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Evaluation of doses in radiotherapy using solid-state composites based on natural colourless topaz

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Abstract

The thermoluminescent properties of composites containing powdered topaz embedded in Teflon or glass were studied and compared with the corresponding properties of the well known TLD-100 commercial dosimeters. Relative sensitivity, TL fading, reproducibility of the sample preparation process, and possibility of re-utilisation of the dosimeters were investigated. Measurements of absorbed doses in simulated radiotherapy treatments were also taken. The irradiations were performed using γ rays from a ^{60}Co source in the dose range from 1 cGy to 2 Gy. The dosimeters were installed in badges and attached inside acrylic plates of various thicknesses in the radiation fields. The dose profiles obtained with this procedure are very similar to the ones obtained with equivalent tissues. It is concluded that these composites of natural colourless topaz crystals can be efficiently used as TL dosimeters.

Keywords: Topaz; Thermoluminescence; Dosimetry

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1. Introduction

Thermoluminescence (TL) has proved to be useful in measuring doses to a particular material. Part of the radiation energy gets absorbed inside the materials forming charge carriers trapped in localised energy levels in the band gap. During the heating process, the detrapping and recombination of the charge carriers give rise to the typical TL emission of the material. The intensity of light is proportional to the amount of radiation, up to saturation. The colour of the emitted light and the temperature at which it occurs are related to the properties of the defects responsible for the traps and the recombination centres.

TL of many different materials, including artificially grown and natural crystals, has been studied with the aim of developing new solid-state TL dosimeters (TLD). An advantage of artificially grown crystals is the possibility to control the composition of the starting materials. On the other hand, natural crystals can be cheaper and available in reasonably large quantities.

According to Keifer et al. (1969), the properties that are desirable for a good solid-state TL dosimeter are: (i) a good TL sensitivity in the dose range from 10^{-4} to 100 Gy (doses typically involved in personal dosimetry and radiotherapy), (ii) a possibility to manufacture a small and easy-to-handle dosimeter, (iii) mechanical resistance under conditions of normal manipulation (temperature, ambient light and humidity), (iv) low cost, (v) known dependences of the response on the type and energy of the radiation, (vi) independence of the TL response of the dosimeter orientation, (viii) independence

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of the response of the time elapsed between the irradiation and reading. To our knowledge, all of these features cannot be found yet in any single solid-state TL dosimeter, and, in many practical cases, a combination of different dosimetric techniques is the only way of getting the desirable information.

Topaz is aluminium fluorosilicate of the composition $Al_2(SiO_4)(F,OH)_2$. It crystallises in a rhombohedrical structure of the space group Pbnm. Topaz is a very hard mineral, typically yellow, blue or colourless. Gandini et al. (1993) reported the average crystal lattice parameters for topaz found in Minas Gerais, Brazil as $a = 4.61$ Å, $b = 8.78$ Å, and $c = 8.41$ Å. The main crystal defects in topaz are the OH^- group substituting F^- ions. The main impurities in the coloured Brazilian topaz, namely, V, Cr, Fe, and Mn, are believed to be the source of its colour.

The first report of TL in topaz was provided by Moss and McKlveen (1978). The saturation dose for their natural samples from the Topaz Mountain in Utah, USA was found to be 700 Gy, and the TL signal intensity increased by 20% after 40 cycles. Azorin et al. (1982) studied TL properties of various minerals from Mexico and found topaz to be the most sensitive. They reported two main TL peaks on a typical topaz TL glow curve. Lima et al. (1986), on the other hand, found four TL peaks in their studies of natural topaz from Governador Valadares, Minas Gerais, Brazil. They also reported that the number and positions of the TL peaks depend on the time between the irradiation and the TL measurement.

In a previous work (Souza et al., 1995, 1997), we studied the TL emission of natural topaz samples of various colours from various areas of Brazil. We found that: (i) there are up to six TL peaks (at 80°C, 140°C, 170°C, 230°C, 280°C, and 330°C) in some samples, (ii) colourless samples are more sensitive to gamma radiation than coloured ones, and (iii) UV light can promote filling of some TL traps. We determined the kinetic parameters of the peaks and found that some of them fit the first order model. Also, we found that the 170°C TL peak of the natural samples displays an anomalous fading.

TL dosimeters have been used in radiotherapy to measure the entrance and exit doses in various tissues, including the skin and the lymphatic system. TL dosimeters are small and can be easily adapted to in vivo measurements without a significant change in the actual radiotherapy treatment (Zehetmayer et al., 1997; Duggan and Johnson, 2000). Kron et al. (1996) found it possible to evaluate the dose in a thin layer of about 1.0 mm of the epithelial tissue using TL dosimeters of different thicknesses. This information is crucial for predicting initial and long-term reactions in the skin in some radiotherapeutic treatments and also for evaluating the doses in the lymphatic tissue in order to avoid future recursive diseases.

TL dosimeters are also useful for calibration of other dosimetric devices. Hayne et al. (2001) have shown that a Scanditronix rectal probe containing five n-type photon-detecting diodes, which is used to measure radiation doses received by anorectum during pelvic radiotherapy, could be accurately calibrated using a set of TL dosimeters. The corrected doses could than be compared with the doses predicted by a GE TARGET(TM) treatment planning system.

New solid-state TL dosimeters were developed for precise determination of dose profiles in the irradiation fields used in radiotherapeutic treatments (Furetta et al., 2001). Beitler et al. (2001) investigated the depth dose profile from the rear surface wall of the phantom's tracheostomy stoma. They used a water-equivalent phantom; the doses were obtained by film dosimetry and thermoluminescent dosimeters oriented parallel to that surface.

The aim of the present work was to investigate the potential of using composites based on natural colourless topaz as TL dosimeters in radiotherapy. Dose response and the ability of the new dosimeters to reproduce the equivalent tissue dose profiles will also be discussed.

2. Experimental

We studied natural colourless topaz samples from Brazil. The composite samples were prepared in two ways: by mixing powdered topaz with Teflon and by embedding topaz microcrystals into a glass matrix. Preparation of the topaz–Teflon composites was described in a previous paper (Souza et al., 2000). The topaz–glass composites were prepared using a 1:1 (wt) mixture of the powdered topaz crystal and powdered commercial glass, the grain size was in the 0.045–0.075 mm range for both materials. Pellets (1 mm thick, 6 mm in diameter) were made by uniaxial pressing followed by sintering at 750°C for 3 h. The dosimeters were irradiated with gamma rays on a ^{60}Co radiotherapy unit at the distance of 80 cm from the source. TL measurements of the topaz–Teflon composites were taken with Harshaw 3500 equipment, which operates in the temperature range from 50°C to 330°C; the measurements for the topaz–glass composites were performed at temperatures up to 400°C; in both cases, a linear heating program was used with a rate of 2°C/s. In most cases, at least 10 TL readings were taken under the same conditions; the results reported below are mean values with error bars representing standard deviations.

3. Results and discussions

In Fig. 1, the TL emission curves of the topaz–Teflon and topaz–glass composites are compared with the

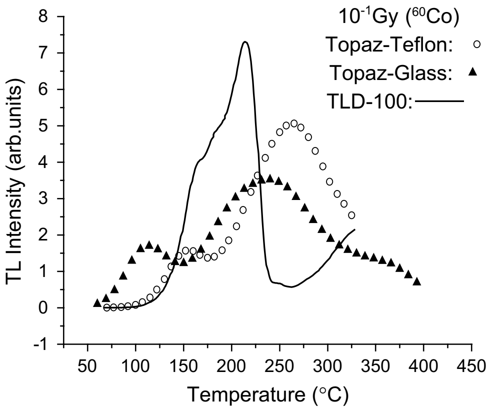


Fig. 1. TL glow curves for the topaz–Teflon and topaz–glass composites compared with the TL curves for the commercial TLD-100 (LiF:Mg,Ti) dosimeters. The samples were irradiated to 0.1 Gy with gamma rays (^{60}Co) at room temperature. The temperature increase rate in the TL experiments was 2°C/s.

emission curves of the TLD-100 dosimeters (Harshaw, LiF:Mg,Ti). The main difference between the TL emissions of the two composites and of the TLD-100 samples irradiated with γ-rays (^{60}Co) is in the peak positions. The topaz composites feature TL peaks at higher temperatures than the TLD-100 dosimeters, with almost 50% of the overall TL emission area located above 220°C. These results indicate that the TL of the composites may be more stable at room temperature. This result is important for certain applications, such as environmental dosimetry, where the TL of the samples is normally measured long after the sample were irradiated.

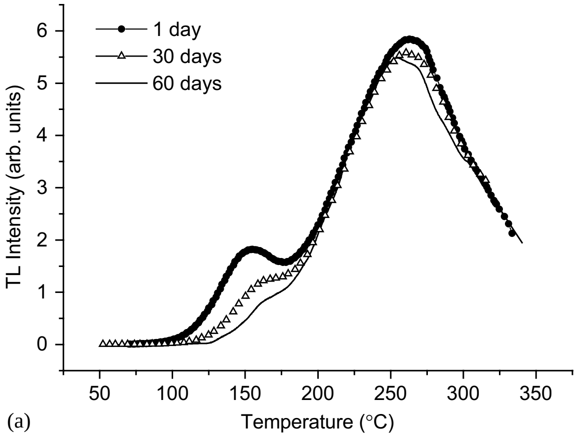
In comparing the TL emission intensities for the composites and the TLD-100 pellets, one should take into account the differences in the masses of the dosimeters. The mass-normalised TL sensitivity of the topaz–Teflon composite is 2.2 times lower than the sensitivity of TLD-100, and the TL sensitivity of the topaz–glass composite is twice as low as the sensitivity of TLD-100. The sensitivities of the topaz composites cannot be enhanced because it is not possible to significantly increase the concentrations of the topaz grains in the samples. The mechanical stability of the composites critically depends on the Teflon/topaz and glass/topaz concentration ratios.

Fig. 2 shows the fading of the TL glow curves for the topaz powder thermally treated at 400°C and irradiated with gamma rays (0.1 Gy). Fig. 2a provides the TL glow curves for different aliquots of the same sample measured after different time intervals between the irradiation and the read-out. In Fig. 2b, the area of the total TL curve and the area of the second TL peak are plotted as a function of that time interval. While the whole area decreases to 83% of its initial intensity after 60 days, the decrease in the area of the second TL peak is only 9%. In 30 days after irradiation, the second TL peak is only 7% weaker than immediately after irradiation, which is well below the 10% limit acceptable for dosimetry applications (CEI/IEC 1066, 1991). As can be seen from Fig. 2a, the fading is mainly due to the low temperature peak, which is not normally used in dosimetric applications.

In a previous paper (Souza et al., 1997), we showed that colourless natural topaz glow curves feature at least 6 peaks in the range from room temperature to 400°C, which overlap to produce 2–4 apparent TL peaks. The number of these "apparent" peaks depends on the thermal treatment of the samples. For the topaz–Teflon composites, the maximum temperature in TL measurements was about 330°C, and the high-temperature peaks of topaz could not be clearly observed. The topaz–glass composites, however, are thermally stable, and their high-temperature peaks are shown in Fig. 1. The observed differences between the topaz–Teflon and the topaz–glass composites in the peak positions and intensities cannot be attributed to the differences in topaz compositions, because the microcrystals used in both the composites were from the same sample batch (the same material as we used in our previous work, Souza et al., 1997). They can be explained, however, on the basis of our previous results. The thermal history of the microcrystals contained in both the composites is not the same, as the sintering processes used for both the composites were different. That resulted in different intensities of the 6 overlapping TL peaks, which produced the apparent TL peaks at different positions.

Souza et al. (1997) showed that the dose dependence of the area under the TL glow curve of the topaz–Teflon composites irradiated with gamma rays (^{60}Co) has the following features: (i) the dose dependence is sublinear for the low doses (10^{-4}–10^{-2} Gy), (ii) the dose dependence has a linear range from 10^{-2} to 20 Gy, followed by a supralinear range and then, again, by a sublinear range. It reaches saturation at doses about 2000–3000 Gy. These results indicate that the topaz–Teflon composites are suitable for various applications in TL dosimetry.

A number of different samples of the topaz–Teflon and topaz–glass composites were prepared, irradiated, and measured under the same controlled conditions. The results of this procedure are shown in Fig. 3. The reproducibility of the topaz–Teflon composites is better than the topaz–glass dosimeters. The average TL areas obtained by fitting the histograms to Gaussian-type functions were (115 ± 5) a.u. and (68 ± 6) a.u. for the topaz–Teflon and topaz–glass samples, respectively. This indicates that the process of the topaz–Teflon sample preparation is better controlled and improvements are needed in the process of the topaz–glass composite preparation. Nevertheless, for both



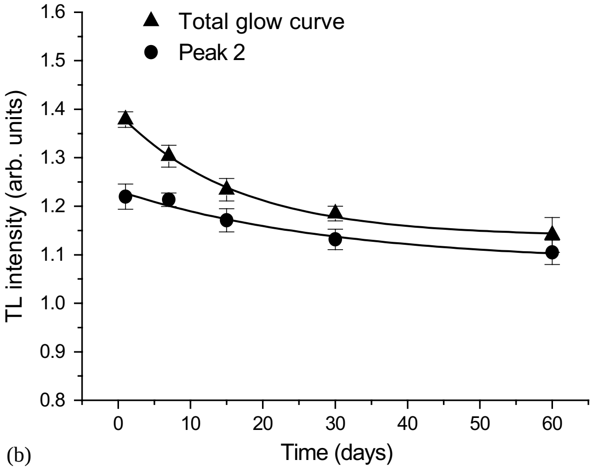


Fig. 2. (a) TL glow curves of the topaz–Teflon pellets measured at different time intervals after irradiation (0.1 Gy), (b) decrease in the total area under the TL glow curve and in the area of Peak 2 at room temperature. The lines show the exponential functions fitted to the experimental data.

composites, a better reproducibility can be easily achieved if dosimeters are pre-selected before use (one should expect that most of the topaz–Teflon samples will be selected while a reasonably large number of the topaz–glass samples will be rejected).

Fig. 4 shows the effect of reusing the topaz–Teflon dosimeters. A number of successive cycles of irradiation—reading—annealing were performed, and the TL intensities of peaks 1 and 2 were plotted as a function of the cycle number. The experiment was conducted in the following way: (i) a batch of samples was produced and irradiated with gamma rays to 1 Gy, (ii) TL readings of all the dosimeters were taken under the same experimental conditions (heating rate and maximum temperature), (iii) the samples were thermally treated at 300°C/h; (iv) steps i–iii were repeated, and the average values of the TL peak intensities were calculated in each cycle. The reproducibility of the TL glow curve is very good: the TL intensities remain unchanged up to 10 cycles.

Batches of the previously selected topaz–Teflon and topaz–glass dosimeters were irradiated with gamma rays (^{60}Co) in the following way: (i) 10 different dosimeters were attached to a 5-mm thick acrylic plate and irradiated to 1 Gy in a $10 \times 10\,\text{cm}^2$ field, (ii) an extra set of acrylic plates of various thicknesses was used to obtain different thicknesses of acryl between the source and the dosimeters, (iii) for each thickness, a new batch of dosimeters was used, and the average of the TL glow curve areas was plotted as a function of the thickness of acryl (Fig. 5). For comparison, the depth dose profiles obtained with an equivalent tissue material (BJR, 1983) are also shown. In the case of the topaz–Teflon dosimeters, the depth dose profile shows a very good agreement with the profile of the equivalent tissue material. For the topaz–glass composites, the agreement

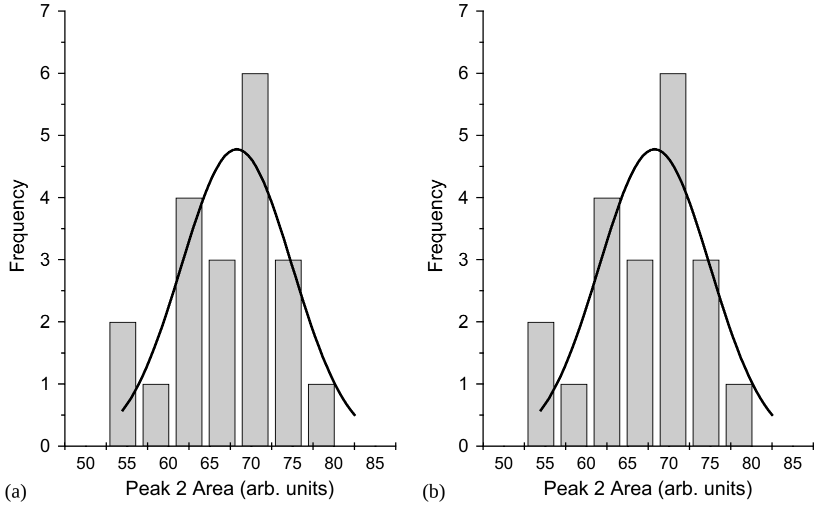


Fig. 3. Histograms of the distributions of the Peak 2 areas for the topaz–Teflon (a) and topaz–glass (b) composites. The data correspond to different samples that were produced, irradiated and measured under the same experimental conditions.

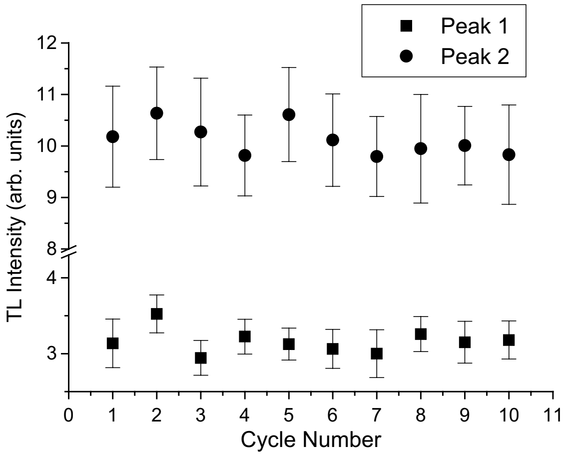


Fig. 4. Intensity of the two main TL peaks of the topaz–Teflon composite as a function of the number of irradiation—reading—annealing cycles. The composites were always irradiated to the same test dose (1 Gy) and measured under the same conditions.

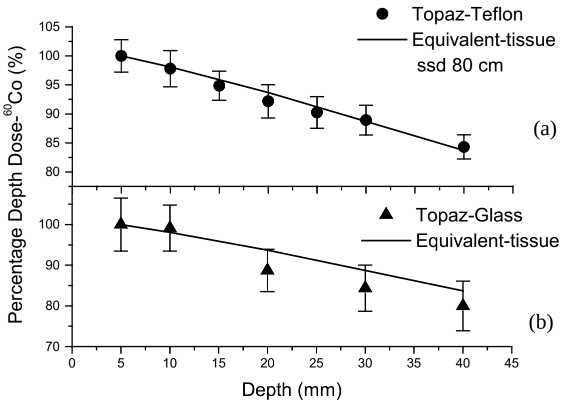


Fig. 5. Dose depth profiles for the topaz–Teflon (a) and topaz–glass (b) composites mounted in acrylic plates of different thicknesses compared with the equivalent-tissue material depth profile.

is not as good as for the topaz–Teflon composites, but it is still reasonable. These results demonstrate that a combination of the topaz-based composite dosimeters with acrylic plates can be a good simulating device for evaluating depth dose profiles in human tissues.

4. Conclusions

The following conclusions can be drawn from the results: (i) The composites of topaz with Teflon or glass are good candidates for TL dosimeters providing TL emission intensities comparable to those of the TLD-100 dosimeters. Their advantages are TL peaks at higher temperatures and a lower cost. (ii) The fading of the TL peak 2 at room temperature is about 7% in 30 days, which is well below the limits of fading acceptable for dosimetry. (iii) The topaz pellets can be used in successive irradiation—reading—annealing cycles without an appreciable change in their sensitivity. (iv) The depth dose profiles obtained with an arrangement of the topaz-based composite dosimeters can be directly compared to the depth dose profiles in human tissue. These conclusions, combined with our previous results, indicate that the composites prepared from natural colourless topaz are very promising new solid-state TL dosimeters. They are suitable for various applications, such as personal dosimetry, where low doses have to be measured, or radiotherapy, where doses between 0.1 and 100 Gy are normally used. They are also suitable for use in quality control programs for radiation emitting devices, where fairly high doses may be provided.

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