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Radiation dosimetry using decreasing TL intensity in a few variety of silicate crystals

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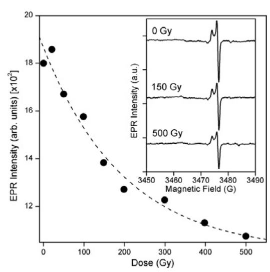
HIGHLIGHTS

- The TL dose responses of four different natural silicate minerals have been investigated.
- After the saturation dose, all minerals show a decreasing TL intensity signal with gamma ray dose.
- These crystals may be used to measure gamma dose that varies from low values up to 400–500 Gy.

GRAPHICAL ABSTRACT



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ABSTRACT

This study shows that there are some ionic crystals which after irradiation with high gamma dose D_m and subsequent irradiation with low doses ranging up to 500 Gy present a decreasing TL intensity as dose increases. This interesting feature can be used as a calibration curve in radiation dosimetry. Such behavior can be found in green quartz, three varieties of beryl and pink tourmaline. In all these silicate crystals it can be shown that irradiation with increasing γ-dose there is a dose D_m for which the TL intensity is maximum. Of course, D_m varies depending on the crystal and irradiated crystal with the dose D_m is stable. If one of these crystals is taken and irradiated with doses from low values up to 400–500 Gy, a curve of decreasing TL intensity is obtained; such a curve can be used as a calibration curve.

1. Introduction

The Radiation Dosimetry based on some ionic crystals had a considerable progress since LiF:Mg,Ti was introduced successfully for that purpose around 1950. Many other crystals have been explored and some of them gave very good result, particularly for routine dosimetry (Mesterházy et al., 2012; Teixeira et al., 2011; Vila and Caldas, 2011; Inrig et al., 2008; Rocha et al., 2003; Wieser et al., 1994). In the last two decades we investigated natural silicate minerals and found many of them exhibiting high sensitivity (Watanabe et al., 2015; Barbosa et al., 2014). The physical properties that were used are thermoluminescence (TL) and electron paramagnetic resonance (ESR). In few cases optical absorption was also used (Camargo and Isotani, 1988). In the usual TL or EPR dosimetry, the TL or EPR intensity is measured as function of radiation dose. The common feature is that the TL or EPR intensity increases with the dose, linearly for low doses, but different kind of dependence can be observed at high doses as reported by Watanabe et al. (2015). In quartz it is usual to deal with E'_1-center, which is formed in the following way. In quartz it is known that stable oxygen vacancies are formed even at RT (Feigl

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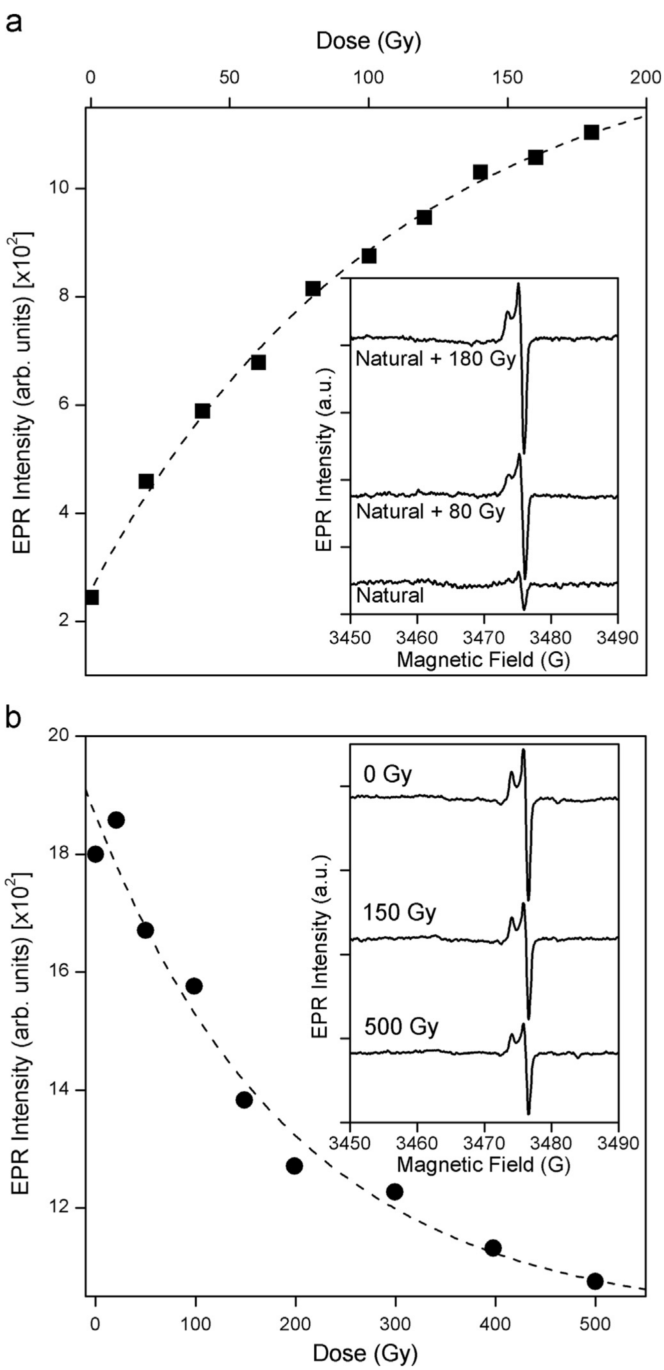

Fig. 1. (a) EPR intensity of E′$_1$-center as a function of dose. (b) EPR intensity of E′$_1$-center in 500 Gy irradiated green quartz.

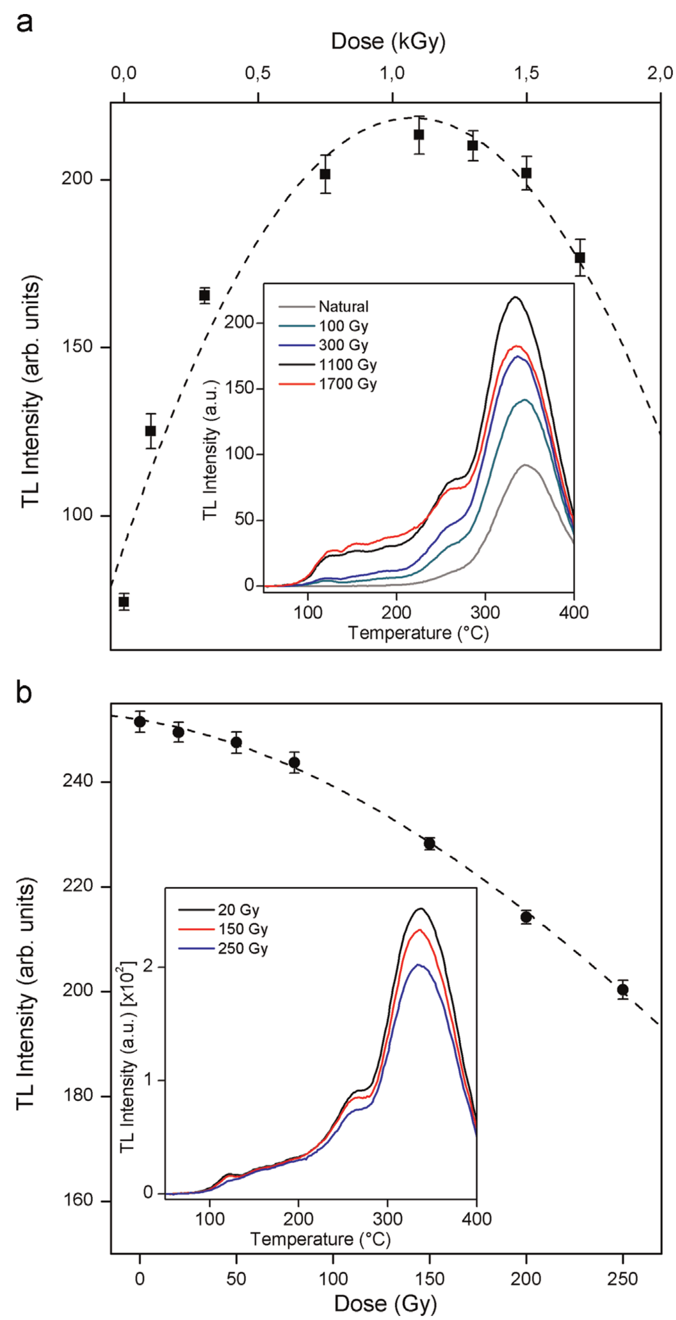

Fig. 2. TL vs. dose curve of a 1100 Gy irradiated green quartz subsequently irradiated with low to 250 Gy γ-rays. Inset: TL response curve of green quartz in the interval 0–2000 Gy.

et al., 1974; Rudra and Fowler, 1987; Toyoda and Ikeya, 1991). When the crystal is irradiated with γ-rays electrons from liberated electron–hole pairs can be captured by oxygen vacancies. One electron captured by oxygen vacancy constitutes an E′$_1$-center, which can be detected by EPR (Toyoda and Schwarcz, 1997).

The radiation dose response of several minerals has shown that the TL intensity or EPR intensity decreases with dose (Watanabe et al., 2015). This decrease is observed after the minerals are subjected to pre-irradiation to high doses in the range of kGy. Therefore, this feature can be used in radiation dosimetry as much as TL is used.

In the present work some silicates minerals samples were studied using TL and EPR techniques to investigate the potential applications in gamma radiation dosimetry using decreasing TL intensity or EPR intensity. It is the first study on silicate crystals for applications in radiation dosimetry using decreasing intensity by TL or EPR techniques.

2. Materials and experimental

Green quartz, three varieties of beryl (uncolored-goshenite, aquamarine-blue/green, morganite-pink) and pink tourmaline have been investigated in this work. We have examined also a non-silicate crystal, namely, LiF:Mg,Cu,P (MCP).

Electron Spin Resonance experiments were carried out using a Bruker EMX EPR spectrometer operating at X-band frequency with 100 kHz modulation frequency. The *g* factor of signals were

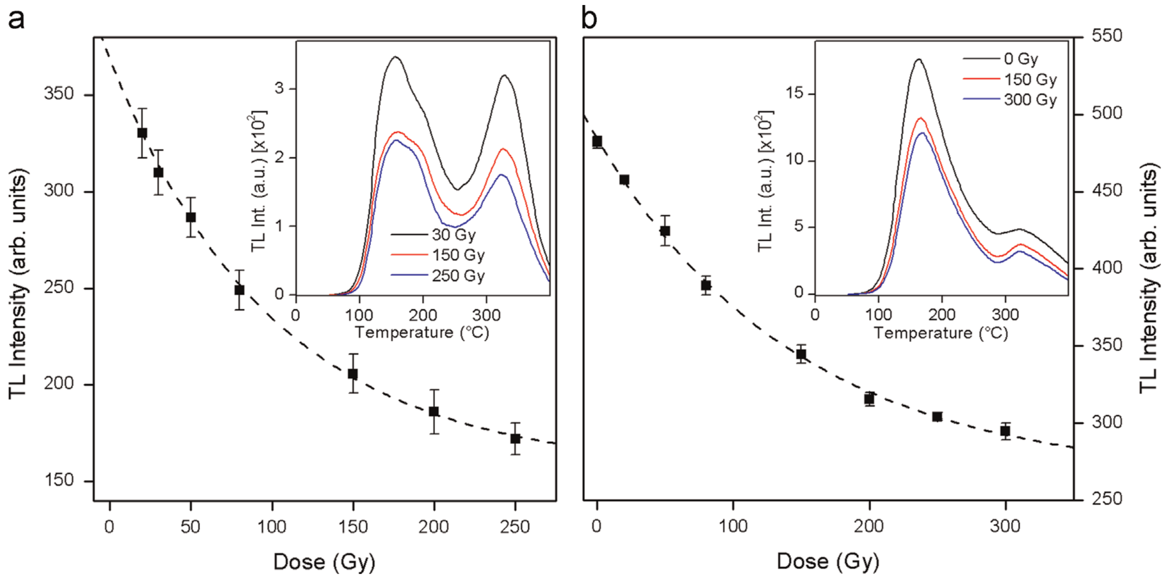

Fig. 3. (a) Aquamarine irradiated with 250 kGy γ-ray and then irradiated with low up to 250 Gy γ-ray. (b) For goshenite previously irradiated with 1100 kGy and then to low up to 300 Gy γ-ray. In inset: TL glow curve of aquamarine and goshenite after pre-irradiated and irradiated with additional gamma dose.

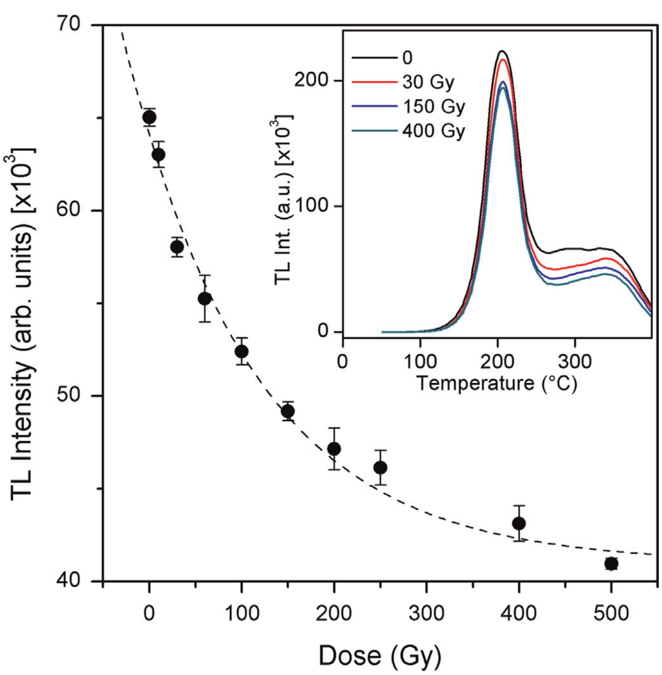

Fig. 4. TL vs. dose curve of pink tourmaline pre-irradiated with 100 kGy and then irradiated with low to 500 Gy γ-ray. Inset: TL glow curve of pink tourmaline after pre-irradiated and irradiated with additional gamma dose.

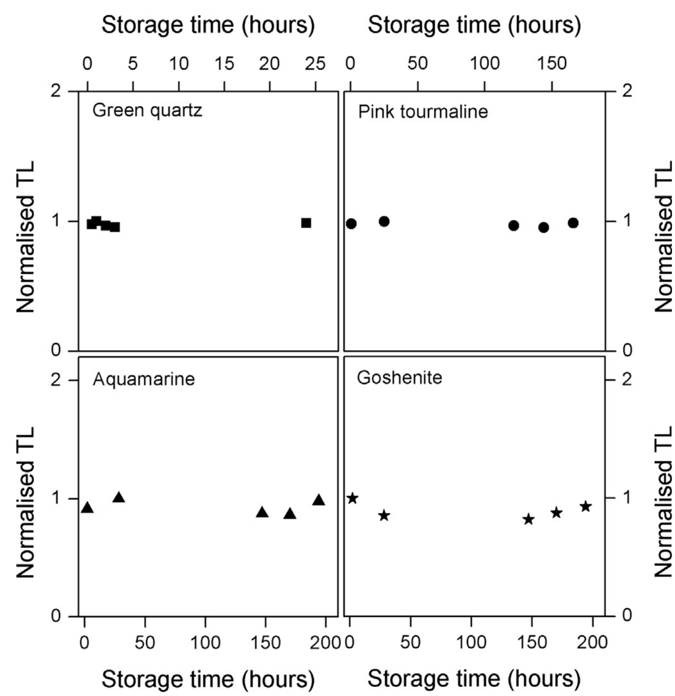

Fig. 5. Reproducibility of the TL peaks after irradiation at room temperature (20–25 °C).

determined using a reference sample of Diphenyl Picryl Hydrazyl (DPPH).

The TL measurements were carried out in a nitrogen atmosphere with a Daybreak model 1100 reader and a model 4500 Harshaw TL reader. The heating rate used in the TL measurements was 4 °C/s. Each point in the glow curve represents an average of five readings.

For irradiation, Institute for Energy and Nuclear Researches Radiation Center's ^{60}Co source was used for γ-doses below 50 kGy. Higher dose irradiation was carried out at CBE-EMBRARAD.

3. Results and discussion

To begin with EPR measurements were carried out on green quartz irradiated with γ-rays with low doses up to 500 Gy. Fig. 1 (a) shows the results of these measurements. As already commented the main signal is that of E_1'-center. Note that the EPR intensity increases, reaching a maximum between 200 and

Table 1
Fading of the samples after gamma dose irradiation, at room temperature (20–25 °C).

Materials	Gamma dose	Glow peak (°C)	Fading
Green quartz	1100 Gy	340	2% in 2 days
Aquamarine	45 kGy	330	10% in 7 days
Goshenite	1100 kGy	320	10% in 7 days
Pink tourmaline	100 kGy	330	5% in 7 days

300 Gy. Of course, this result can be used in radiation dosimetry for doses between low and about 200 Gy. Subsequently green quartz was irradiated initially with 300 Gy dose and then with doses from low to 300 Gy. Fig. 1(b) shows the behavior of the EPR intensity with the dose. This result has shown that 300 Gy irradiated green quartz, which is stable (Ikeya, 1993), can be used for radiation dosimetry in the range 0–500 Gy. The experimental points follow the equation

$$I = I_R + I_0 \exp(-\beta D)$$

I_R is the residual value for large D value and I_R+I_0 is the value at $D=0$. In the present case of the green quartz we obtained $I_R=1005$ (a.u.), $I_0=857$ (a.u.) and $\beta=0.005$/Gy.

We carried out similar measurements on two varieties of beryl and on pink tourmaline, and this time we used, however, TL measurements. We included green quartz again. We started with green quartz and like in the EPR case, we first measured TL of the sample irradiated with doses up to 1600 Gy.

Fig. 2(a) shows the dose–response curve of the 340 °C TL peak of green quartz irradiated with doses between 100 and 1700 Gy. This figure shows that the resulting curve is a parabola with a maximum around 1100 Gy. A sample is irradiated with 1100 Gy dose and then irradiating it with doses up to 250 Gy leads to the curve shown in Fig. 2(b).

Watanabe et al. (2015) have shown that TL intensity in morganite grows linearly up to about few thousand Gy and then saturates. We, therefore, did not carry out the experiment of irradiating the morganite with, say, 1000 Gy. We did, however, such experiment with aquamarine irradiated with 250 kGy and goshenite irradiated with 1250 kGy. Fig. 3(a) shows the result for aquamarine and Fig. 3(b) for goshenite for the TL peaks at 330 and 320 °C, respectively.

Watanabe et al. (2015) studied the behavior of TL intensity vs. dose of TL peak at 190 °C and at 330 °C of pink tourmaline sample. Here, we consider only the second TL peak, due of its stability at room temperature. Irradiating such a sample with 100 kGy dose and subsequently with doses up to 500 Gy we obtain the curve shown in Fig. 4.

To show that these results are reproducible, measurements were repeated not only on the same day, but also after 7 days, same results were obtained in both the measurements (see Fig. 5). The fading characteristics of the samples were analyzed under experimental conditions of storage, the results are shown in Table 1. The shape of the glow curves of the silicates samples did not change but the intensity of glow peak oscillated between 2 and 10% after 7 days of storage at room temperature.

It is usual to use increasing TL or EPR intensity as a function of radiation dose. However, we have shown that green quartz, aquamarine, uncolored beryl (goshenite) and pink tourmaline irradiated with a dose D_m for which these crystals have maximum TL or EPR value, a subsequent irradiation with increasing doses from low to about 400 or 500 Gy, their TL or EPR intensity decreases regularly with the dose. This is a calibration curve and it can be used in radiation dosimetry. The process is reproducible.

4. Conclusion

This work has shown that many TL materials exhibit a TL response as a function of radiation dose that has a maximum at some dose D_m. If such materials are pre-irradiated with the dose D_m and subsequently irradiated with increasing gamma dose starting from a low value and reaching up to 300–500 Gy, a decreasing TL response is observed. In the present study, green quartz, aquamarine, goshenite, pink tourmaline (all are silicate minerals) have been investigated. Such decaying TL as a function of dose can be used in radiation dosimetry. This interesting feature can be used as a calibration curve in radiation dosimetry. Crystals irradiated with gamma dose D_m is stable. The silicate minerals can be efficiently used as dosimeters after applying the suitable pre-irradiation gamma dose D_m.

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