Luminescence properties of feldspars from the Northeast region of Brazil

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Abstract. The aim of this study is to investigate the thermoluminescence (TL) and infrared-stimulated luminescence (IRSL) properties of three types of feldspars from the districts of Solonópole and Parelhas located in the Northeast region of Brazil in order to propose a preheat procedure to minimize the anomalous fading effect in these materials. The feldspar samples were characterized by X-ray diffraction (XRD) and X-ray fluorescence spectrometry (XRF). The XRD analysis showed that the feldspar from Solonópole and one of the samples from the Parelhas district were classified as microcline (K-feldspar). The other sample from Parelhas was classified as albite (Na-feldspar) mixed with low concentration of muscovite and quartz. Studies of the fading effects in TL and IRSL signals for 30 days and preheating effects from 50 to 250°C for 10 minutes were carried out with these samples. The results show that preheating at 125°C is sufficient to avoid the 30 days fading. The TL and IRSL dose responses for ⁶⁰Co gamma radiation were studied in the range of 1 to 400 Gy. The results showed a linear response in the range of 1 to 100 Gy for the two types of Parelhas feldspars and from 50 to 400 Gy for the Solonópole feldspar. The low sensitivity showed by the feldspar from Solonópole is discussed in relation to its mineralogical composition. It is concluded that the TL and IRSL sensitivities are not associated with K- and Na- types of feldspar. The results of this paper will contribute to a standardized moderate annealing treatment for retrospective dosimetry and dating.

1. Introduction

The luminescence emission properties of quartz and feldspars have been widely used in dosimetry. Natural quartz and feldspar are generally chosen as dosimeters for radiation accidents since they are substances abundantly found on the earth’s crust and are used in the fabrication process of building materials such as roofs, bricks, tiles and many ceramic objects [1, 2]. In the last decade, most retrospective luminescence dosimetry research has focused on developing methods based on quartz. It was observed that the quartz dose response curve generally saturates at 200 Gy, limiting its use for dating studies to approximately 100-150 ka [3].

Feldspars are another type of natural luminescence dosimeters. They have many advantages compared to quartz, that include; a higher intrinsic luminescence sensitivity resulting in a lower...
detection limit; a luminescence signal which has been shown to saturate at a higher dose than that of quartz, and, unlike quartz, they are sensitive to IR stimulation [4, 5]. Unfortunately, the feldspar luminescence signal decreases with time faster than expected from a consideration of trap depth and ambient temperature. This feldspar anomalous fading is weakly dependent on temperature [6, 7].

The aim of this paper is the evaluation of TL and IRSL responses of three types of Brazilian feldspars and the effect of preheat temperature in TL and IRSL curves, in order to propose a preheat procedure to minimize the anomalous fading effect in these materials. The results of this paper will contribute to a standardized moderate temperature treatment for retrospective dosimetry and dating.

2. Materials and methods

The feldspars used in this study are from Brazil; they were taken from two deposits located in the Parelhas district (Rio Grande do Norte State) and from one deposit located in Solonópole district (Ceará State). The rocks were crushed with a laboratory grinder and carefully pulverized using a mortar and pestle. The grains were sieved and the particle size classified. Sizes under 75 μm were used for mineralogical characterization by X-ray diffraction (XRD) and X-ray fluorescence spectrometry (XRF). The mineralogical phases were determined by XRD with a Bruker-AXS D5005 instrument using Co Kα radiation. The XRD patterns were obtained scanning from 5° to 80° (2θ) in steps of 0.02° with a measurement time of 1 s per step. Each pattern was analyzed using a Bruker DiffracPlus software comparing the sample with a standard in the PDF02 database (ICDD, 1996) [8]. The semi-quantitative analysis of the oxides in each sample was carried out with a Rigaku-RIX 3000 XRF spectrometer, equipped with a Rh tube.

Feldspar fractions between 75 and 150 μm were selected for TL and IRSL studies. These samples were annealed at 500 °C for 1 hour in a programmable temperature-controlled muffle-furnace at atmospheric pressure. The samples were separated in aliquots of 150 mg, and placed in opaque bags, to be protected from the ambient light. The feldspar aliquots were irradiated with a 60Co Gammacell.

For TL and IRSL fading studies, 1000 mg of each type of feldspar were irradiated with 100 Gy and were stored in the dark, at room temperature. The TL and IRSL signals of the 150 mg irradiated aliquots were read after different periods, ranging from 1 hour to 30 days. In order to evaluate the preheat temperature effect on TL and IRSL signals, a sample of each feldspar was irradiated with 100Gy. The aliquots of 150 mg were thermally treated for 10 minutes at different temperatures in the range of 50 °C to 250°C. After each heat treatment, the luminescence signals were read under the same conditions described above. To evaluate the dose response of the TL and IRSL curves, the aliquots were given a preheat treatment at 125°C for 10 minutes following each irradiation. The dose response curve for feldspars from Parelhas and Solonópole were, respectively, analysed at 1 to 100 Gy and at 50 to 400 Gy.

The TL signal was read using the Harshaw 3500 reader, with a heating rate of 4°C.s⁻¹ in a temperature range from 50 to 400°C. The IRSL measurements were performed at room temperature with a continuous wave stimulation (CW) instrument generated by infrared LEDs (λ=850 nm). The luminescence detection was made with a bialcaline Hamamatsu photomultiplier tube (PMT) using a BG-39 filter 2 mm thick [9]. The IRSL signal was defined as the integrated counts from the beginning of stimulation until reaching 50% of the maximum.

3. Results and discussion

The XRD patterns are shown in Fig.1. Using the PDF02 database it was possible to obtain the phase indexation of the feldspar samples. The results show that the feldspars from Solonópole (MS) and one of the feldspars from Parelhas (MP) present a microcline phase, and were classified as microcline (KAlSi₃O₈). The other sample from Parelhas (AP) was classified as albite ((Na,Ca)Al(Si,Al)₃O₈) mixed with low concentration of muscovite (KAl₂[Si₃AlO₁₀](OH,F)₂) and α-quartz (SiO₂).

Table 1 shows the XRF analysis. The MP and MS samples showed high concentration of K₂O confirming the potassium characteristic of these feldspars. The feldspar from Solonópole shows higher Fe₂O₃ concentration compared with the other samples. The high concentration of SiO₂ showed in the
AP sample is due to the presence of quartz as well as of muscovite, observed in the XRD analysis. The presence of muscovite in the AP sample can also explain the high percentage of K$_2$O compared with Na$_2$O in this sample.

![Diagram of XRD patterns](image)

**Fig. 1** - XRD patterns of feldspars from Parelhas (AP and MP) and Solonópole (MS) deposits (Co-K$_{\alpha}$ radiation).

| Sample | SiO$_2$ | Al$_2$O$_3$ | Fe$_2$O$_3$ | MgO | TiO$_2$ | CaO | Na$_2$O | P$_2$O$_5$ | K$_2$O | RbO$_2$ | Cr$_2$O$_3$ |
|--------|---------|------------|------------|-----|---------|-----|--------|----------|------|-------|---------|
| AP     | 72.2    | 16.7       | 0.5        | -   | -       | 1.9 | 1.0    | 2.7      | 2.0  | 0.02  | -       |
| MP     | 54.5    | 12.2       | 0.3        | -   | -       | 0.7 | 0.2    | 1.3      | 29.3 | 1.4   | 0.3     |
| MS     | 64.7    | 18.3       | 1.5        | 0.2 | 0.02    | 0.07| 1.4    | 0.04     | 13.9 | -     | -       |

- not determined

**Table 1** - Chemical composition of major constituents found in feldspar samples obtained by XRF

![Graph of TL and IRSL curves](image)

**Fig. 2** – (a) TL and (b) IRSL curves of feldspars AP, MP and MS irradiated with $^{60}$Co.

Fig. 2 (a) shows typical TL curves obtained for the three feldspars studied. The microcline samples (MP and MS) present a large TL peak with a maximum at about 100°C and a slow decay composed of several overlap peaks between 100 and 400 °C, in agreement with the literature [10]. The albite sample (AP), presents a broad peak and two additional peaks observed at 90 °C and 340 °C. These peaks are probably due to the presence of quartz in this sample, as shown in the XRD pattern. Fig. 2(b)
shows the IRSL decay curves for the feldspars, where it is possible to observe that the three samples present a typical IRSL signal decay.

Figure 3a shows the decay in the TL glow curve of the feldspar MP due to the fading at room temperature and in the dark, during the period of one month. The integrated TL signal from 50 °C to 400 °C after 30 days, was 50% lower than the signal on the first day for the MP sample, and 37% for the AP and 56% for the MS samples, respectively. These results are similar with those reported in the literature [6].

Fig. 3 (b) shows the MP feldspar IRSL signal decay as a function of the time elapsed after irradiation, where the fading effect is evident after 10 seconds. A similar behaviour was observed for the AP and MS samples. Integrating the IRSL signal counts from the beginning of the stimulation until the signal reached 50% of the maximum, the unwanted fading effect is minimized. In fact, after 30 days storage at room temperature, the integral IRSL signals did not change for any of the feldspar samples studied.

![Fig. 3](image-url)

(a) (b)

Fig. 3 – Fading effect at various storage times in (a) TL and (b) IRSL curves of MP feldspar.

Fig. 4 (a) shows the behaviour of the TL glow curve for the feldspar MP sample after different preheat treatments, ranging from 50 °C to 250 °C for 10 minutes, after irradiation. The effect of preheating in the other feldspars was normalized and summarized in Table 2. Preheating temperatures higher than 150 °C diminish significantly the TL signal of all the samples, and, hence, may only be used for applications with a long exposure time, such as in archaeological dating [11]. The effect of preheating in the IRSL signals of the MP feldspar is shown in Fig. 4 (b). It is observed that until reaching a preheating temperature of 150 °C, there is no significant alteration of the IRSL signal during the first 10 seconds of the luminescence decay. Preheat temperatures from 200 to 250°C affect significantly all the IRSL signals.

![Table 2](table-url)

| Sample | 50°C | 100°C | 125°C | 150°C | 200°C | 250°C |
|--------|------|-------|-------|-------|-------|-------|
| AP     | 100  | 53    | 44    | 38    | 17    | 10    |
| MP     | 100  | 35    | 26    | 21    | 6     | 2     |
| MS     | 100  | 32    | 22    | 15    | 5     | 1     |

Since the IRSL signal was defined as the integrated counts from the beginning of the stimulation until reaching 50% of the maximum, it is possible to conclude that fading (30 days after the irradiation) doesn’t influence significantly the integrated IRSL signal. However, for TL measurements, a preheat treatment at 125°C for 10 minutes was chosen as part of the protocol for these feldspar samples to simulate natural fading.
Fig. 4 – Preheat effect at various temperatures for 10 minutes in (a) TL and (b) IRSL curves of MP feldspar.

Fig. 5 shows the TL (5a) and the IRSL (5b) dose response curves. Results show a linear dose response, indicating that these feldspars can be used as natural dosimeters regardless of the type or origin. However, the AP and MP feldspars have higher sensitivity than the MS feldspar.

For doses above 25 Gy, a sub-linear behaviour of the IRSL curves for the Parelhas feldspar (MP and AP) samples was observed. This effect was eliminated reducing the PMT high voltage from 700 to 600 V. Thus, the sub-linear behaviour observed in Fig. 5 (b) was attributed to a decrease in the efficiency of the PMT rather than an effect of sample saturation. The PMT high voltage adjustment opens the possibility to use the equipment in a large range of doses and permits the investigation of samples with different sensitivities. In the MS feldspar case, which shows a weaker luminescence than the other samples, it is possible to measure the IRSL signal at doses under 50 Gy just by increasing the PMT high voltage.

4. Conclusions
The three feldspars showed many overlapping TL peaks between 50 and 400 °C. In addition, two peaks were observed in the AP samples due to the presence of quartz. The preheat study showed that treating the samples at 125 °C for 10 minutes eliminates part of TL unstable signals and thus minimize the fading effect for a 30 day storage time. This treatment of 125 °C for 10 minutes may be used to
standardize both TL and IRSL techniques, despite the fact that a restriction on the integration of the IRSL curve would be enough.

The results showed linear TL and IRSL responses of feldspars from Parelhas (AP and MP) at dose ranges from 1 to 100 Gy and from 50 to 400 Gy for the feldspar from Solonópole (MS). We believe that the higher concentration of iron oxide measured in the MS sample can explain its lower sensitivity because, with increasing total Fe$_2$O$_3$ concentration, the substitution of Fe$^{3+}$ at tetrahedral positions increases, whereas the concentration of Al-O-Al centres decreases [12]. Many papers [13, 14, 15, 16, 17] reported that the Al-O-Al centres were correlated with the UV-blue emission band and the Fe$^{3+}$ in the feldspar is associated with the red-IR emission. Due to the high concentration of Fe$_2$O$_3$, the TL emission will have a high contribution of the red-IR band, located in the lower PMT efficiency region.

The standardized moderate annealing treatment proposed in this study will facilitate TL and IRSL techniques using different feldspar types of different origin, thus contributing to retrospective dosimetry and archaeological dating.

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