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Characterization of α-spodumene to OSL dosimetry

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HIGHLIGHTS

- The α-spodumene can be exploited for high-doses OSL dosimetry.
- The α-spodumene has linear OSL response over the dose range from 30 Gy to 10 kGy.
- The reproducibility of the OSL response of α-spodumene/Teflon® pellets is 2.1%.

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ABSTRACT

The aim of this paper is to evaluate the optically stimulated luminescence (OSL) response of spodumene ($LiAlSi_2O_6$) silicate and its potential use for gamma radiation dosimetry. A natural Brazilian crystal of α-spodumene was used in this study. After the crystal grinding, pellets with the diameter of 6.0 mm were prepared using a mixture of α-spodumene and polytetrafluoroethylene (Teflon®) (1:2). To study the OSL response, the samples were irradiated with gamma radiation beam of Co-60 and the response was measured using an OSL Reader in two modes of operation: Continuous-wave and pulsed stimulation. The results of the integrated OSL curve of the pellets irradiated with the dose of 30 Gy showed that their response is reproducible within ±2%. The variation of the OSL response upon the dose exhibits a linear response in the range from 30 Gy to 10 kGy, with a correlation coefficient of 0.99. It is possible to conclude that the α-$LiAlSi_2O_6$/Teflon® has a great potential for OSL dosimetry of high gamma doses.

1. Introduction

Luminescence applied to radiation dosimetry is used in different application fields. Recently the optically stimulated luminescence (OSL) became very popular and it is used in many areas of the applications of ionizing radiation. The OSL phenomenon corresponds to the light emission of a given material, previously irradiated, during stimulation with photon beams of specific wavelengths. The light emitted by the material is proportional to the received radiation dose. In optically stimulated luminescence (OSL) dosimetry the most used materials are quartz, feldspar and Al_2O_3:C (Yoshimura and Yukihara, 2006).

The OSL technique has different modes of stimulation and light collection. The main ones are the continuous and pulsed mode. In continuous mode, the sample is stimulated with a constant light source and the light emitted by the sample is collected simultaneously. In the pulsed mode, the sample is stimulated with light pulses and its luminescence is detected after the stimulation.

In this work the natural crystal of α-spodumene, lithium aluminum silicate ($LiAlSi_2O_6$), was chosen because it is relatively abundant in nature and present relatively high intensity of luminescent emission. Spodumene is a pyroxene mineral consisting of lithium aluminium inosilicate, $LiAl(SiO_3)_2$, and is a source of lithium. The normal low-temperature form α-spodumene is in the monoclinic system whereas the high-temperature β-spodumene crystallizes in the tetragonal system. Some of its thermoluminescence (TL) properties are already studied for applications in dating and ionizing radiation dosimetry (Brovetto et al., 1993; d'Amorim et al., 2012; Salis, 1995; Souza et al., 2004; Souza et al., 2007), but no studies discuss its OSL response. For this reason, the aim of this work is to study its OSL response and evaluate its application for dosimetry.

2. Experimental

A natural crystal of α-spodumene from Minas Gerais, Brazil, was investigated. After grinding the natural crystals in an agate mortar,

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the grains between 75 and 180 μm were selected to prepare the α-spodumene/Teflon® pellets, with 0.2 mm thick and 6.0 mm in diameter, using a mixture of α-spodumene and polytetrafluoroethylene (Teflon®) (1:2) according to the procedure described by d'Amorim et al. (2012).

X ray diffraction (XRD) of natural grains of the α-spodumene was performed to confirm its structure. The measurements were performed with a powder diffractometer PANalytical (XPert Pro MPD), with CoK_α radiation (λ=1.7889 Å), with the tube operating at 40 kV/30 mA in the continuous mode with steps of 2° min^{-1} at room temperature. The identification of the crystalline phases was analyzed by the Rietveld refinement by the program FullProf (Young et al., 1995). The Rietveld refinement refers to adjustment the model parameters used in calculation of a diffraction pattern which is closest to the observed (Will, 2005). The refinement is carried out by minimizing the sum of the differences between the calculated and observed intensities.

To study the OSL response, the pellets were irradiated with gamma source of Co-60 with a dose rate of 5.20 kGy/h in March/2012 and the response was measured using a custom made OSL Reader, utilizing blue LEDs for the stimulation of the samples. The response of this OSL reader was evaluated and compared to the Risø OSL reader. The OSL measurements were performed in two modes of operation: continuous-wave (CW) and pulsed (POSL) stimulation.

To evaluate the sensibility of the OSL response of a batch of 23 dosimeters, they were irradiated simultaneously with a dose of 30 Gy. Five cycles of optical treatment-irradiation-reading were performed and then the mean value of readings (x) and the respective standard deviations (s) were calculated. The coefficient of variation (*CV*), also known as relative standard deviation was calculated by the ratio of the standard deviation σ to the mean μ:

$$CV = \frac{\sigma}{\mu} \quad (1)$$

It shows the extent of variability in relation to mean of the population.

3. Results and discussion

Fig. 1 presents the XRD pattern of the powder of α-$LiAlSi_2O_6$. The XRD patterns were compared to experimental parameters reference ICSD 9668, being verified that the used sample corresponds to α-spodumene. This XRD pattern also is described by d'Amorim et al. (2012). The α-spodumene belongs to the monoclinic crystal system having the space group C2/c. The crystallographic parameters are: a=9.4490 Å, b=8.3860 Å, c=5.2150 Å, α=90°, β=110° and γ=90°.

The result of the Rietveld refinement was obtained from the XRD analysis and calculated by the least squares method and expressed as index indicating disagreement, in percentage. This index is as values of permitted error (R_P), obtained error (R_{WP}), expected error (R_E) and the ratio R_{WP}/R_E (or, simply, S) (Young et al., 1995). The S value is of critical importance; S=1 means that the calculated spectrum is perfectly adjusted to the experimental spectrum. Distortions of quality measured during the sample preparation can lead to the significant value of high-R_E, which is empirically considered excellent if below 10%.

Fig. 2 presents the results of the OSL response of the 23 pellets of α-$LiAlSi_2O_6$/Teflon® irradiated with the absorbed dose of 30 Gy. Each point corresponds to the average of five measurements with the indication of the standard deviation. The solid line represents the overall average of the measurements; the outermost dotted lines correspond to the range ($\overline{x} \pm 2s$) while the innermost represent the range ($\overline{x} \pm 1s$). Considering these results, the pellets that presented the variation coefficient (*CV*) above 10% were removed from the group. This acceptance criterion is established by ISO 12794:2000 standard (ISO, 2000), according to which the seven pellets were removed. Then the pellets that showed an average value out of the range of ($\overline{x} \pm 1s$) were checked. In this case no pellets were discarded. Thus, the total number of selected pellets for the OSL study was 16. Their new average value and standard deviation were calculated. The corresponding *CV* was 2.1%.

Fig. 3 shows the OSL signal of α-$LiAlSi_2O_6$/Teflon® irradiated with different doses, obtained in the continuous stimulation mode during 4000 s. The analysis of the OSL signal indicates that it is composed of two components: a fast (1), assigned to electrons that recombine directly with holes, and a slow (2), due to the presence of shallow traps in the structure where electrons are retrapped for several seconds before being recombined with holes.

Fig. 3 also shows that the decay rate becomes slower with increasing dose. Yukihara et al. (2004) observed that the rate of OSL decay curves increases with increasing dose in samples of Al_2O_3:C, irradiated with a beta source of Sr-90/Y-90. The opposite phenomenon, observed here, may be explained by the different ionization power of gamma and beta radiation. Beta radiation produces locally a large amount of ions in comparison to gamma radiation, corresponding to the component (1) of OSL curve. Thus, it is possible to conclude that the center responsible for component (1) is occupied firstly, becoming saturated in the region of the high ionization. Due to this

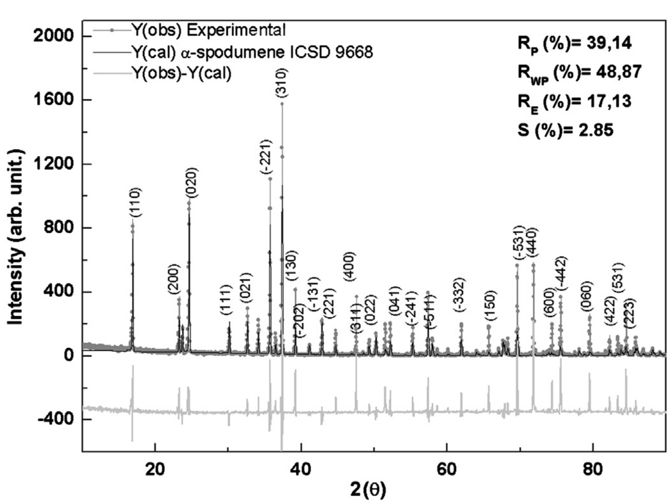

Fig. 1. *Y*(obs) X-ray diffraction pattern from a sample of natural spodumene and *Y*(cal) spectra calculated by Rietveld method. The result of refinement is the difference *Y*(obs)−*Y*(calc).

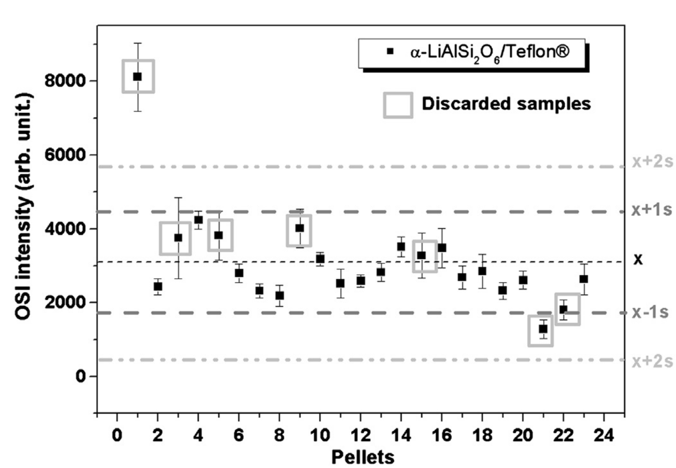

Fig. 2. OSL reproducibility of α-LiAlSi2O6/Teflon® irradiated with absorbed dose of 30 Gy with Co-60.