



# Comparing the monochromatic TL response of a high sensitivity natural quartz irradiated with β and γ rays

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## ABSTRACT

This study investigates the effect of the dose-rate in the thermoluminescent glow curves of a single crystal of quartz. The samples were sensitized by high dose of  $\gamma$  radiation combined with heat-treatments. The glow curves were registered in zeroed (unsensitized) and sensitized conditions using an optical filter centered in violet spectral region. Tens mGy test doses were administered with one  $\beta$  ( $^{90}$ Sr/ $^{90}$ Y) source and two  $\gamma$  radiation sources ( $^{60}$ Co and  $^{137}$ Cs). The TL curves were deconvoluted using a first-order kinetic model. Differences in the glow curve patterns and trapping parameters were observed between zeroed and sensitized samples. Differences were found in the TL curves comparing the three radiation sources. The principal variation is the remarkable increase in the TL signal above 350 °C, which is observed only in sensitized samples with the minor dose-rate source ( $^{137}$ Cs). This signal seems to be associated with deep trapping states. The intensities of the components defining the first peak and the high temperature signal show a dependence on the dose-rate. The dose-rate dependence of the first-peak components is explained by the competing effects that may take place during the excitation stage. The components that fitted the sensitized peak (~260 °C) do not exhibit a clear dependence on the dose-rate of radiation source.

Keywords: thermoluminescence, quartz, sensitization, dose-rate, deep trap

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#### **1. INTRODUCTION**

Quartz is among the most useful minerals for thermoluminescence (TL) dating. This is due to their abundance in sediments at most geological settings, as well as their ability to fulfill the requirements of sensitivity to radiation dose, resistance to weathering, and relatively well investigated luminescence properties [1]. Due to these features, most research has focused on the development of procedures to determine the palaeo- or equivalent dose [2]. Since the introduction of quartz-inclusion technique and the pre-dose effect [3], and due to the automatic devices usually including a  $\beta$  radiation source, these sources have commonly been used in quartz dating protocols even though  $\gamma$  rays are the main component of natural radiation [4].

Thermoluminescence is a technique that has applications in both retrospective dosimetry and dating. To determine the  $\gamma$  ray equivalent natural dose in TL dating, it is necessary to measure the TL response per dose. In general, the dating protocols assume that laboratory radiation sources, with relatively high dose-rates compared to natural radiation, have similar efficiency to generate the latent TL signal [5]. For instance, the natural dose-rate to which the sample is exposed is approximately  $3x10^{-11}$  Gy/s, whereas the laboratory dose-rate corresponds to several Gy/s. Thus, the dating protocols employed to determine the equivalent dose are usually performed at a higher dose-rate than those found in geological and archaeological sites [6,7]. However, it is well accepted that further efforts are required to progress in the understanding of the dependence of quartz TL signal with dose-rate and radiation sources [6,8].

The overall analysis of dose-rate effects in some luminescent materials suggested that the TL response as a function of the dose is not completely independent from the dose-rate. In the case of quartz, it was also noticed that the TL emissions related to high temperature glow peaks change differently with the increase of the dose-rate [7,9]. Another study showed that the variation of the dose-rate from  $1.4 \times 10^{-3}$  to 3.3 Gy/s caused a decrease in TL signal in a factor of five in powdered samples of Brazilian quartz irradiated with  $^{60}$ Co  $\gamma$  rays [5]. A model for dose-rate dependence of TL intensity, including the stage of heating phase of TL readout, considering one trapping state and two recombination centers, showed that dose-rate effects may occur for two TL peaks, where one peak may increase with the dose-rate whereas the other decreases [7]. Valladas and Ferreira [9] found different behaviors for the TL emission at three components (UV, blue and green) giving the same

dose with two different dose-rates. For the blue component, the authors observed that the low doserate yielded more TL signal than the high one, and the UV and green components exhibited a higher intensity for high dose-rate. In contrast, an increase in the TL signal was observed using dose-rates from  $2x10^{-5}$  to  $2x10^{-2}$  Gy/s in milky Brazilian quartz irradiated with <sup>60</sup>Co  $\gamma$  rays [10].

This brief synthesis made clear that further studies are required to understand the dependence of the TL signal of quartz on dose-rate effect using different radiation sources. To progress in this direction, the effect of the dose-rate was investigated in a single crystal showing a high TL sensitivity above 200 °C, with test-doses as lower as 63 mGy. To achieve this, the samples in zeroed and sensitized conditions were irradiated with one  $\beta$  radiation source and two  $\gamma$  sources, with a range of dose-rate from 63 to 0.008 mGy/s. The TL glow signals were obtained with an optical setup to collect TL emissions in the violet region.

#### 2. MATERIALS AND METHODS

#### 2.1. Samples and sensitization

The natural quartz used in the present study corresponds to a single crystal extracted from one deposit located in the district of Solonópole (Ceará State, Brazil), which was used in previous studies [11-13]. It was a semi-euhedral and lightly smoky specimen with two defined prismatic (*m*-) faces. Six samples measuring  $5\times5\times1$  mm were prepared from plates cut parallel to the crystal plane ( $10\overline{10}$ ). Each sample was lapped and optically polished with Al<sub>2</sub>O<sub>3</sub> abrasives and cleaned in acetone. Previously, optically stimulated luminescence (OSL) measurements with stimulation of blue LEDs produced partial desensitization in the samples [11] and the batch of samples were submitted to several test-dose irradiation and heating cycles [12,13]. Therefore, to reuse the samples in the TL measurements, it was necessary to define a new starting point by submitting all samples to a prolonged heat treatment. For this purpose, the samples were heat-treated at 700 °C for 3 h in a muffle furnace at atmospheric pressure, with a heating rate of 5 °C/min, and then coolled up to room temperature inside the furnace. This condition is defined as the zeroed condition (Z). This temperature value used to clean-up the residual signals is in accordance with the isochronal annealing experiments carried out in samples of the same crystal [14].

Three samples at zeroed condition were resensitized (ReS) using the procedure described by Khoury et al. [15]. For this, the samples were irradiated at room temperature with 30 kGy with <sup>60</sup>Co ( $\gamma$  rays) in a  $\gamma$  cell irradiator (dose-rate of 2.14 kGy/h) and subsequently submitted to three cycles of heat treatment at 400 °C for 1 h using the heating and cooling rates mentioned above. The three remaining (unsensitized) samples were kept in the zeroed (heat-treated at 700 °C) condition.

#### 2.2. Test-dose irradiations and TL measurements

The TL glow curves were recorded from 25 to 425 °C (heating rate 2 °C/s) using an automated *Lexsyg* SMART reader equipped with a Hamamatsu H7360-02 bi-alkaline photomultiplier tube (PMT) and with an internal  $\beta$  particle source ( ${}^{90}$ Sr/ ${}^{90}$ Y). The TL curves were acquired with a combination of interference filters, Schott-BG 39 and AHF-HC 414/46, that corresponds to a violet detection window centered in the 411(51) nm. This filter set is the most adequate to register the TL signals in the whole temperature range for this sample [16].

To characterize the differences in the glow curves between Z and ReS samples, three radiation sources were employed, two of them are  $\gamma$  radiation and one is  $\beta$  ray source. In dosimetry, linear energy transfer (LET) is defined as the amount of energy that an ionizing particle transfers to the material per unit of crossed path. In other words, the LET describes the rate at which the energy is transferred per unit length of tracking (keV/µm).  $\beta$  particles (0.3 keV/µm) and  $\gamma$  rays (0.2 keV/µm) are low-LET radiation as compared with alpha particles (50-200 keV/µm), which are high-LET radiations [17]. The specific information of the test-dose of each radiation source is summarized in Table 1.

Source radiation	Dose-rate (mGy/s)	Irradiation time (s)	Test-dose (mGy)	Energy (MeV)
γ ( <sup>137</sup> Cs)	0.008	1228	10	0.661
γ ( <sup>60</sup> Co)	0.299	33	10	1.173
β ( <sup>90</sup> Sr/ <sup>90</sup> Y)	~63	1	63	0.546

 Table 1: Radiation source and test-dose used for the TL measurements.

Before each TL measurement, in order to eliminate the residual signal of the previous TL reading, the samples were annealed at 400 °C for 1 h. To avoid the thermal fading of the so-called 110 °C glow peak, the  $\gamma$  irradiated samples were put into an ice-bath immediately after the irradiation with the test-dose. The sequence of test-dose irradiation, TL reading, and annealing was repeated three times for each sample. In order to remove the instrumental noise and the incandescence signal contributions, the background was measured after each TL measurement and subtracted from the principal TL signal.

#### 2.3. Glow curve deconvolution

In order to characterize the kinetic parameters of the components of the glow peaks in Z and ReS conditions, the glow curves were scrutinized using a glow curve deconvolution method using the *GlowFit* deconvolution software [18] based on the Randall-Wilkins first-order kinetics model.

Previously, the first order kinetics was satisfactory used to model the TL mechanism giving rise to the quartz glow peak at 110 °C as well as those occurring above 300 °C [19,20]. From the theoretical point of view, Bos [21] assumed that the first order kinetics is appropriate to depict the TL processes that are controlled by localized transitions. On turn, Sunta, Yoshimira and Okuno [22] suggested that the first order kinetics prevails in quartz TL emissions is related to the presence of interactive deep traps. In our previous studies with high sensitive quartz, the TL mechanism was tentatively explained considering the existence of localized transitions of ionic and electronic charges between active electron-hole traps and the existence of deep traps acting ans competitors [14,16]. Due to these reasons, only the first order kinetics was used to model the TL curves in this work.

Since quartz glow curves consist of several overlapping components, several initial fittings were tested making rational guesses to the number of glow peaks without restricting the peak temperature. Based on the TL glow curves obtained with very small test-dose (1 mGy) [16] and the initial set of fits, six components were established as well as the peak temperatures that best fit all TL curves. The deconvolution procedure consisted of the following steps: (i) include the heating rate; (ii) stipulate the initial values of the fit (amount of peaks, temperature associated with the maximum intensity of each peak); (iii) fix the temperature of the peaks except the first peak TL, which could be freely adjusted by *Glowfit*; (iv) restrict the variation of the activation energy with increasing temperature, i.e., the activation energy can only have increasing values with the increasing the peak maxima ( $T_m$ )

of each peak. This protocol is based on the assumption that TL emission is produced only by electron trap transitions [23]. To correct for slight shifts of the peak maxima, due to the thermal lag between the sample and the hot planchet, small corrections were applied to the  $T_m$  and, consequently, to the energy values. The reliability of the fitting was assessed using the figure of merit (FOM) given by the *GlowFit* software and the *Cb* parameter calculated by the mathematical expression suggested by Kitis [24].

#### 3. RESULTS AND DISCUSSION

# 3.1. Effect of ionization radiation: $\beta$ (<sup>90</sup>Sr/<sup>90</sup>Y) vs. $\gamma$ (<sup>60</sup>Co)

Typical TL glow curves of zeroed (Z) and resensitized (ReS) samples registered with  $\beta$  (<sup>90</sup>Sr/<sup>90</sup>Y) and  $\gamma$  (<sup>60</sup>Co) radiation sources are shown in Figure 1. In Figure 1(a), it is observed that the Z samples are characterized by the glow peaks at 100 °C, also called the first peak, and the broad structure of high-temperature peaks, frequently defined as 325 and 375 °C TL peaks. These peaks are usually observed in glow curves of quartz grains separated from sand, granitic rocks, pegmatite deposits, hydrothermal veins as well as synthetic quartz [25].

The Figure 1(b) shows the glow curves for ReS samples. It is observed that the occurrence of the sensitized peak is completely different than the broad signals observed in Figure 1(a). The sensitized peak occurs around 260 °C, as previously observed in samples of the same origin [14]. The sensitization produced additional changes in the glow curves. Comparing Figure 1(a) and 1(b), it is observed that the position of the first peak changed from approximately 100 to 90 °C, as well as a substantial increase in the intensity for the first peak was noticed in ReS samples. Previously, this increase was not observed because the samples were stored at room temperature after the test-dose irradiation [11,15].

In general, for both Z and ReS samples, it is observed that the greatest intensity in the entire TL curve occurs with the  $\gamma$  (<sup>60</sup>Co) radiation compared to  $\beta$  (<sup>90</sup>Sr/<sup>90</sup>Y) rays. The whole curve for Z samples shows the same behavior, i.e., the first peak and a peak centered around 140 °C are more intense for the  $\gamma$  (<sup>60</sup>Co) radiation. For ReS samples (Figure 1(b)), the first peak shows a very similar intensity for both radiation sources, whereas the intensity of the sensitized peak occurring around 260 °C is higher in the glow curve recorded with  $\gamma$  (<sup>60</sup>Co) radiation.



**Figure 1:** Net TL glow signals of zeroed (a) and resensitized (b) samples of quartz crystals registered after test-dose irradiation with  $\beta$  ( ${}^{90}Sr/{}^{90}Y$ ) and  $\gamma$  ( ${}^{60}Co$ ) rays. Detection window: 411(51) nm. Heating-rate: 2 °C/s.

# 3.2. Effect of dose-rate: $\beta$ (<sup>90</sup>Sr/<sup>90</sup>Y) vs. $\gamma$ (<sup>60</sup>Co) vs. $\gamma$ (<sup>137</sup>Cs)

In order to assess the effect of the dose-rate on the behavior of the TL curve, it was decided to use another radiation source available. Therefore, a third  $\gamma$  source (<sup>137</sup>Cs) was employed, which has a dose-rate of 0.008 mGy/s. Figure 2 shows the typical TL glow curves of zeroed (Z) and resensitized (ReS) samples irradiated with the three sources obtained for the same samples. In Figure 2(a), it is possible to observe that the two curves obtained with the  $\gamma$  sources present the same behavior and intensity above 200 °C. The curve resultant from  $\beta$  irradiation shows a lower intensity. In the region of the first peak, the smallest signal resultant from  $\gamma$  irradiation with 0.008 mGy/s is affected by the competing effects of thermal fading (half-life is about 35 minutes at room temperature) of the shallow traps due to the prolonged period of irradiation.

Typical glow curves of ReS samples are shown in Figure 2(b). The first peak occurs in the same position for the three radiation sources, and, similarly as to the zeroed condition, each of them has different intensity. As observed in Figure 2(a), this peak is more intense for the glow curve registered with the <sup>60</sup>Co source, while the lowest intensity was obtained with the other  $\gamma$  source (<sup>137</sup>Cs). Compared with Figure 2(a), the difference between signal intensities decreased suggesting that the thermal fading during irradiation with 0.008 mGy/s is lower for sensitizatized samples. The sensitized peak around 260 °C also shows different intensities for each curve. Comparing the intensities in the

region of 250-300 °C, it is noted that for both the zeroed condition and resensitized condition, the glow curve associated with the  ${}^{90}$ Sr/ ${}^{90}$ Y source shows the lowest intensity.



**Figure 2:** Net TL glow curves of zeroed (a) and resensitized (b) samples of quartz crystals registered after test-dose irradiation with three radiation sources:  $\beta$  ( $^{90}Sr/^{90}Y$ ),  $\gamma$  ( $^{60}Co$ ), and  $\gamma$  ( $^{137}Cs$ ) rays. Detection window: 411(51) nm. Heating rate: 2 °C/s.

In Figure 2, it is observed that the high temperature region of the glow curves was affected with the sensitization procedure because the signals strongly increased above 350 °C. This notable increase in TL signal is observed only in the glow curves for ReS samples with  $\gamma$  (<sup>137</sup>Cs). The glow curves registered with the  $\beta$  (<sup>90</sup>Sr/<sup>90</sup>Y) rays do not show this increment. As noted in Figure 2(b), an increase in TL signal above 350 °C is not observed with the other  $\gamma$  source, being observed only with minor dose-rate of the <sup>137</sup>Cs source. This remarkable increase in the TL signal was previously observed with test-dose of 1 mGy (<sup>137</sup>Cs) in ReS samples and it was associated with deep traps [16]. Thus, this behavior seems to be much more associated with the magnitude of the dose rate than the radiation source itself.

#### 3.3. Kinetic analysis

In order to evaluate the contribution of the glow peaks of Z and ReS samples, it was determined the peak maximum intensities for samples irradiated with  $\beta$  ( ${}^{90}$ Sr/ ${}^{90}$ Y),  $\gamma$  ( ${}^{60}$ Co), and  $\gamma$  ( ${}^{137}$ Cs) radiation sources and test-doses of 63 and 10 mGy, respectively. The results are summarized in Table 2. The comparison of intensities between Z and ReS conditions shows the sensitization of the first peak for

the three sets of TL curves. The first peak in the Z condition is the most intense for the higher doserate  $\gamma$  source (<sup>60</sup>Co). After sensitization, the contribution of the first peak is more significant for the same source of radiation.

No direct comparison can be made between the sensitized peak (~260 °C) and the 330 °C peak, as they correspond to different electronic traps. Therefore, each of them must be studied separately. For the 330 °C peak, the comparison between the intensities registered with the three radiation sources shows that the contribution of the  $\gamma$  radiation is dominant and the highest intensity is observed with the minor dose-rate source (<sup>137</sup>Cs).

The values of the intensities for the sensitized peak are comparable for both  $\gamma$  radiation sources, among which it is highlighted that the <sup>60</sup>Co source (highest dose-rate among  $\gamma$  sources) presents a slightly higher intensity. Thus, both the first peak and the sensitized peak of the TL curve corresponding to ReS condition are more intense for the <sup>60</sup>Co source. It is also observed that the signal registered with the highest dose-rate ( $\beta$  source) is more dominant for the first peak of Z and ReS samples than for the 330 °C and sensitized peak.

TL peak	sample condition	β ( <sup>90</sup> Sr/ <sup>90</sup> Y) (63 mGy/s)	γ ( <sup>60</sup> Co) (0.299 mGy/s)	γ ( <sup>137</sup> Cs) (0.008 mGy/s)
first peak	Z	48.2 ± 17.8	$78.8 \pm 30.3$	$21.4 \pm 7.6$
	ReS	$223.9\pm20.9$	$277.8\pm27.0$	$169.5\pm20.5$
330 °C peak	Z	4.1 ± 1.9	$15.2 \pm 4.5$	$24.6\pm5.1$
sensitized peak	ReS	214.9 ± 17.1	334.2 ± 16.8	$302.2 \pm 17.8$

**Table 2**: TL peak-maximum intensities (a.u.) registered with three radiation sources in zeroed (Z) and resensitized (ReS) quartz crystals. The error corresponds to one standard deviation related to measurements in three different samples. The intensities were divided by the respective test-dose.

The findings of peak intensities at 330 °C and the sensitized peak summarized in Table 2 are in agreement with Valladas and Ferreira [9]. The authors studied rock crystal (hyaline) samples annealed at 600 °C using two radiation sources with dose-rate of  $1.33 \times 10^{-4}$  Gy/s (<sup>137</sup>Cs) and 0.15 Gy/s (<sup>60</sup>Co). The glow curves were recorded using a test-dose of 32 Gy, heating rate of 5 °C/s, and different optical filters (ultraviolet, blue and green filters). It was observed the decrease (increase) of the 320 °C (360 and 370 °C) glow peaks with the higher dose-rate (0.15 Gy/s; <sup>60</sup>Co). The peak at 320 °C was best observed through the use of the blue filter, which has greater intensity with lower dose-rate (1.33 × 10<sup>-4</sup> Gy/s; <sup>137</sup>Cs). The peak at 360 °C observed with the UV filter, is more intense with the higher dose-rate (0.15 Gy/s; <sup>60</sup>Co).



**Figure 3:** Result of deconvolution into six first-order components of TL glow curves of zeroed (a, b, c) and resensitized (d, e, f) quartz samples irradiated with  $\beta$  ( $^{90}Sr/^{90}Y$ ; 63 mGy) and  $\gamma$  ( $^{60}Co$ ,  $^{137}Cs$ ; 10 mGy) sources. Detection window: 411(51) nm. Heating rate: 2 °C/s.

The kinetic analysis of the glow curves was made according to the procedure described in section 2.3. Typical fitting curves are illustrated in Figure 3. For the three radiation sources, the FOM values of the TL curves of ReS samples were always better than 5%, which indicates a good fitting according to the study by Balian and Eddy [26]. For Z samples, the FOM values varied around 10% for both  $\gamma$ 

sources and 7% for  $\beta$  radiation source. These values show the satisfactory fitting and a probable effect due to the scattering of TL intensities.

For the ReS samples, the figures 3(d, e, f) show that the first peak can be adjusted by a single component whereas the sensitized peak is composed of two components. The presence of the 5<sup>th</sup> component at 335 °C was supported by the TL measurements carried out in samples irradiated with test-dose of 1 mGy [16]. Note that for both sources with the highest dose-rate (<sup>90</sup>Sr/<sup>90</sup>Y and <sup>60</sup>Co), the sixth component appears with maxima at approximately 420 °C.

The mean values for the peak temperature and activation energy calculated for all glow curves are shown in Tables 3 and 4, respectively. In Table 3, it can be seen that the peak temperature related to each component exhibits small differences between the three lots for Z samples, with the highest dose-rate  $\gamma$  source (<sup>60</sup>Co) showing the greatest values. For the ReS condition, the peak temperatures are comparable for the three dose-rate sources, except for the last component. The significant difference is related to the peak temperatures of intermediate and last components, which show higher values for the lowest dose-rate source (<sup>137</sup>Cs).

Zeroed				ReS			
c*	β ( <sup>90</sup> Sr/ <sup>90</sup> Y) (63 mGy/s)	γ ( <sup>60</sup> Co) (0.299 mGy/s)	γ ( <sup>137</sup> Cs) (0.008 mGy/s)	β ( <sup>90</sup> Sr/ <sup>90</sup> Y) (63 mGy/s)	γ ( <sup>60</sup> Co) (0.299 mGy/s)	γ ( <sup>137</sup> Cs) (0.008 mGy/s)	
<b>c</b> 1	$98.3\pm3.5$	$98.6 \pm 1.9$	$97.0\pm2.0$	$87.4\pm1.3$	$88.4\pm0.3$	$87.3\pm0.3$	
c2	$126.8\pm2.8$	$128.4\pm0.4$	$125.1\pm0.0$	$129.1\pm4.9$	$130.3\pm2.7$	$130.0\pm0.0$	
c3	$284.8\pm10.8$	$290.9\pm4.5$	$263.4\pm0.0$	$258.2\pm1.3$	$253.0\pm1.3$	$264.1\pm0.1$	
c4	$312.1\pm1.4$	$322.9\pm7.3$	$311.3\pm0.0$	$278.0\pm0.6$	$274.9 \pm 1.4$	$286.2\pm0.1$	
c5	$337.0\pm5.7$	$346.6\pm2.6$	$327.8\pm0.0$	$333.6\pm1.4$	$333.8\pm2.3$	$330.1\pm0.0$	
c6	$385.9\pm3.4$	$389.2\pm0.5$	$379.6 \pm 1.4$	$411.9\pm5.6$	$401.2\pm8.2$	$525.1\pm25.2$	

**Table 3:** Peak temperature (°C) of trap depth obtained by computerized deconvolution of TL glow curves using three radiation sources for zeroed (Z) and resensitized (ReS) samples of quartz crystal.

\* Glow curve component.

The activation energies are summarized in Table 4. For Z condition, slight differences are noticed from one radiation source to another, but these variations are within the measurement uncertainty.

Therefore, it is possible to suggest that in the zeroed condition, the activation energies estimated for the three groups of TL curves are statistically the same.

For ReS condition, the activation energy is statistically the same for the three groups, i.e., the variations are within the uncertainty of measurement, except for the 6<sup>th</sup> component for the lowest dose-rate source (<sup>137</sup>Cs), which has a higher energy value than the other two sources. Thus, for the first five components, the three radiation sources reveal the same charge trapping states.

			quartz cr	ystal.	D C	
		Zeroed			ReS	
c*	β ( <sup>90</sup> Sr/ <sup>90</sup> Y) (63 mGy/s)	γ ( <sup>60</sup> Co) (0.299 mGy/s)	γ ( <sup>137</sup> Cs) (0.008 mGy/s)	β ( <sup>90</sup> Sr/ <sup>90</sup> Y) (63 mGy/s)	γ ( <sup>60</sup> Co) (0.299 mGy/s)	γ ( <sup>137</sup> Cs) (0.008 mGy/s)
<b>c</b> 1	$1.03\pm0.01$	$1.01\pm0.02$	$1.00\pm0.05$	$1.11\pm0.01$	$1.10\pm0.00$	$1.10\pm0.02$
c2	$1.04\pm0.01$	$1.02\pm0.02$	$1.01\pm0.05$	$1.12\pm0.01$	$1.11\pm0.00$	$1.11\pm0.02$
c3	$1.05\pm0.01$	$1.03\pm0.02$	$1.02\pm0.05$	$1.13\pm0.01$	$1.12\pm0.00$	$1.12\pm0.02$
c4	$1.06\pm0.01$	$1.04\pm0.02$	$1.03\pm0.05$	$1.14\pm0.01$	$1.14\pm0.00$	$1.18\pm0.07$
c5	$1.07\pm0.01$	$1.05\pm0.02$	$1.04\pm0.05$	$1.15\pm0.01$	$1.15\pm0.01$	$1.19\pm0.07$
c6	$1.08\pm0.01$	$1.06\pm0.02$	$1.08\pm0.04$	$1.16\pm0.01$	$1.16\pm0.00$	$2.17\pm0.03$

**Table 4:** Activation energies (eV) of trap depth obtained by computerized deconvolution of TL glow curves registered with three radiation sources for zeroed (Z) and resensitized (ReS) samples of guartz crystal.

Comparing the trapping parameters between Z and ReS samples for the three radiation sources it is observed that the sensitization process caused some changes in the values of the kinetic parameters, which can be summarized as follows: (i) the activation energy of all components of the ReS samples are higher than the respective values calculated for the Z samples; (ii) the peak temperature and activation energy of one of the components that constitute the sensitized peak (c4) and the last component (c6) obtained with the lowest dose-rate (<sup>137</sup>Cs) source has the most different values for ReS samples, compared to the other two sources; (iii) after sensitization, the peak temperature of the 5<sup>th</sup> component has lower values; (iv) after sensitization, the peak temperature and activation energy related to the 6<sup>th</sup> component is different only to the lowest dose-rate (<sup>137</sup>Cs) source.

The most significant difference between the glow curves registered with the three radiation sources is the increase of TL signal above the 350 °C, which is observed only with the lowest dose-

rate source (<sup>137</sup>Cs) in ReS samples. This change can be better quantified using the relative values of the contribution of each component in relation to the total signal, which is summarized in Table 5.

	Zeroed			ReS		
c*	β ( <sup>90</sup> Sr/ <sup>90</sup> Y) (63 mGy/s)	γ ( <sup>60</sup> Co) (0.299 mGy/s)	γ ( <sup>137</sup> Cs) (0.008 mGy/s)	β ( <sup>90</sup> Sr/ <sup>90</sup> Y) (63 mGy/s)	γ ( <sup>60</sup> Co) (0.299 mGy/s)	γ ( <sup>137</sup> Cs) (0.008 mGy/s)
<b>c</b> 1	0.64	0.52	0.17	0.25	0.20	0.04
c2	0.10	0.09	0.02	0.02	0.02	0.00
c3	0.07	0.08	0.05	0.29	0.16	0.14
c4	0.01	0.05	0.14	0.27	0.40	0.04
c5	0.09	0.14	0.24	0.10	0.13	0.05
c6	0.09	0.12	0.37	0.08	0.10	0.72

**Table 5:** Relative values of the contribution of integral area (a.u.) of the peak components in relation to the total signal, calculated by CGCD. The area values were divided by the test-doses adopted for each case.

It is observed that for the Z and ReS samples the c1 and c2 components are higher for the highest dose-rate (<sup>90</sup>Sr/<sup>90</sup>Y), which is explained by the thermal decay of the shallow traps, which is more dominant in long irradiation times, showing a dependence on the time elapsed between excitation by ionizing radiation. This effect is considerably smaller for the ReS condition, which suggests that the trapping levels related to the first peak became more stable under sensitization. This observation is in good agreement with the increase of the activation energies shown in Table 4.

The components in the high temperature region ( $\geq 300$  °C) for Z and ReS samples, which represent the deep traps, are dependent on the dose-rate. Thus, the lower the dose-rate, the higher the area contribution per component. This effect is evident in the Z samples for the components that occur above 300 °C (c4-c6), and for the ReS samples whose component occurs above 400 °C (c6). The intensities of the components that represent the traps responsible for the sensitized peak (c3 and c4) do not show a clear relationship with the dose-rate.

The results found for the c1 and c2 components and the components above 300 °C seem to be similar to those obtained by Chen and Leung [7]. The authors compared the behavior of two TL peaks

in a dose-rate interval, noting that one emission of them, increases with increasing dose-rate, whereas another emission decreases with the dose- rate with a constant total dose.

## 4. CONCLUSION

The results of the present study can be summarized as follows:

- (i) The same set of trap depths were found in zeroed (Z) and sensitized (ReS) samples irradiated with  $\beta$  and  $\gamma$  rays. The activation energies of the trap depths were systematically higher for ReS samples. Besides the creation of the strong peak at ~260 °C, the sensitization process caused a significant reduction in the intrinsic thermal fading of the first TL peak.
- (ii) For both conditions, Z and ReS, the intensity of the components responsible for the first peak was significantly affected by the fading occurring simultaneously with excitation with 0.008 mGy/s ( $\gamma$ ; <sup>137</sup>Cs) dose-rate. The competition between trap filling and emptying was lower for ReS samples and was not perceptible when samples were irradiated with 0.299 mGy/s ( $\gamma$ ; <sup>60</sup>Co) dose-rate.
- (iii) The contribution of the component occurring above 350 °C to the whole TL signal increases with the decreasing of the dose-rate. This effect is higher for the ReS sample. For some reason that is not yet clear, the filling of the deep traps with charge carriers is favorable when test-doses are administered with dose-rates lower than 0.01 mGy/s.

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