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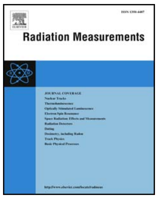

# OSL dosimetric properties of synthetic topaz

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## ABSTRACT

The optically stimulated luminescence (OSL) properties under beta radiation of synthetic topazes produced from three different reactants are reported for the first time. Topazes were synthesized by chemical vapor deposition using sodium hexafluorosilicate ($Na_2SiF_6$) compacts as solid precursor and either aluminum oxide, aluminum hydroxide or kaolinite compacts as reactants. The OSL dosimetric characteristics of the most promising composition, namely, topaz synthesized using aluminum oxide, were determined and analyzed. The alumina-derived topaz exhibited a linear response in the dose range from 0.2 up to 20 Gy, a satisfactory repeatability and a fading of approximately 30% in the first hours, after which the OSL response remained constant. These characteristics, together with a minimum detectable dose (MDD) of 0.01 Gy – corresponding to a 3σ background signal – suggest the potential of synthesized topaz as an OSL dosimeter. The OSL mechanism might be explained by at least two trap types, possibly related to topaz shallow traps. A decreasing behavior – featuring a single exponential decrease –, similar to that of OSL signal was observed in ultraviolet and visible light bleaching measurements.

## 1. Introduction

Optically stimulated luminescence (OSL) is a good alternative to thermoluminescence (TL) for applications in geological dating as well as personal dosimetry due to several advantages. One of these advantages is that the stimulation method is completely optical, which makes it unnecessary to use a heating system for stimulating irradiated samples. For the same reason no thermal quenching occurs and more robust plastics encased OSL dosimeters can be easily manufactured. Moreover, high sensitivity of OSL allows multiple readings because it is no necessary to stimulate all of trapped charges and the readout process can be made very fast by increasing the stimulating light intensity (McKeever, 2001).

At present, only a few materials to be applied in OSL dosimetry have been developed and commercialized. The first one, which could be considered as the standard material for OSL in practical dosimetry, is the C-doped alumina ($Al_2O_3$) (Perks et al., 2007) and the other one is the BeO, which has advantage of having high efficiency and a nearly tissue equivalence (Sommer et al., 2008). Nevertheless, there is a permanent interest in searching for new materials with improved OSL dosimetric properties.

On account of previous investigations where it was shown the potential application of synthetic topaz in thermoluminescence dosimetry (Trujillo-Vázquez et al., 2016), the aim of this work is to study the effect of reactant compact (either aluminum oxide ($Al_2O_3$), aluminum hydroxide ($Al(OH)_3$) or kaolinite ($Al_2Si_2O_5(OH)_4$)) and processing conditions (temperature, time, atmosphere and angle position of the compact with respect to the gas flow direction) on topaz formation and its OSL properties. Finally, the dosimetric characteristics of the most efficient composition, namely topaz synthetized using $Al_2O_3$ as reactant, have been analyzed and the feasibility of use this compound as OSL dosimeter has been evaluated.

## 2. Experimental procedure

Samples were synthesized by chemical vapor deposition (CVD) using sodium hexafluorosilicate ($Na_2SiF_6$) compacts as solid precursor and aluminum oxide ($Al_2O_3$), aluminum hydroxide ($Al(OH)_3$) and kaolinite ($Al_2Si_2O_5(OH)_4$) compacts as reactant. Samples were ground in an agate mortar and sieved to-100 mesh. Details on synthesis procedure and characteristics of the topazes synthesized using the different reactants can be found in references Trujillo-Vázquez et al. (2016) and Trujillo-Vázquez and Pech-Canul (2017).

Samples were placed at 1 cm from a 10 mCi ophthalmic Sr-90 beta-source and irradiated at room temperature. The Sr-90 beta-source rendering a dose rate of 0.022 Gy/min at the sample position. For optical stimulation a Luxeon V Star green LED with maximum emissions at 530 nm, a Luxeon V Star blue LED with maximum emissions at 470 nm

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and a Luxeon III Star red LED whit maximum emission at 627 nm, were used. LEDs were driven at 500 mA with a Laser Diode Driver model 525B of Newport rendering an effective luminous flux of 128, 38 and 56 lm at the sample position for the green, blue and red LEDs, respectively. In each case the LED light was filtered by means of two 3 mm thick Schott long-pass filters before reaching the sample, namely, OG570, OG530 and GG420 long-pass filters by red, green and blue stimulation, respectively. Each long-pass filter features a maximum transmission of about 0.9 for wavelengths higher than the cutoff wavelength (570, 530 and 420 nm, respectively) and a transmission less than $10^{-6}$ at shorter wavelengths. In order to get rid of the stimulation light, two 3 mm thick Hoya B-390 or two 3 mm thick Hoya U-340 band-pass filters were interposed between the sample and the light detector. The B-390 filter has non-zero transmission between 320 and 500 nm and maximum transmission (0.77) at 390 nm and the U-340 filter has non-zero transmission between 250 and 390 nm and maximum transmission (0.80) at 340 nm. OSL was measured by means of an Electron Tube P25PC-02 photon counting head having sensitivity between 180 and 630 nm and maximum response at 350 nm. For all measurements both irradiation and stimulation were applied to the same face of the sample from which the emitted light was detected.

UV and visible bleaching of the samples were carried out by using either a NICHIA chip type UV LED model NCSU033A (T) with maximum emissions at 365 nm, or a Luxeon V Star green LED with maximum emissions at 530 nm, respectively. In both cases, LEDs were placed at 50 cm of the sample and they were driven at 50 mA with the same Laser Diode Driver model 525B.

## 3. Result and discussion

### 3.1. Filters and LEDs

In order to determine the optimal combination of filters to maximize the collection of the light emitted by the stimulated samples, a study of the emission spectra of topazes is necessary. Since the light emitted during OSL is not stationary, obtaining its spectrum is not easy without resorting to multichannel highly sensitive detector. Instead, it is possible to obtain the radioluminescence (RL) spectrum, i.e., the spectrum of the light emitted during irradiation, which generally is the same to the OSL spectrum because it is expected that the luminescence center involved in both OSL and RL is the same.

In this context, we use the RL spectra obtained and published and a previously work (Trujillo-Vázquez and Pech-Canul, 2017). In Fig. 6 of reference Trujillo-Vázquez and Pech-Canul (2017) it is possible to see that three samples, namely $Al_2O_3$, $Al(OH)_3$ and $Al_2Si_2O_5(OH)_4$ compacts as reactant, show a broad band between 250 and 560 nm and centered at 380 nm. These spectra are similar to those measured by Yukihara et al. (1999) and Souza et al. (2006), for Brazilian natural topazes. Yukihara et al. found that the emission spectra of different topazes have a very broad and similar band ranging from 300 to 560 nm whereas Souza et al. reported in 2006 various emission spectra in the range between 350 and 550 nm for several natural topazes.

From these spectra, two configurations of filters (emission filters) were selected to interpose between the sample and the light detector, i.e., two Hoya B-390 and two Hoya U-340 band-pass filters with transmission between 320 and 500 nm and, 250 and 390 nm, respectively. Regarding the stimulation light, three light sources were selected, namely, in the red, green and blue region, respectively, as it was detailed in the Experimental Procedures Section.

When the samples were stimulated with red and green light, both configurations of emission filters, namely Hoya B-390 and Hoya U-340 band-pass filters were evaluated, whereas when they were stimulated with blue light, only the configuration with the Hoya U-340 filters was investigated because of the overlapping of the wavelength.

Of all the investigated configurations, only the last one – i.e., blue stimulation and Hoya U-340 band-pass as emission filters – showed OSL response, irrespective of the studied topaz. In this context, results shown hereafter were obtained with the last mentioned configuration.

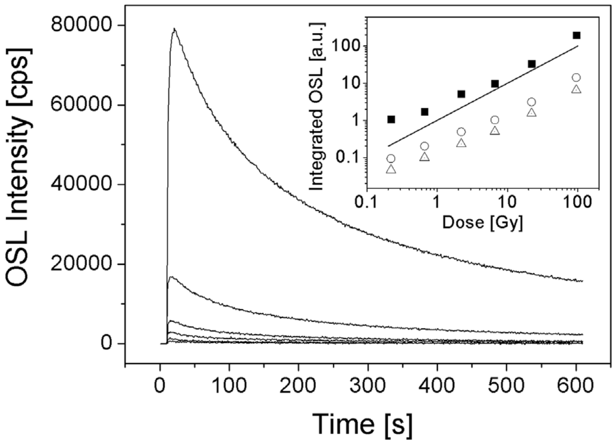

**Fig. 1.** OSL curves from topaz synthesized using $Al_2O_3$ compacts as reactant for different irradiation doses, ranging from 0.22 up to 100 Gy from bottom to top one after another. In the inset: dose response when it is integrated the first 10 (dot center triangles), 20 (hollow circles) and 600 (filled squares) seconds, respectively. Error bars are smaller than the symbol size. A solid line with the 1:1 linear behavior has been included for eye-guiding purposes.

### 3.2. OSL response as a function of reactant

Of the studied topazes, it was found that the topaz synthesized using aluminum oxide as reactant show the highest OSL response, whilst samples processed from aluminum hydroxide and kaolinite exhibit responses that are two orders of magnitude lower than that of the former. This is in accordance with a previous work (Trujillo-Vázquez et al., 2016) where the maximum TL response was obtained for samples synthesized with the same procedure. Hereafter, this work will focus on the characterization of the dosimetric properties of topaz with the highest OSL response, i.e., topaz synthesized using $Al_2O_3$ as reactant. The F/OH ratio results in synthetic topaz with the stoichiometry, $Al_2F_{1.44}(OH)_{0.56}SiO_4$.

### 3.3. Dose response

Fig. 1 shows the OSL curves from topaz synthesized using aluminum oxide compacts as reactant. As it can be seen from the figure, a good linearity is observed when the sample is irradiated with different doses of beta radiation, namely 0.22, 0.66, 2.2, 6.6, 22 and 100 Gy, from bottom to top one after another. In the inset of the figure, the dose response when it is integrated the first 10, 20 and 600 s of the OSL curve is presented. A good linearity in the dose range of 0.22–22 Gy and a supralinear behavior in the last dose is observed. In this context, when a linear regression is performed on the experimental data, a regression coefficient equal to 0.999 is found for the cases where it was integrated the first 10 and 20 s of the OSL curve, whereas if the whole OSL curve is integrated, a value of 0.996 is found. In the latter case, it is possible that the regression coefficient was lower than that of the others because while for low doses the OSL curves reached zero, for high doses this was not the case. It is possible to improve the regression coefficient when the highest dose is removed of the regression and the dose range of 0.22–22 Gy is analyzed.

### 3.4. Repeatability and fading of the OSL signal

In order to assess the feasibility of using this compound as OSL dosimeter, other OSL dosimetric properties such as repeatability and fading of the OSL signal and the minimum detectable dose were determined. Fig. 2 (a) shows the repeatability of the OSL signal when it is

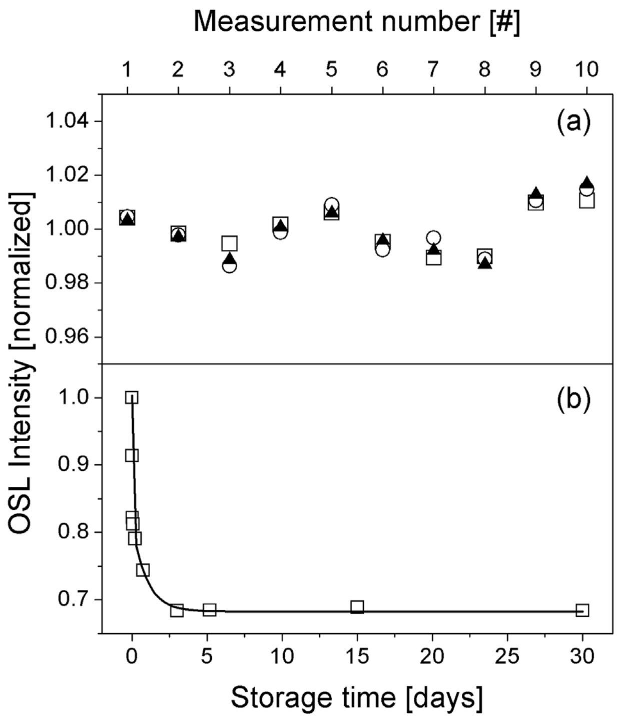

**Fig. 2.** (a) Repeatability of the OSL response (normalized by the corresponding mean value) when it is integrated the first 10 (open circles), 20 (solid triangles) and 600 (open squares) seconds, respectively. In (b) Integrated OSL signals as a function of the storage time. Continuous line has been obtained by fitting with a double exponential decay curve.

integrated the first 10, 20 and 600 s of the OSL curve, finding a standard deviation of 1.5, 1.7 and 1.1%, respectively. In all cases, samples were irradiated with a dose of 2.2 Gy and read 2 h after irradiation in order to eliminate the fast fading component completely. The lifetime of this fast fading component was found to be of 0.23 h (see Fig. 2 (b)). Results have been normalized by the corresponding mean value.

Fig. 2 (b) shows results of the fading of the OSL signal for different storing times in darkness and at room temperature. Sample was irradiated with a beta dose of 2.2 Gy. As it can be seen in the figure, the OSL signal decreases down to 70%, after which the signal remains constant. It is apparent from the figure that the OSL decay curves are not simple exponential functions. In order to characterize the OSL decay, a fitting with a double exponential function was carried out. The best-fitting for the lifetimes resulted in a fast component characterized by a lifetime of approximately 0.23 h and a slow component having a lifetime of 23 h, respectively. This fact evidences that at least two types of traps are involved in the OSL mechanism. The same behavior was observed in a previous work (Trujillo-Vázquez et al., 2016) for the study of the thermoluminescence fading of this compound and this partial emptying of OSL traps could be related to the shallow traps present in the topaz.

### *3.5. Minimum detectable dose (MDD)*

According to the definition (Yukihara and McKeever, 2011), the minimum detectable dose (MDD) is defined as MDD = 3 $\sigma_{BG}$, being $\sigma_{BG}$ the experimental standard deviation of the background signal recorded by using the same detectors and a non-irradiated sample. Accordingly, a MDD = 0.01 Gy was found for topaz synthesized using $Al_2O_3$ as reactant.

### *3.6. UV and visible bleaching*

Finally, a study of the influence of ultraviolet (UV – 365 nm) and visible (green – 530 nm) light on OSL curves of synthetic topaz was analyzed. In both cases, samples were irradiated with a dose of 2.2 Gy.

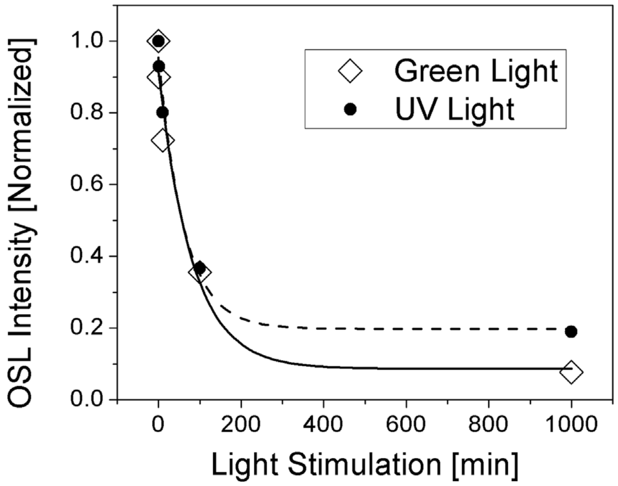

**Fig. 3.** Integrated OSL signals as a function of green (open diamonds) and UV (solid circles) light stimulation. Solid and dashed lines have been obtained by fitting with a single exponential decay curve for each case.

Firstly, irradiated samples were exposed to green light for different periods of time. Fig. 3 shows the OSL intensity, defined as the area under the OSL curve during the first 20 s, as a function of green exposure time (open diamonds). The integrated OSL signal decreases as a single exponential function with a lifetime of approximately 62 min.

Secondly, the influence of the UV light on the OSL curves was also analyzed (see Fig. 3, solid circles) and a similar behavior with the green bleaching was observed, namely, the OSL intensity decreases as a single exponential function with a lifetime of 80 min. In the case of UV bleaching, the OSL intensity decreases down to 20% of the curve measured immediately after irradiation whereas that for green stimulation, the OSL intensity decreases down to 10% of the main signal.

If it is compared the effect of UV and green light bleaching on OSL intensity with the fading study, i.e., OSL response of the samples storage in darkness and at room temperature, it is possible to see some differences.

First, the fading study was characterized by a double exponential function with two lifetimes of approximately 0.23 and 23 h, respectively and, the OSL signal decreased down to 70% of the OSL signal recorded immediately after irradiation. Then, the OSL intensity remains constant. This fact also was observed in a previous work (Trujillo-Vázquez et al., 2016) for the study of the thermoluminescence (TL) fading of this compound where the TL signal decreases down to 75% of the TL glow curve measured immediately after irradiation. As mentioned above, this partial emptying of OSL traps could be related to the shallow traps present in the synthesized topaz.

On the other hand, both green and UV bleaching have been characterized by a single exponential function with lifetimes of 62 and 80 min and the OSL signals decreases down to 10 and 20%, respectively. This could be explained by assuming that both visible and UV light have enough energy for emptying both shallow and deep traps of the samples at the same time. These behaviors make that close attention must be paid to the light that can reach the sample between irradiation and reading.

As it was reported by other authors (Souza et al., 2002; Dantas et al., 2006), the emission bands of both topaz and quartz are similar and two color centers are mentioned, namely, $(AlO_4)^0$ and $(H_3O_4)^0$ emitting at 460 and 380 nm, respectively. In Fig. 6 of a previous work (Trujillo-Vázquez and Pech-Canul, 2017), it is possible to see that samples show a broad band between 250 and 560 nm and centered at 380 nm, which could be attributed to $(H_3O_4)^0$ centers, although it is possible to observe a shoulder between 400 and 500 °C which could be ascribed to $(AlO_4)^0$. Although the same color centers and consequently the same emission

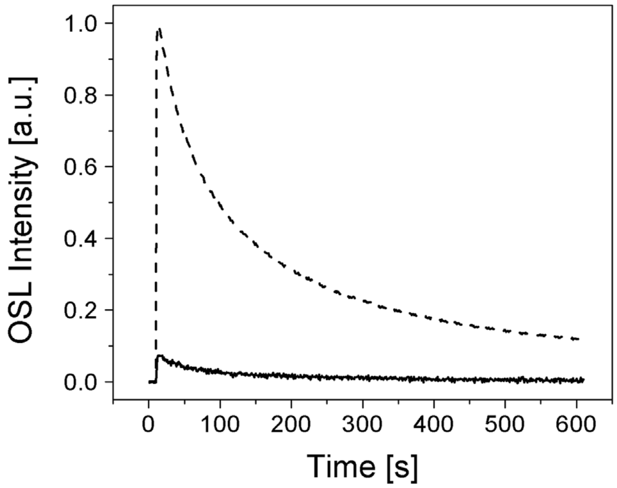

**Fig. 4.** Comparison between the OSL curves of commercial $Al_2O_3$:C (dashed line) and topaz synthesized using $Al_2O_3$ compacts as reactant (solid line). Both OSL signals have been recorded under the same experimental conditions and normalized to the sample weight.

wavelength were observed in both, RL and TL of topazes (Trujillo-Vázquez et al., 2016; Souza et al., 2004; Ribbe and Gibbs, 1971), the emission efficiency depends strongly on the reactant used in the synthesis of the topaz (Trujillo-Vázquez et al., 2016; Trujillo-Vázquez and Pech-Canul, 2017).

In this work, it was found that topaz synthesized using aluminum oxide as reactant shows the highest OSL response, whilst samples processed from aluminum hydroxide and kaolinite exhibit responses that are two orders of magnitude lower than that of the former. This is in accordance with a previous work (Trujillo-Vázquez et al., 2016), where the F/OH ratio resulted in synthetic topaz with the stoichiometry $Al_2F_{1.44}(OH)_{0.56}SiO_4$ that showed the maximum TL response. On the other hand, in reference Trujillo-Vázquez and Pech-Canul (2017) it was found that topaz formed using aluminum hydroxide showed the maximum RL response whilst samples processed from aluminum oxide and kaolinite exhibited responses two and five times lower than those of the first one. These results could be evidence that during the synthesis of topaz using aluminum oxide as reactant, a greater number of trap centers are created whereas in that using aluminum hydroxide as reactant, a greater number of recombination centers are formed.

### 3.7. *Comparison with $Al_2O_3$:C*

In Fig. 4 it is possible to see the comparison between the OSL curve of the standard commercial $Al_2O_3$:C dosimeter (Landauer, Inc.) and topaz synthesized using $Al_2O_3$ compacts as reactant. Samples were irradiated with a beta dose of 2.2 Gy and OSL curves were normalized to the sample weight.

It is possible to see from the figure that $Al_2O_3$:C is twenty times greater than synthetic topaz. This fact in principle could be considered as a disadvantage. On the other hand, the OSL signal of synthetic topaz decays faster than that of $Al_2O_3$:C and fades out completely after 200 s of stimulation. This result implies that the OSL readout empties most of the trapped charges in this material so that no any additional optical bleaching process is necessary before reutilization. In the case of $Al_2O_3$:C, the long decay time of its OSL signal could become a drawback if the total depletion of traps is necessary before each OSL measurement (Gaza et al., 2005). A solution consists in only deplete partially the accumulated OSL signal during each stimulation period and to devise correction algorithms to account for the degree of depletion from all previous optical stimulations (Gaza et al., 2005).

## 4. Conclusions

The OSL dosimetric properties of several synthetic topazes have been investigated for the first time. Of the studied samples, topaz synthesized using $Al_2O_3$ as reactant showed the highest OSL response whilst samples processed from aluminum hydroxide and kaolinite exhibit response two order of magnitude lower than the first one.

Regardless the studied topaz, only the configuration with blue stimulation and two Hoya U-340 band-pass as emission filters, showed an OSL response. With this configuration, it was found that topaz synthetized using $Al_2O_3$ as reactant presented a linear response in the studied dose range from 0.2 up to 22 Gy and a minimum detectable dose of 0.01 Gy. Besides, the repeatability study of the OSL signal showed a standard deviation of 1.5, 1.7 and 1.1% when it is integrated the first 10, 20 and 600 s of the OSL curve, respectively, A fading study showed that the OSL signal decreases down to 70% and then the signal remains constant. The OSL decay curves are characterized by a double exponential function with two lifetimes of approximately 0.23 and 23 h, respectively. This fact evidences that at least two types of traps are involved in the OSL mechanism which could be related to the shallow traps present in the topaz. Ultraviolet and visible light bleaching measurements were carried out and a similar decreasing behavior of OSL signal was observed. In both cases, a single exponential decrease was observed, with a lifetime of approximately 62 and 80 min, respectively.

Although the OSL efficiency of synthetic topaz was twenty times lower than that of the $Al_2O_3$:C, the quickly fades out of OSL signal could be an advantage in order to restore the sample between dose measurements.

The results of this work show that synthetic topaz could be considered as a promising OSL dosimeter, worthy of further research.

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