discs of 6 mm diameter, 1 mm thick and weigh approximately 50 mg. Figure 1 shows quartz pellets used in this study. Ten pellets were used, five for each of the conditions (zeroed and sensitized).

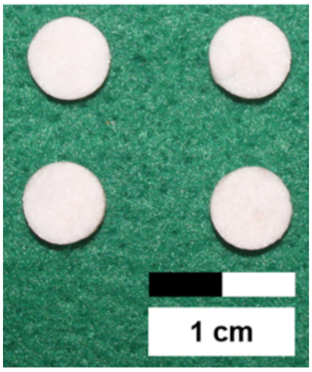

Figure 1: Quartz pellets produced with 75 x 150 µm quartz grains.

2.2. TL measurements and optical stimulation

The glow curves were recorded from 25 to 400 °C (2 °C.s^{-1}) with an automated *Lexsyg* SMART reader equipped with $^{90}Sr/^{90}Y$ source (~63 mGy.s^{-1} at sample position). Table 1 shows optical and thermal conditions used to carry out three different bleaching experiments before the TL measurements. Optical stimulations were performed in continuous wave (CW-OSL) mode with blue LEDs (465 ± 5 nm; 80 mW.cm^{-2}) under two thermal conditions: (i) at room temperature and two stimulation times (160 and 480 s) and (ii) preceded by preheating (200 °C; 10 s) and thermally assisted (125 °C). For each condition, the measurements were performed in five samples with a test dose of 500 mGy.

Table 1: Optical and thermal conditions used to carry out different bleaching experiments.

condition/experiment	preheat	thermal assistance	stimulation time
b1	200 °C (10 s)	125 °C	160 s
b2	-*	room temperature (RT)	160 s
b3	-*	room temperature (RT)	480 s

* Without preheating to eliminate the effect of thermal stimulation.

2.3. Glow curve deconvolution

The method of glow curve deconvolution was used to assess the effect of blue light bleaching in each TL component on the whole TL curve. For this, the *tgcd* package [12] of the *R* programming language [13] was used to perform the glow curve fitting. The glow curves were fitted using the first-order equation proposed by Bos et al. [14]. The number of TL components was determined considering the results obtained by Caicedo Mateus et al. [15] and the assessment of the resolution parameter proposed by Kitis and Pagonis [16] in unbleached TL signals. This procedure defined the number (minimum) of seven components to fit the whole glow curve. The fitting procedure comprised the fixing of the activation energy (E) assuming a distribution of trapping energies as proposed by Hornyak, Chen and Franklin [17]. The reliability of the fitting was assessed by the figure of merit (FOM) provided by *tgcd*, which is defined as:

$$FOM = \sum |Y_{exp} - Y_{fit}|/A \quad \textbf{(1)}$$

Where Y_{exp} is the experimental glow curve, Y_{fit} is the deconvoluted curve and A is the area of the deconvoluted curve.

3. RESULTS AND DISCUSSION

Figure 2 shows typical TL (a) and CW-OSL (b) signals from powdered quartz in zeroed and sensitized conditions. For the TL signal in zeroed (unsensitized) condition, it is possible to observe an intense peak at ~100 °C and a series of low-intensity peaks above to 200 °C, especially the characteristic quartz TL peak at 325 °C. For the sensitized signal, it is possible to observe the sensitized TL peak (~275 °C) already reported by Guzzo et al., Khoury et al., and Carvalho Junior et al. [9,10,11]. For the samples used in this work, the intensity of TL peaks above 200 °C is ~25 times higher for the sensitized condition. Comparing the OSL signals shown in Fig. 2 (b), it is possible to observe that the signal of the zeroed sample has the same intensity as the background signals, which shows the effect of the sensitization procedure on the OSL signal. Ferreira de Souza et al. [18] also reported the sensitization of the OSL signal in single crystal samples from two provenances, one of them the MC pegmatite. The higher TL intensity observed in the first peak for zeroed condition is probably due to thermal activation. Chen and Pagonis [19] reported an increase in sensitivity to thermal treatments at this temperature (400 °C) for the 110 °C TL peak.

Figure 3 shows TL curves for unbleached (full TL signal) and the different bleaching conditions. In Figure 3 (a) the first TL peak is removed by a preheat (200 °C; 10 s) and optical stimulation at 125 °C. It is possible to observe that blue light does not specifically affect the region of glow curve above 200 °C. Comparing the signals shown in Fig. 3 (b) it is possible to observe that bleaching at RT resulted in a progressive reduction of the first TL peak with the increase of the time of stimulation. Smith and Rhodes [20], using ^{40}Ar laser (514 nm; 2.1 mW.cm^{-2}), observed an initial increase of the first peak followed by a reduction in its intensity. For a stimulation time of 160 s with experimental arrangement in this study, the energy deposited by blue light is ~6 times the used by Smith and Rhodes [20] with 1000 s. For this reason, it is not possible to be sure if the signal observed at the first TL peak is PTTL (phototransferred thermoluminescence) or the remaining TL. For TL peaks above

200 °C, the different bleaching resulted in similar glow curves; similar to the result obtained by Smith and Rhodes [20] that did not observe signal reduction due to bleaching with times greater than 1000 s. For this region, the intensity reduction of signal when compared with unbleached signal was ~22% on average to the different bleaching. This reduction is like that got by Kitis et al. [8] (10-20%) who used a test dose 200 times higher and optical energy deposited in the sample 6 times lower. This result suggests that optical stimulation has a limited capacity for depleting charges from the traps responsible for the TL peaks above 200 °C.

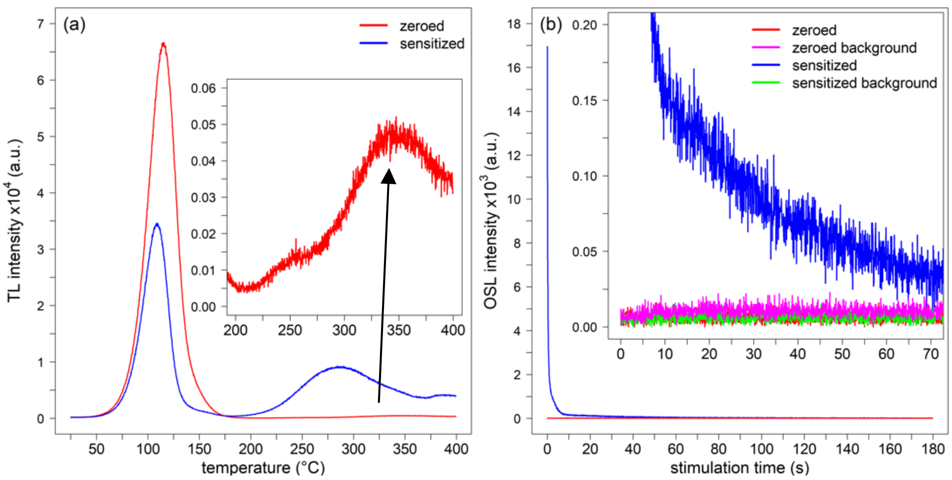

Figure 2: *Typical TL (a) and CW-OSL (b) signals from quartz pellets in zeroed and sensitized conditions. TL: test-dose: 0.5 Gy; heating rate: 2 °C.s^{-1} . CW-OSL: test-dose: 30 Gy; preheat: 200 °C (10 s); thermal assistance: 125 °C. The background OSL signals are also shown.*

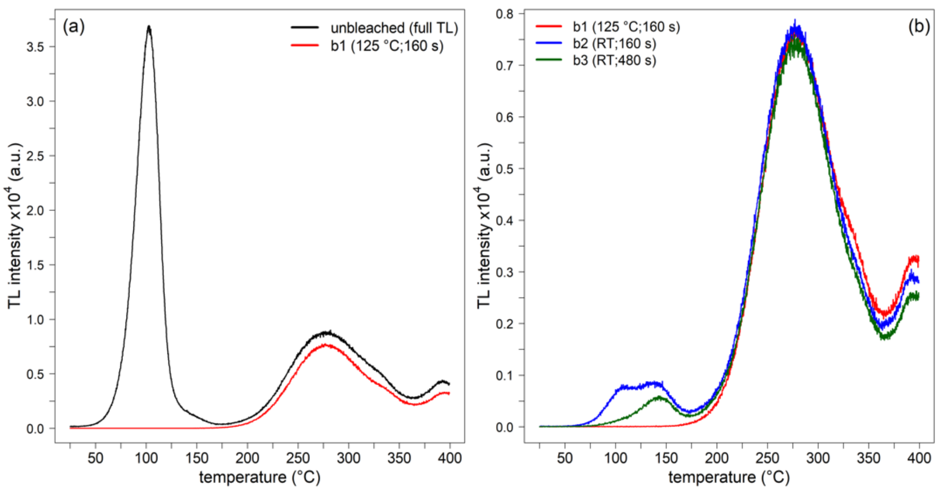

Figure 3: *Unbleached (full TL) and b1 TL signals (a) and TL obtained after different bleaching conditions (b). Test-dose: 0.5 Gy*

To progress with this investigation, the glow curves were deconvoluted to observe the bleaching effect on each TL component. Figure 4 shows typical results of fitted glow curves composed of seven first-order components for unbleached (a) and bleached (b) conditions. FOM values were always better than 3.20%.

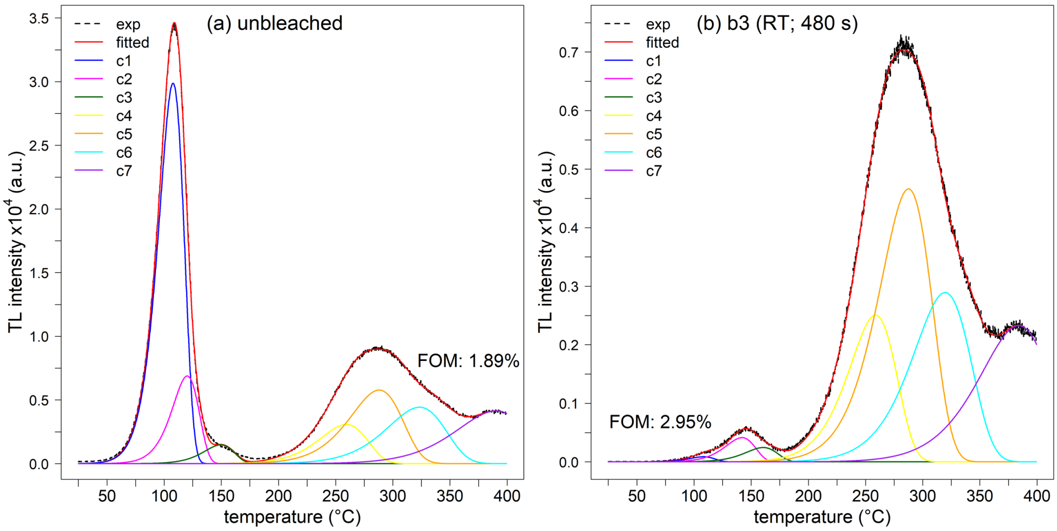

Figure 4: *Typical results of deconvolution into seven components for unbleached (a), and b3 (RT; 480 s) (b).*

Table 2 shows the averages values for peak maximum intensity (I_m) and temperature of the maximum peak intensity (T_m) of components obtained from bleached and unbleached signals. It is possible to observe that the first TL peak comprises three components: c1, c2 and c3. For all specimens, the c1 was the most intense component. The sensitized peak is compound of c4, c5 and c6 components. The component c7 describes a high-temperature peak. This peak was previously observed by Caicedo Mateus et al. [15] in a single crystal sample of quartz from the same provenance and it was associated with deep traps filled during the sensitization process.

Table 3 shows the residual TL intensity by component for different bleaching conditions. The residual TL intensity was calculated using the ratio of bleached and unbleached component intensity. It is possible to observe that c1 is the most affected component related to the first peak. This component did not show any residual signal after 480 s of optical stimulation at room temperature. In turn, c2 and c3 components also showed a progressive and significant intensity reduction of approximately 91% for c2 and 86% for c3. This result suggests that c2 and c3 components can be completely bleached by prolonged optical stimulation. For the sensitized peak, c4 and c5 components are less affected than c6 (associated with the 325 °C peak) component. The signal intensity reduced by approximately 35% for the c6 component, while for c4 and c5 it was a lower decrease by approximately 12%. The c7 component also shows a significant intensity reduction of approximately 32%. This result suggests that the TL peak at 110 °C and the highest temperature peaks at 320 and 390 °C are more sensitive to blue light.

Table 2: Average values for maximum peak intensity (I_m) and temperature of the maximum peak intensity (T_m) of components obtained by deconvolution from bleached and unbleached signals.

component	**unbleached**		**b1 (125 °C; 160 s)**		**b2 (RT; 160 s)**		**b3 (RT; 480 s)**	
	T_m (°C)	**I_m**	**T_m (°C)**	**I_m**	**T_m (°C)**	**I_m**	**T_m (°C)**	**I_m**
c1	102 ± 4	32806	-	-	101 ± 3	524	105 ±4	92
c2	115 ± 4	6253	-	-	126 ± 4	568	137 ± 4	474
c3	142 ± 5	1474	-	-	147 ± 4	569	157 ± 4	189
c4	251 ± 6	3354	252 ± 4	2975	249 ± 4	3133	250 ± 7	2874
c5	280 ± 6	5673	280 ± 6	4705	279 ± 4	5236	279 ± 6	4939
c6	317 ± 5	4630	317 ± 8	3059	315 ± 4	3182	315 ± 5	2919
c7	394 ± 9	3931	389 ± 7	2871	394 ± 9	2620	391 ± 9	2256

Table 3: Residual intensity for each TL component for different bleaching conditions.

	Residual TL intensity		
TL component	**b1 (125 °C; 160 s)**	**b2 (RT; 160 s)**	**b3 (RT; 480 s)**
c1	-*	0.02	0.00
c2	-*	0.10	0.09
c3	-*	0.40	0.14
c4	0.89	0.93	0.84
c5	0.83	0.92	0.87
c6	0.66	0.68	0.63
c7	0.78	0.65	0.60

* The components were completely erased by preheating (200 °C; 10 s) and thermal assistance (125 °C).

4. CONCLUNDING REMARKS

For the high sensitive MC natural quartz crystal, it was concluded that the blue light bleaching affected all TL peaks regardless of the thermal conditions used. The deconvolution of TL signals showed some components were more affected by blue light. For the 110 °C TL peak, we observed a significant depletion of this peak, especially the component c1. Exposure during 160 s at room temperature almost erased completely the signal from the c1 component. For the TL peaks above 200 °C, we observed no significant difference in glow curves from the three bleaching conditions. For all bleaching conditions, the reduction in TL intensity was ~22% when compared with the unbleached signal. This result suggests that only part of the traps associated with those TL peaks are optically sensitive. For the TL peaks above 200 °C, the TL components c6 (320 °C) and c7 (390 °C) showed greater depletion than c4 (250 °C) and c5 (280 °C).

ACKNOWLEDGMENT

This work was supported by FACEPE – *Fundação de Amparo à e Tecnologia do Estado de Pernambuco* [APQ-0648-3.09/14] and CNPq – *Conselho Nacional de Desenvolvimento Científico e Tecnológico* [P: 306374/2018-8]. BRS is grateful to FACEPE [IBPG 0231 3.09/18] and CAPES [88887.596316/20] for the scholarship provided.

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