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HIGH-DOSE DOSIMETRY USING NATURAL SILICATE MINERALS

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ABSTRACT

In the present study, certain natural silicate minerals such as aquamarine (AB), morganite (PB), goshenite (WB), white jadeite (JW), green jadeite (JG), pink tourmaline (PT) and two varieties of jadeite-like quartz, denoted here by JQ1 and JQ2, were investigated using the thermoluminescence technique to evaluate their potential for use as very-high- and high-dose dosimeters. These minerals respond to high doses of γ-rays of up to 1000 kGy and often to very high doses of up to 3000 kGy. The TL response of these minerals may be considered to be satisfactory for applications in high-dose dosimetry. Investigations of electron paramagnetic resonance and optically stimulated luminescence dosimetry are in progress.

1. INTRODUCTION

A number of materials have been investigated not only for low-dose dosimetry, which is useful in medical applications, but also for high-dose dosimetry, which is important in retrospective or industrial dosimetry [1-6]. There are various scenarios in which very high doses are involved, such as very-high-energy particle accelerators, nuclear power plants and very-high-dose irradiation facilities for food preservation or the modification of the properties of solid materials. One such type of detector is based on LiF, doped either with Mg and Ti or with Mg, Cu and P [7-9].

A group of physicists at the Institute of Nuclear Physics (IFJ), Krakow, Poland, has been investigating the effects of high radiation doses on LiF doped with Mg and Ti (called MTS) and with Mg, Cu and P (called MCP) since the beginning of this century; the low-dose regime has been under investigation at the IFJ since the 1960s in the case of MTS and the 1980s in the case of MCP materials. This group has tested these materials under irradiation with γ-rays, electrons, protons, alpha particles and also neutrons. Their most important result is that these materials, particularly the MCP materials, exhibit interesting and unexpected

behavior when irradiated with high doses of up to 500 kGy. A high-temperature TL peak is observed at above 300 °C, and this peak shifts to higher temperatures of up to 460-470 °C as the dose reaches approximately 500 kGy. These authors refer to this peak as peak B [10]. High neutron fluences on the order of 10^{15} per cm^2 also produce similar results. In the article published by Obryk et al. [11], references prior to that date can be found.

Camargo and Isotani [12] have measured the optical bands in natural pink tourmaline and have identified bands at 8500, 14800, 19500 and 25500 cm^{-1}, with intensity of all of them growing uo to 13000 kGy. These authors irradiated tourmaline at this very high dose value, and their results indicated that with further irradiation, these bands could continue to intensify. This is an extraordinary result; no other researchers have performed similar ultra-super-high-dose irradiation experiments.

In the study reported here, we investigated certain natural silicate minerals and found that some of them responded to high to very high radiation doses. Here, 'high dose' refers to a dose of up to 1000 kGy, whereas 'very high dose' refers to doses above this value; such doses are typically of up to 2000 or 3000 kGy, but in one case a much higher dose has been applied. In this work, we report high-dose dosimetry conducted using several silicate minerals; these materials were irradiated with γ-ray doses from a few tens of kGy up to 3000 kGy.

2. MATERIALS AND EXPERIMENTS

The following natural silicate minerals were acquired from stone dealers in Teofilo Ottoni, State of Minas Gerais, Brazil, for the present study: morganite (PB), goshenite (WB), aquamarine (AB), white jadeite (JW), green jadeite (JG), pink tourmaline (PT) and two varieties of jadeite-like quartz, (JQ1) and (JQ2). Here, we studied the TL of these crystals to determine their dosimetric behavior. For this purpose, we first crushed the samples and then sieved them, retaining grains of between 0.080 and 0.180 mm in size. These powders were then annealed at 500 °C for 30 min and subsequently irradiated with γ rays at doses in the range of hundreds of kGy up to 2000 kGy. The TL measurements were performed in a nitrogen atmosphere using a model 4500 Harshaw TL reader equipped with two photomultiplier tubes, which could record luminescence signals independently. The reader was controlled by WinREMS Software, which was supplied with the spectrometer and was run on a Windows computer. The heating rate used in the TL measurements was 4 °C/s. Each point in the glow curve represents an average of five readings.

3. RESULTS

Samples JG, JW, JQ1 and JQ2 we acquired as samples of jadeite. X-ray fluorescence analysis indicated that JG and JW were in fact jadeite, but JQ1 and JQ2 were much more similar, although not identical, to quartz. The JQ1 glow curve exhibited TL peaks at 110, 260 and 340 °C, whereas JQ2 presented peaks at 190, 260 and 340 °C. Both these materials were initially irradiated with γ rays up to 70 kGy. Subsequently, however, JQ2 was irradiated to a higher dose of 3000 kGy. Figure 1 presents the TL response of the 260 and 340 °C peaks as functions of the dose for JQ1 and JQ2. The maximum peak height was used to quantify the TL response. The TL glow curves of JQ1 and JQ2 samples that were pre-heated at 500 °C for 30 min and then γ irradiated at a dose of 2 kGy are presented in the insets of Fig. 1(a) and Fig. 1(b), respectively.

The JW sample exhibited peaks at 110, 190 and 240 °C, whereas JG exhibited peaks at 140, 210, 250 and 330 °C. The intensity of the peak at 190 °C in JW increased by two orders of magnitude as the dose increased from 50 Gy to approximately 10 kGy and then saturated (see Fig. 2a). Figure 2(b) presents the behavior of the TL intensity (based on the peak height) as a function of dose; it is observed that the 140 °C peak grew slowly with increasing dose, whereas the 210 °C peak grew more rapidly and 330 °C peak grew very rapidly with increasing dose. The response of this last peak increased by nearly three decades as the dose was increased to approximately 20 kGy. With the exception of the 330 °C TL peak, all other TL peaks grew linearly for doses of up to 200 Gy and then supralinearly for γ doses of up to approximately 20 kGy, saturating thereafter. The linear-supralinear behavior and the stability at room temperature of the TL peaks at 210, 240 and 330 °C allows the use of natural jadeite as a dosimetric material.

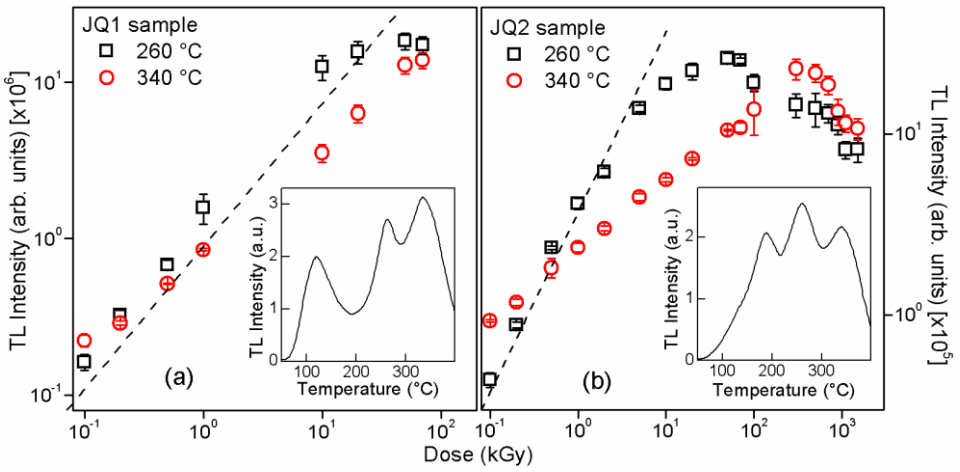

Figure 1: TL intensity versus γ dose for the TL peaks of (a) JQ1 and (b) JQ2; the dashed line indicates linearity. In the insets are presented the glow curves of JQ1 and JQ2 samples irradiated with 2 kGy of γ-rays.

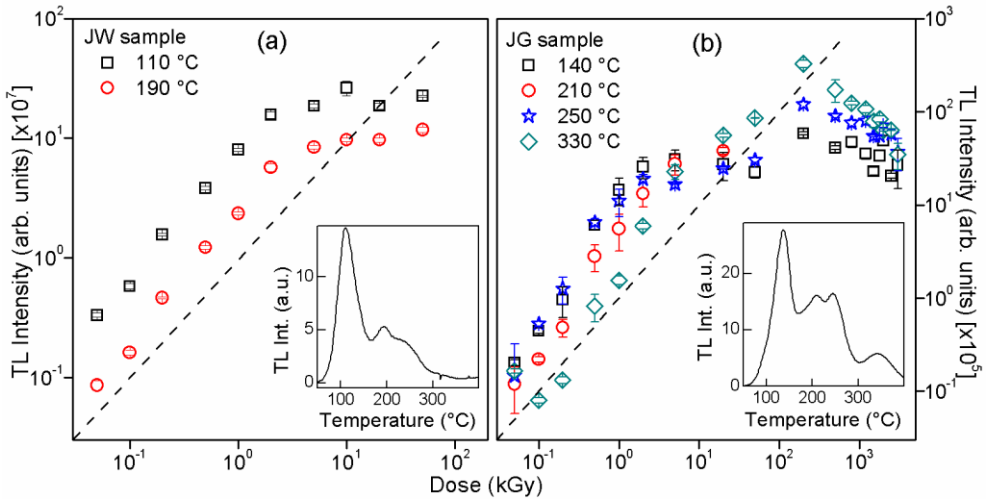

Figure 2: TL intensity behavior of the TL peaks as a function of γ radiation dose for the JW and JG samples, respectively; the dashed line indicates linearity. In the insets are presented the TL glow curves of JW and JG samples pre-annealed at 500 °C in air and then irradiated with a γ dose of 2 kGy.

The TL glow curves of beryl samples that were pre-annealed at 500 °C for 30 min and then irradiated to a γ dose of 10 kGy are presented in Fig. 3(a). Figure 3(b,c,d) presents the TL response curves as functions of dose for all three studied types of beryl: PB, WB and AB. In PB, the TL response saturated at approximately 5 kGy; in WB, the TL response described a parabola-like with a maximum at approximately 1250 kGy and then decreased beyond this dose value, although the TL intensity at 2000 kGy remained high. In AB, the TL response increased for doses of up to approximately 250 kGy, rapidly decreased for doses of up to approximately 1300 kGy and then decreased slowly beyond that point.

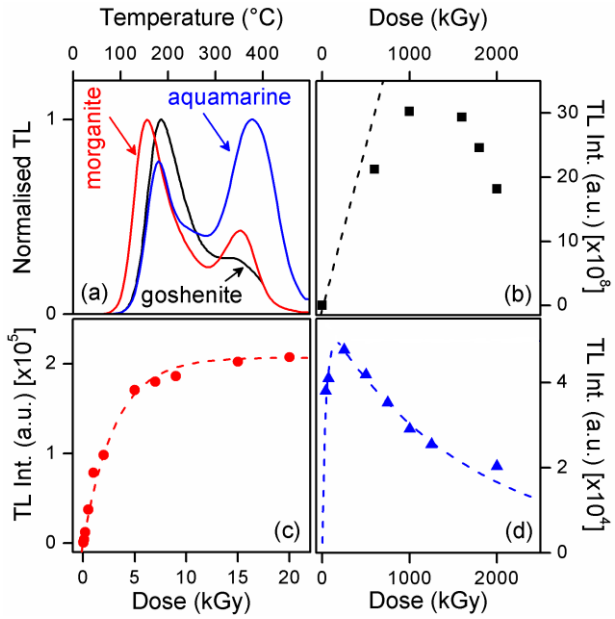

Figura 3. (a) TL glow curves of morganite, goshenite and aquamarine and TL response curves as functions of radiation dose of (b) goshenite, (c) morganite and (d) aquamarine.

Figure 4 reveals an interesting result. For a sample of pink tourmaline irradiated at high doses, the TL reading reached a high value of close to $9x10^8$ a.u. for a dose close to 100 kGy, but when this sample was irradiated at a very high dose, from hundreds of kGy up to 3000 kGy, the TL response exhibited a well-behaved decrease in value (curve). The inset of Fig. 4 presents the TL glow curves of pink tourmaline irradiated at doses varying from 10 to 1000 Gy. The TL versus dose curve is already shown in Fig. 4. It is observed that the TL intensity is still increasing for doses of >1000 Gy, indicating that the TL growth curve extends into the region corresponding to hundreds of kGy

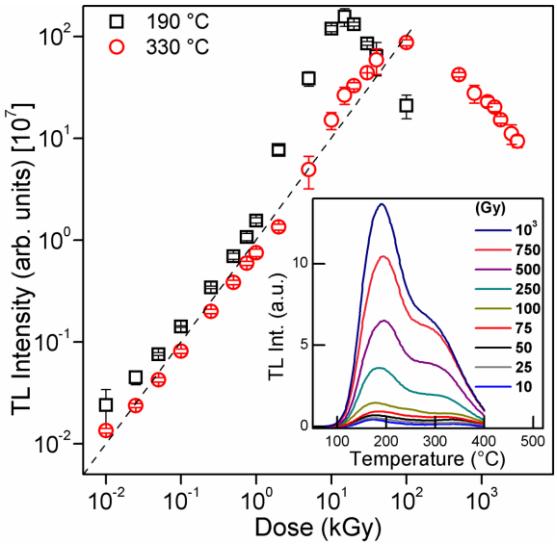

Figura 4. TL response curves as functions of radiation dose for pink tourmaline. In the inset are presented the TL glow curves of pink tourmaline irradiated with 10, 25, 50, 75, 100, 250, 500, 750 and 1000 Gy of γ-rays.

4. COMMENTS AND CONCLUSION

Although JW and JG were both jadeite crystals and morganite, goshenite and aquamarine were all beryl crystals, they presented distinct TL behaviors under heavy irradiation. They differed in the impurities they contained, but it is difficult to attribute the various behaviors described above solely to impurities. The differences among the responses of these materials require further study.A summary of the saturation doses of the investigated thermoluminescence dosimeters, as determined in the present study, is provided in Table 1. All silicate minerals that were investigated here exhibited dose-dependent TL responses upon high- or very-high-dose (γ) irradiation at doses up to the order of 2000 kGy, but in the case of JG and pink tourmaline, this behavior extended to 3000 kGy; in other words, these materials can be used for very-high-dose dosimetry.

In the case of tourmaline, both the 190 and 330 °C peaks increased in intensity with increasing dose for doses of up to approximately 10 and 100 kGy, respectively, and then began to decrease, describing a parabola-like similar to those observed in the JQ samples, JW and JG. In all these cases, the TL response was observed for high (1000 kGy) to very high doses (2000 to 3000 kGy).

In the case of a TL versus dose curve with one increasing branch (A) and one decreasing (B) branch, if the measured TL value could be representative of either (A) or (B), we offer the following suggested method of distinguishing the correct dose in the ambiguous region of the calibration: use at least two aliquots to measure TL, one for the actual TL value measurement and the second subsequently irradiated with an additional dose. If an increase in the TL response is measured, the original measurement lies on the (A) branch; otherwise, it lies on the (B) branch. In this manner, the entire TL vs. dose curve can be used for high-dose dosimetry.

Table 1 TL peaks and dose response of investigated materials.

Material	TL peak (°C)	Saturation dose
JQ1	260	70 kGy
	340	70 kGy
JQ2	260	70 kGy
	340	500 kGy
JW	110	10 kGy
	190	10 kGy
JG	140	20 kGy
	210	20 kGy
	250	20 kGy
	330	200 kGy
AB	380	250 kGy
PB	330	5 kGy
WB	330	1000 kGy
PT	190	10 kGy
	330	100 kGy

5. ACKNOWLEDGEMENTS

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