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# TL and OSL dosimetric properties of Opal gemstone for gamma radiation dosimetry

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## HIGHLIGHTS

- The XRD, SEM and EDX techniques were used to investigate powdered Opal.
- Pellets of three different concentrations of Opal and Teflon were studied.
- The dosimetric properties of the Opal + Teflon pellets were verified.
- TL and OSL techniques were used to analyze the characteristics of the pellets.
- Pellets of concentration of 2:1 (Opal:Teflon) presented the most adequate results.

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## ABSTRACT

In this work, the response of the natural material Opal was studied in relation to its thermoluminescence (TL) and optically stimulated luminescence (OSL), after exposure to the gamma radiation of a $^{60}$Co source. The structure of the powdered Opal was verified using the X-ray diffraction, scanning electronic microscopy and energy-dispersive X-ray spectroscopy techniques. The material, in its stone form, was turned into powder and mixed to Teflon (also in powder) in three different concentrations, and then pellets were manufactured. The aim of this work was to evaluate the response of these pellets in high-doses of gamma radiation beams, and to observe their possible application as dosimeters, using the TL and OSL techniques. The dosimetric properties of the samples were analyzed by means of different tests, as: TL emission curves and OSL signal decay curves, reproducibility of TL and OSL response, minimum detectable dose, TL and OSL dose–response curves (5 Gy–10 kGy), and fading. The results obtained in this work, for the TL and OSL phenomena, demonstrated that the pellets of Opal + Teflon present an adequate performance e possibility of use as dosimeters in beams of high-dose gamma radiation.



## 1. Introduction

Opal samples were frequently obtained from the mines of Czechoslovakia until the 19th century, but nowadays they mostly come from Australia; for this reason, Opal is considered as the Australian national gemstone. This material is basically composed of silica and water, and can mainly be of four types: white, black, fire and water Opal. Its main characteristics are: the opalescence, in which it is possible to observe a blue aspect in the stone, as a consequence of the reflection phenomena, the opalization, a color set of the Opal that varies according to the angle in which the gemstone is observed, and also the iridescence, a phenomenon caused by the decomposition of light into fractures and flaws that result in rainbow colors. In addition to the four main types of Opal, they are also classified as common or precious stones, and only the precious

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ones present the iridescence phenomenon (Hall, 1997; Schumann, 2002).

The Opal stone was already studied in the natural and synthetic forms, in relation to the luminescent response emission after its stimulation by heating, that consists in the thermoluminescence (TL) technique, so much used in several research areas involving solid-state dosimetry. TL glow curves were obtained presenting peaks around 230°, 300° and 360 °C; the first one was related to the water in Opal and the third one is characteristic of some Australian sedimentary fields. This study also demonstrated that no effect after gamma irradiation was verified in these samples (Meakins et al., 1978). Another study performed was a comparison between the microcrystalline and non-crystalline Opal of the structural point of view, using different techniques, as magic-angle spinning nuclear magnetic resonance (MAS NMR), X-ray diffraction (XRD) and infrared absorption spectroscopy (IR) (Graetsch et al., 1994). Opal was also investigated in nanocrystalline form, and it was synthesized, morphologically and structurally characterized, and its response was compared with that from natural Mexican Opal (Hernández-Ortiz et al., 2015).

The purpose of this work was to evaluate the response of the Opal + Teflon pellets in high-dose gamma radiation beams. Another objective was to verify the possibility of its application in high-dose gamma dosimetry, using the TL technique and also that in which the luminescent signal is emitted after optical stimulation, the process of optically stimulated luminescence (OSL). This technique has been applied in several studies (Yukihara et al., 2014).

## 2. Materials and methods

Teflon powder was added to the powdered Opal stone in proportions of 1:1, 1:2 e 2:1 (Opal:Teflon), and round sintered pellets were manufactured with dimensions of 6.0 mm in diameter and 0.8 mm in thickness. The pellets were then thermally treated at 350 °C for 30 min and 400 °C for 3 h, to ensure good uniformity and mechanical strength in the material.

The Opal + Teflon pellets were irradiated at the Radiation Technology Center, IPEN, using a Gamma-Cell 220 System, model 200, Atomic Energy of Canada LTD, with a $^{60}$Co source (47.64 TBq, Oct./2014).

The response of the pellets was evaluated in terms of their TL and OSL signals, using the reader system composed by the TL/OSL meter Risø, model TL/OSL-DA-20. The parameters used in the TL measurements were: heating rate of 10 °C/s and a final temperature of 300 °C. In the case of the OSL readings, the conditions were: optical power of 90% for blue LEDs, stimulation time of 50 s, and the use of a filter basket Hoya U-340 in front of the photomultiplier.

Initially, the OSL reading was taken, and afterwards the TL measurement of each sample. After these two readings, the pellets were submitted to the thermal treatment of 300 °C during 1 h, to erase any residual signal of the samples and to allow their reutilization.

X-ray Diffraction (XRD) measurements were taken using a diffractometer Equinox 1000 INEL, with CuKa radiation ($\lambda$ = 1.5418° A), with the tube operating at 300 kV/30 mA in the continuous mode with steps of 2°min$^{-1}$. The identification of the crystalline phases was performed using the software X-Pert High-Score (Panalytical) and the crystallography data for all phases were obtained using the Inorganic Crystal Structure Database (ICSD).

Samples of Opal powder used in the preparation of the pellets were analyzed by dispersive X-ray spectroscopy (EDX) and scanning electron microscope (SEM) with a scanning electron microscope, model Vega 3 SEM Tescon, operating with a tungsten filament.

## 3. Results

Inititally, the physical and chemical characteristics of the powdered Opal were verified by the XRD, SEM and EDX techniques. After the production of the Opal + Teflon pellets in all three concentrations, their dosimetric properties were studied using the TL and OSL responses through the characterization tests, as: TL glow curve and OSL signal decay, reproducibility of response, dose–response curves, lower detection limits and fading of the signals.

### 3.1. X-ray diffraction (XRD)

The XRD results obtained can be observed in Fig. 1, which shows the XRD standard of the powdered Opal combined with the reference standard ICSD 200728, corresponding to the silicon oxide ($SiO_2$). The $SiO_2$ signal belongs to the hexagonal crystal system and to the spatial group P3121. The crystallographic parameters were: a = 4.8170 Å, b = 4.8170 Å, c = 5.3280 Å, $\alpha$ = 90°, $\beta$ = 90° and $\gamma$ = 120°.

### 3.2. Scanning electron microscope (SEM)

The analysis performed using the scanning electron microscope (SEM) identified the presence of microcrystals. It was also possible to observe microspheres of silica, an Opal characteristic, in an ordered structure forming a kind of layers, according to Fig. 2. Thus, it can be observed that the Opal sample presents irregular morphology and defined edges.

### 3.3. Energy dispersive X-ray spectroscopy (EDX)

The experiment performed through analysis of energy dispersive X-ray spectroscopy showed a predominant presence of $SiO_2$ (94.6%) and a variety of impurities in small quantities, as Na (1.6%), Fe (1.4%), Mn (1.1%), Cu (1.0%) and Ca (0.4%). This result, with the analysis by XRD, proves the composition of $SiO_2$ present in Opal. This description can be better visualized by Fig. 3.

### 3.4. TL glow curve and OSL signal decay

The TL glow curve and the OSL signal decay (Fig. 4) of the Opal + Teflon pellets were obtained after exposure of the samples to the $^{60}$Co source, with an absorbed dose of 500 Gy. Fig. 4a shows the TL glow curves for all three concentrations of material (1:1, 1:2 and 2:1 -Opal:Teflon): a dosimetric peak can be observed at about

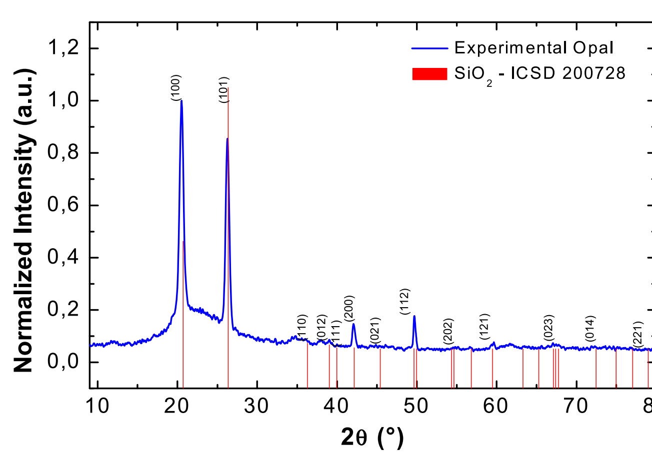


**Fig. 1.** X-ray diffraction pattern of the powdered Opal, compared to the reference standard ICSD 200728 of $SiO_2$.

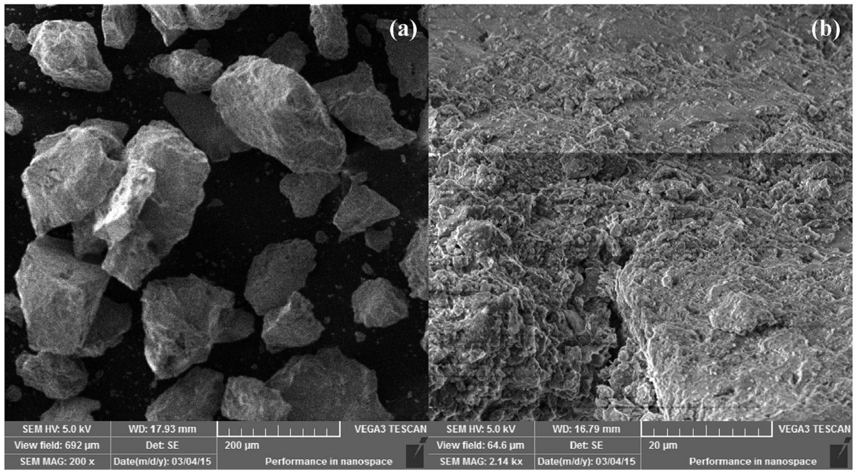


**Fig. 2.** Micrographs of powdered Opal: (a) increase – 200 x; and (b) increase – 2.14 kx.

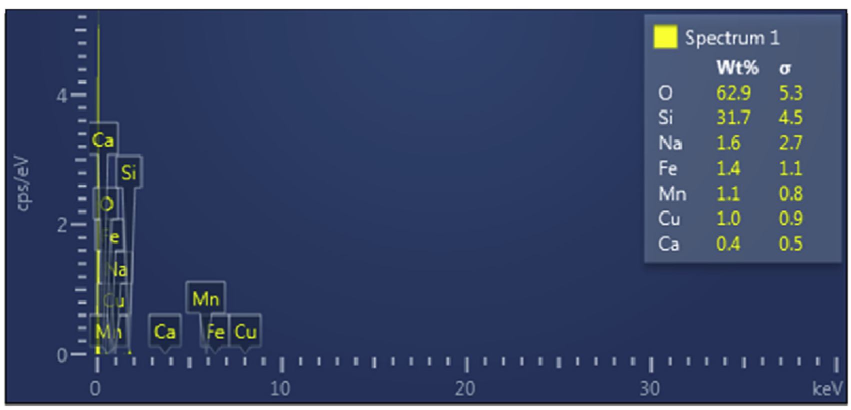


**Fig. 3.** Energy dispersive X-ray spectroscopy of powdered Opal.

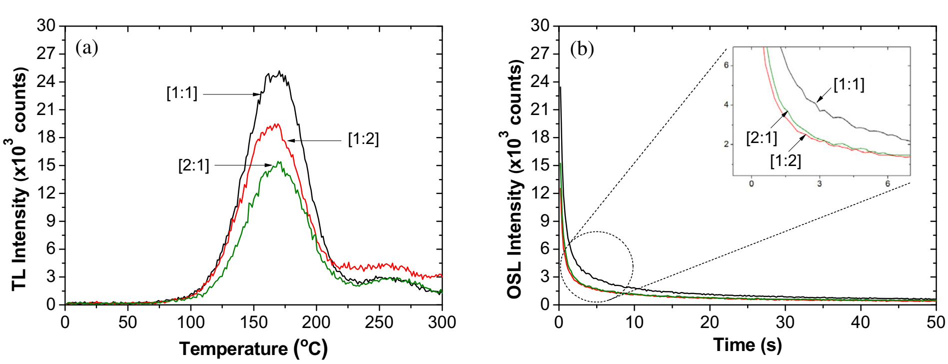


**Fig. 4.** TL and OSL response of the Opal + Teflon pellets: (a) TL glow curve; and (b) OSL signal decay, obtained after an irradiation to 500 Gy ($^{60}$Co).

170 °C. Fig. 4b presents the OSL signal decay for all three types of pellets.

### 3.5. Reproducibility of TL and OSL responses

The reproducibility study of the TL and OSL responses of the Opal + Teflon pellets was performed using 15 samples of each concentration: 1:1, 1:2 and 2:1 (Opal:Teflon). The measurements were taken in 5 cycles of irradiation, signal evaluation and thermal treatment. For this characterization test, the pellets were irradiated with an absorbed dose of 1 kGy. Table 1 presents the results

**Table 1**
Reproducibility study performed with Opal + Teflon pellets.

| Luminescent technique | Reproducibility (%) | | |
|---|---|---|---|
| | [1:1] | [1:2] | [2:1] |
| TL | 1.4 | 1.7 | 1.2 |
| OSL | 4.5 | 3.4 | 1.8 |

obtained for both luminescent techniques.

From the results showed in Table 1, it was possible to conclude that the pellets of proportion 2:1 (Opal:Teflon) present the best reproducibility in their TL and OSL responses, although the pellets of the other two concentrations can also be considered reproducible (lower than 4.5%).

The reproducibility study performed with the Opal + Teflon pellets provided comparable results to those already reported by Antonio and Caldas (2015) for Spectrolite + Teflon samples, irradiated with an absorbed dose of 1 kGy of $^{60}Co$, with reproducibility results of 4.2% and 4.9% for concentrations of 1:1 and 1:2 of Spectrolite:Teflon, using the TL technique, and 2.5% and 2.4% (1:1 and 1:2) using the OSL technique. D'Amorim et al. (2014) obtained a reproducibility of OSL response of 2.1%, after an irradiation of α-Spodumene pellets with 30 Gy ($^{60}Co$ source).

### 3.6. Lower detection limit (LDL)

The lower detection limit for each one of the three concentrations of Opal + Teflon samples, and for both luminescent techniques (TL and OSL), was determined by the variation of the responses of the non-irradiated pellets, taken after the thermal treatment. This limit was determined using the method of taking three times the standard deviation of non-irradiation detectors, according to Pagonis et al. (2006). The results obtained in this study can be observed in Table 2.

In relation to other studies with silicates and irradiation with the same source ($^{60}Co$), Vila (2012) obtained lower detection limits of 2.0 Gy for Tremolite, with the TL technique, and 52.9 Gy for Rhodonite, with the OSL technique. D'Amorim et al. (2014) reported a LDL of 80 Gy for α-Spodumene using the OSL technique. Therefore, the results obtained in this work agree with some already presented in literature for silicates.

### 3.7. Dose-response curves

The TL and OSL responses were obtained varying the absorbed dose from 5 Gy to 10 kGy using the $^{60}Co$ source.

For the TL technique, the maximum standard deviations obtained in the readings were: 9.3% (concentration of 1:1 of Opal:Teflon, and absorbed dose of 50 Gy), 9.8% (1:2 and 100 Gy) and 8.0% (2:1 and 50 Gy). In the case of the OSL technique, the values of maximum standard deviations were: 28% (1:1 and 50 Gy), 18% (1:2 and 100 Gy) and 9.4% (2:1 and 50 Gy).

For both dose–response curves (TL and OSL), data for 5 Gy, 10 Gy and 50 Gy were not considered with confidence because they were around the minimum detectable doses. Thus, if the interval of absorbed dose from 50 Gy to 10 kGy is taken into account, it is possible to observe a supralinear behavior from 50 Gy to 5 kGy with a tendency to saturation from 5 kGy to 10 kGy, in the case of TL response. For the OSL technique, the response presented a sublinear behavior from 50 Gy to 10 kGy. These descriptions are according to the information showed in Fig. 5a–b, for the dose-TL and OSL response curves, respectively.

The literature already showed results obtained for other materials of the silicate family. D'Amorim et al. (2014) verified a linear behavior from 100 Gy to 10 kGy for the α-Spodumene with the OSL technique. Vila (2012) obtained a linear behavior between 50 Gy and 1 kGy for Tremolite silicate and the TL technique, and a sublinear behavior in a range of 1 Gy–20 kGy for Rhodonite silicate and the OSL technique. Antonio and Caldas (2015) studied pellets of Spectrolite + Teflon; for both TL and OSL responses; an initial linear behavior with tendency to saturation was observed in an interval of 5 Gy–10 kGy.

**Table 2**
Lower detection limits obtained for Opal + Teflon pellets.

| Luminescent technique | Lower detection limit (Gy) | | |
|---|---|---|---|
| | [1:1] | [1:2] | [2:1] |
| TL | 8.75 | 12.9 | 19.5 |
| OSL | 32.6 | 44.6 | 46.2 |

### 3.8. Fading

The study of the TL and OSL signal fading was performed after irradiating the Opal + Teflon pellets, in three concentrations, with an absorbed dose of 1 kGy ($^{60}Co$ source). The measurements were immediately taken after the irradiation and after 24 h, 48 h, 96 h and 168 h. The behavior of the TL and OSL fading can be observed in Fig. 6a–b.

In the case of the TL technique, the maximum standard deviations verified in the measurements were: 34.9% for concentration of 1:1 of Opal and Teflon (168 h after irradiation), 17.4% for 1:2 (0 h), and 7.7% for 2:1 (0 h). For the OSL response, these results were 26.9% for 1:1 (24 h), 16.9% for 1:2 (0 h), and 8.5% for 2:1 (96 h).

Table 3 presents the results of the fading study of the TL and OSL techniques in terms of remaining signal (in %), obtained for the Opal + Teflon pellets.

The results presented in Table 3 show the values determined in each time interval normalized to zero time (initial reading). In relation to the TL response, the pellets of concentration of 1:1 (Opal:Teflon) showed the lowest fading (168 h), because the signal remained as 24.5% from the initial value (0 h). For the OSL response, the samples with concentration of 1:1 presented a considerable oscillation, with measurement values increasing some times. Taking into account the other two concentrations, the pellets of 2:1 showed a higher remaining signal in relation to the other materials, showing that the fading was lower for this material.

Vila (2012) obtained a remaining signal of the TL response of 91% for Tremolite, and of 69% for Rhodonite, for OSL response, and after 168 h post-irradiation. Antonio and Caldas (2015) observed remaining signals of 37.9% and 44.4% for TL response, and 39.1% and 49.1% for OSL response for the Spectrolite + Teflon pellets; all these values were obtained after 120 h of the irradiation. Comparing the results obtained in this work, for the Opal + Teflon pellets and those already showed in literature, it can be considered that the fading for the TL response is considerable high, while for OSL response, the fading is low.

## 4. Conclusions

Analyses of the powdered Opal were performed by means of XRD, SEM and EDX techniques. The first technique allowed to compare the standard of the powdered Opal experimentally obtained with a reference standard ICSD, corresponding to the silicon oxide ($SiO_2$); hexagonal crystal system. The SEM technique revealed the presence of microspheres, and irregular morphology and defined edges in the Opal grains. The predominant presence of $SiO_2$ (94.6%) was verified by the EDX technique.

The dosimetric properties of the pellets were also studied exposing the material to a $^{60}Co$ source. The dosimetric characteristics were obtained with 3 different concentrations of Opal and Teflon (1:1, 1:2 and 2:1), by means of the following characterization tests of the TL and OSL responses: TL glow curve, OSL decay curve, reproducibility of the response, dose–response curve and lower detection limits.

The results obtained in the reproducibility study of TL and OSL responses, in the dose–response curves and in the fading study of TL and OSL signal, showed that the most adequate results were

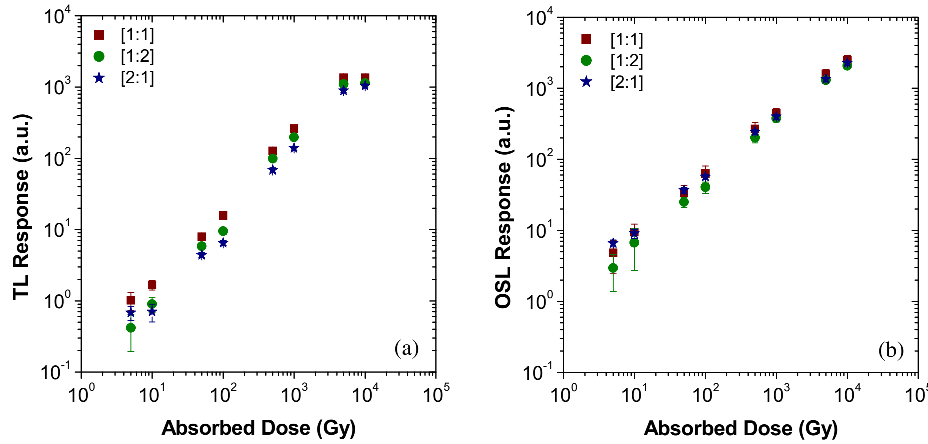


**Fig. 5.** Dose-response curves for: (a) TL; and (b) OSL techniques, after irradiation of Opal + Teflon pellets with a $^{60}$Co source.

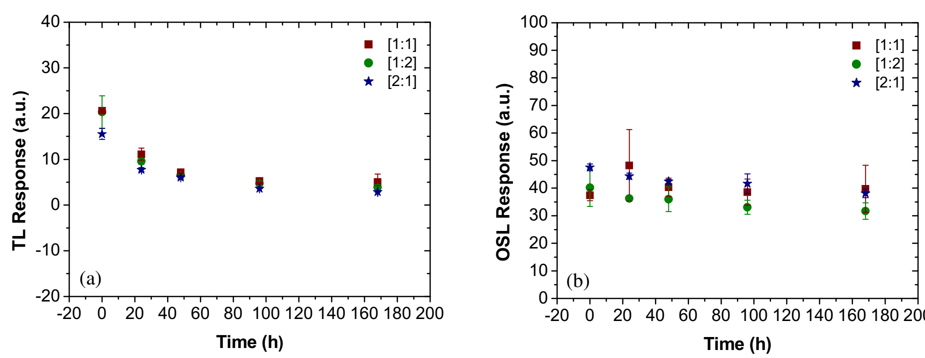


**Fig. 6.** Fading study of the Opal + Teflon pellets after irradiation with a $^{60}$Co source: (a) TL; and (b) OSL techniques.

**Table 3**
Fading study represented by the remaining signal (in %) of the TL and OSL responses of the Opal + Teflon pellets, for three different material concentrations.

| Time (h) | Fading (%) | | | | | |
|---|---|---|---|---|---|---|
| | TL response | | | OSL response | | |
| | [1:1] | [1:2] | [2:1] | [1:1] | [1:2] | [2:1] |
| 0 | 100 | 100 | 100 | 100 | 100 | 100 |
| 24 | 53.8 | 47.0 | 49.8 | 128 | 90.2 | 93.3 |
| 48 | 34.7 | 32.0 | 38.8 | 108 | 89.6 | 89.3 |
| 96 | 25.4 | 23.1 | 23.1 | 103 | 82.3 | 87.5 |
| 168 | 24.5 | 19.1 | 18.3 | 106 | 78.9 | 80.1 |

obtained for the concentration of Opal:Teflon (2:1); therefore, they present application as gamma radiation detectors in high-dose dosimetry.

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