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Correlation of optically and thermally stimulated luminescence of natural fluorite pellets

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HIGHLIGHTS

- CaF_2:NaCl pellets are extremely useful as dosimetric materials.
- Only part of the defects responsible for the TL signal from fluorite is optically active.
- NaCl signals affects the TL and OSL signals from the pellets.
- NaCl OSL signal can be avoided with a pre-heat or after about 1 h waiting time.

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ABSTRACT

Natural fluorite (CaF_2), a dosimetric material of large usage, presents Thermoluminescence (TL) and Optically Stimulated Luminescence (OSL). This study examined the behavior of TL and OSL (stimulated with Blue LEDs) signals from the Brazilian natural fluorite pellets with NaCl as binding agent, as well as their correlations, in order to study and optimize the dosimetric process with this material. A series of experiments were conducted, basically with thermal treatments before OSL acquisition, and optical bleaching before TL readout. The role of NaCl in the TL and OSL emission was investigated. It was observed that natural CaF_2 TL signal is still ample to be used in dosimetric applications, as dose re-assessment in personal dosimetry after an OSL measurement. Also it was verified that the fluorite OSL signal is extinguished by a 350 °C heating and that NaCl has no contribution to the stable part of the OSL signal.

1. Introduction

Both natural and synthetic calcium fluoride are widely used as dosimetric TL material mainly due to their high sensitivity. In particular, the use of natural fluorite has extended to many places because of its abundance and low cost. In Brazil we have developed a solid dosimeter, based on natural calcium fluoride and sodium chloride powders pressed together (CaF_2:NaCl) (Okuno et al., 1977; Trzeniak et al., 1990) for dosimetric applications. These pellets are extremely useful as dosimetric materials because they can provide information on dose and radiation energy with the use of appropriated filters (Guimarães and Okuno, 2003). Recently, it has been observed that both Indian (Sunta, 1970; Chougaonkar and Bhatt, 2004) and Brazilian (Yoshimura and Yukihara, 2006) fluorite also emit OSL when blue light is used for stimulation, and that the emitted light is related to the absorbed dose. In this paper we explore the OSL and TL properties of the fluorite pellets in order to verify the possibility of expanding the dosimetric capabilities with the use of combined OSL and TL emissions. Studies of correlation of OSL and TL signals in fluorite are rare (Polymeris et al., 2006; Chougaonkar and Bhatt, 2004; Bakshi et al., 2009). They show that not all the traps related to TL peaks are optically active and that both photo and thermal transfer processes may be present. The addition of NaCl and the production of pressed pellets with CaF_2 powder may affect the correlation of OSL and TL signals. This occurs because the NaCl is itself a luminescent material and the pellet thickness (1 mm in average) may prevent uniform illumination or homogeneous deposition of radiation energy in the pellet (Moralles et al., 2005). Thus, this work aims to investigate the influence of TL readouts on the OSL response and the effect of blue illumination in the TL intensity of natural CaF_2:NaCl pellet dosimeters. Thus, it is expected that a better performance of this dosimeter will be achieved.

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2. Materials and methods

The materials used in this work are natural fluorite and sodium chloride (≥99% pure). Natural fluorite acquired as-from-the-mine was transformed into powder as described by Trzeniak et al. (1990). Sodium chloride was used as binding agent. Aliquots of pure NaCl or of a mixture containing 60% CaF_2 and 40% NaCl were compressed to make 5 mm diameter × 1 mm thickness (~45 mg mass) pellets. The dosimeters are protected from humidity with storage in dry ambient or with plastic enveloping, to avoid influence of this condition in TL or OSL signals.

Before the first use and between subsequent uses of the same sample in a experiment, the sample was heated at 5 °C/s up to 450 °C, as in a TL acquisition, to empty the traps and erase the TL and OSL signal. The variation of NaCl and CaF_2:NaCl pellets sensitivity to repeated irradiation-readout (TL/OSL) cycles was tested over 100 cycles and the statistical dispersion is compatible with the uncertainties presented, without trends.

Two groups of ten samples each were selected in accordance with the signal intensity after the same irradiation time under the beta source. One group of pellets was made of pure NaCl and the other of CaF_2:NaCl (60:40); each group of selected pellets presented 3.0% standard deviation of the mean OSL signal. The experimental data that are shown in this work are the mean value from these group measurements with their respective uncertainties.

OSL and TL measurements were carried out using a commercial automated TL/OSL reader produced by Risø National Laboratory (model DA-20). Luminescence was stimulated using blue light emitting diodes (470 nm, FWHM = 20 nm) delivering 80 mW/cm^2 at the sample position. The TL/OSL signals were detected with a bialkali photomultiplier tube (PMT) behind an UV transmitting broad-band glass filter (Hoya U-340, 7.5 mm thick × 45 mm diameter) to block the stimulation light while transmitting part of the OSL signal from the samples. Irradiations were performed at room temperature using the built-in $^{90}Sr/^{90}Y$ beta source of the TL/OSL reader (dose rate of 11 mGy/s).

The stimulation time for OSL measurements was 100 s. Integrated OSL signals were calculated as first 50 s counts considering the last 50 s as background. TL glow curves were obtained using a heating rate of 5 °C/s.

3. Results and discussions

The experiments described below involve a series of irradiation-readout cycles. Some types of NaCl have shown vulnerability to these cycles, as shown by Polymeris et al. (2011). However, the TL glow curves from the NaCl samples used on this work show no peaks at temperatures higher than 300 °C, as can be seen in Fig. 1. These TL glow curves are more similar to those presented by Ekendahl and Judas (2011) than by Polymeris et al. (2011) and may explain that no change of sensitivity of NaCl pellets to irradiation-readout cycles was observed in our work.

Fig. 1 shows residual TL glow curves for CaF_2:NaCl pellets after CW-OSL with different illumination times. The results for a NaCl pellet are in the insert, and we see that the TL signal is restricted to the low temperature region, and is completely extinguished after a light stimulation of just 5 s. Almost all the signal of the fluorite pellet comes from fluorite, and part of the TL is bleached by the optical stimulus.

For a more complete analysis, we show in Fig. 2 the difference between the TL glow curve from a non-illuminated sample and TL glow curves after different illumination times. With this data it is possible to conclude that the shallow traps responsible for the low temperature part of the glow curve (below 150 °C) are easily bleached out during the OSL readout – less than 5 s of illumination

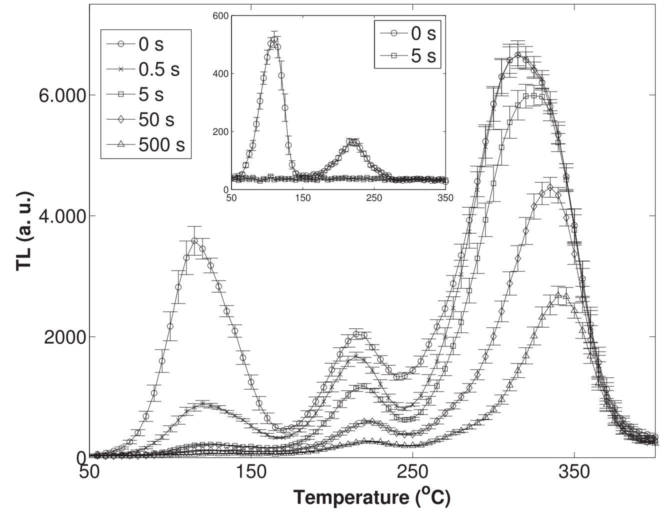

Fig. 1. TL glow curves of CaF_2:NaCl pellets obtained after different optical stimulation times. The lines connecting points are just guides for the eyes. The insert shows the TL signal of a NaCl pellet, freshly irradiated to the same dose, and after blue light stimulation during 5 s.

are necessary to remove these peaks from the glow curve. As far as the fluorite TL dosimetric peak is concerned, the effect of optical stimulation is to bleach the low temperature part of the peak leaving a very stable signal, centered near 350 °C. In fact, after 500 s of illumination, a TL peak centered at 310 °C, usually a part of the main peak, is completely bleached out. A satellite peak at 350 °C survives the bleaching. Polymeris et al. (2006) have seen a similar effect in the study of natural fluorite, and have predicted the existence of such a peak through the deconvolution of the whole TL glow curve. Here we identify its presence by the bleaching out of the TL signal.

A better correlation of TL and OSL signals can be seen in Fig. 3 where the relative integrated TL signal of the fluorite pellets, from room temperature to T_{stop} is compared to the relative residual OSL signal after a preheating till T_{stop}. Again it is possible to observe that the NaCl contribution to the OSL signal is connected with shallow traps. Also, we see that the fluorite shallow traps concentrate the

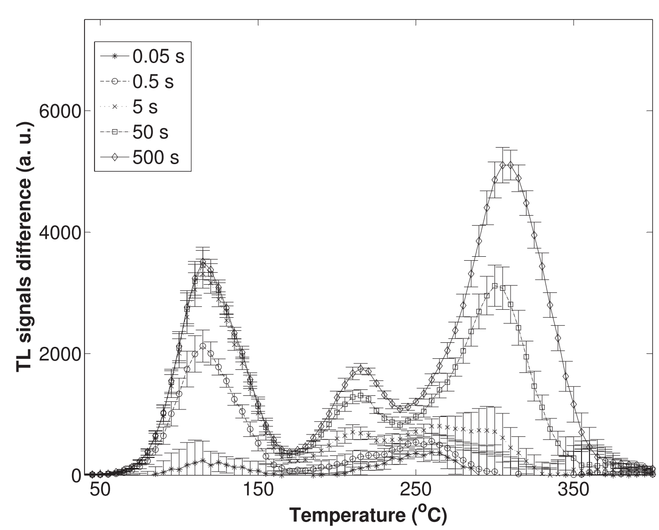

Fig. 2. Differences between the TL glow curve obtained without optical bleach and the TL glow curves after different stimulation times. The lines connecting points are just guides for the eyes.