THE INFLUENCE OF HIGH DOSES OF RADIATION IN CITRINE STONES

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Abstract

The possibility of using samples of Brazilian stones as quartz, amethyst, topaz, jasper, etc. for high-dose dosimetry has been studied in recent years at IPEN, using the techniques of optical absorption (OA), thermoluminescent (TL), optically stimulated luminescence (OSL) and resonance paramagnetic electron (EPR). In this work, the TL properties of citrine samples were studied. They were exposed to different doses of gamma radiation (^{60}Co). The natural citrine stone was extracted from a mine in Minas Gerais state, Brazil; it is a tectossilicate ranked as one of three-dimensional structure, showing clear yellow to golden brown color. The natural citrine stone is classified as quartz (SiO_2), and it has a lower symmetry and more compact reticulum. The TL emission curve showed two peaks at 160ºC and 220^{o}C. To remove the TL peak (160^{o}C) of the sintered citrine pellet glow curves, different thermal treatments were tested during several time intervals. The TL dose-response curve between 50 Gy and 100 kGy, the reproducibility of TL response and the lower detection dose were obtained. The results show that citrine may be useful as high-dose detectors.

Keywords: Citrine; TL technique; High-Dose Dosimetry; Gamma Radiation.

1.- INTRODUCTION

High-doses have been used in industrial processes such as material sterilization, food tuber germination treatments, grain and seed growing, water purification, among other possibilities [Mclaughlin *et al*., 1989; Morrissey and Herring 2002].

Samples of commercial, national and imported glasses [Caldas and Quezada 2002; Rodrigues Jr. and Caldas 2002; Caldas and Teixeira 2002] have been tested at the Radiation Metrology group of IPEN, using the techniques of optical absorption (OA) and thermoluminescence (TL) for high-dose dosimetry. Brazilian beach sand samples [Caldas and Teixeira 2004] and from the city of Descalvado [Teixeira *et al*., 2008] were studied in IPEN, and showed favorable use for high-dose dosimetry.

The possibility of using samples from natural mines at Minas Gerais state, Brazil, such as topaz [Souza *et al*., 2002], amethyst [Rocha *et al*., 2003], jasper [Teixeira and Caldas 2012], and jade from other parts of the world [Melo *et al*., 2012], were studied and tested also at IPEN, using the thermoluminescence (TL), electronic paramagnetic resonance (EPR), optical absorption (OA), thermally stimulated exoelectron emission (TSEE) and optically stimulated luminescence (OSL) techniques.

Citrine belongs to the chemical silica group. This material can be found in several countries, as Brazil, Argentine, Melgaxe Republic, USA, Spain, Namibia, Burma, Russia and Scotland. The citrine can be confused with other stones such as amethyst which becomes pale-yellow when heated to 470°C, or smoky quartz when heated to 300-400 °C. All citrine samples have a warm reddish tone, while natural citrine samples are predominantly of yellow color and when heated they present no pleochroism. [Schumann 2009; Branco 2008; Favacho 2001].

The natural citrine stone studied in this work is a tectossilicate with a three-dimensional structure, with a color from clear yellow to golden brown color, and it was extracted from a mine in Minas Gerais state, Brazil. It is classified as quartz (SiO_2), and occurs naturally in clusters of pyramids on a geode base. This kind of material is trigonal, a less symmetrical subset of the hexagonal system with four crystallographic axes.

The objective of this work was to determine citrine samples for application in high-dose dosimetry, using the thermoluminescence technique.

2.- MATERIALS AND METHODS

The citrine stones were extracted from a Brazilian mine, in the Minas Gerais State. The elements of the citrine samples were obtained by the neutron-activation analysis technique at the Radiochemistry Department of IPEN. The results are presented in Table 1. As can be observed, Fe and Na are the most important elements.

Table 1.- Neutron activation analysis results of citrine samples: concentration of the main elements

Element	Concentration ($mg.kg^{-1}$)	Element	Concentration ($mg.kg^{-1}$)
Co	0.030±0.001	Fe	58±2
Cr	0.30±0.02	Sb	0.035±0.005
Na	44±3	Zn	2.1±0.2

The stones were initially washed, broken and powdered in grains with diameter between 0.075 mm and 0.180 mm. Samples of citrine powder were subjected to an initial thermal treatment at 300°C for one hour. This treatment was chosen for reutilization too.

To simplify the sample handling, sintered citrine pellets (50 mg) were prepared at the Laboratory for Production of Dosimetric Materials, IPEN, using Teflon as binder in a ratio of 2 (Teflon): 1 (citrine). The samples were irradiated using a Gamma-Cell-220 system (^{60}Co, dose rate of 1.52 kGyh^{-1}, Out 2013), of the Center for Radiation Technology, IPEN, between doses of 50 Gy and 100 kGy. The irradiations were made at ambient temperature, and the citrine samples were fixed between 3 mm thick polymethyl methacrylate plates (Lucite), to guarantee the occurrence of electronic equilibrium during the irradiations.

The evaluation of the citrine pellets was carried out with a thermoluminescent reader (RISÖ TL/OSL Reader and Controller, model DA-20) using a heating rate of 10°Cs^{-1} and a constant flow of N_2 of 2.5 L/min. All TL measurements were integrated between 50°C and 300°C

3.- RESULTS

In this work some dosimetric properties of citrine samples were studied: TL emission curve, the thermal treatment to eliminate the first TL peak, TL response reproducibility, the minimum detectable dose, and the dose-response curve for gamma radiation.

Figure 1 shows a TL emission curve of a citrine sample irradiated with 10 kGy, one hour after its irradiation. Two TL peaks at 160°C and 220ºC can be observed. The first TL peak at 160°C of citrine pellets was eliminated through the study of thermal treatments at 130°C for different time intervals (5 - 60 minutes), as shown in Figure 2. The most appropriate thermal treatment (TT) to remove this TL peak was 130°C during 10 min. This procedure was undertaken for the following experiments in this work.

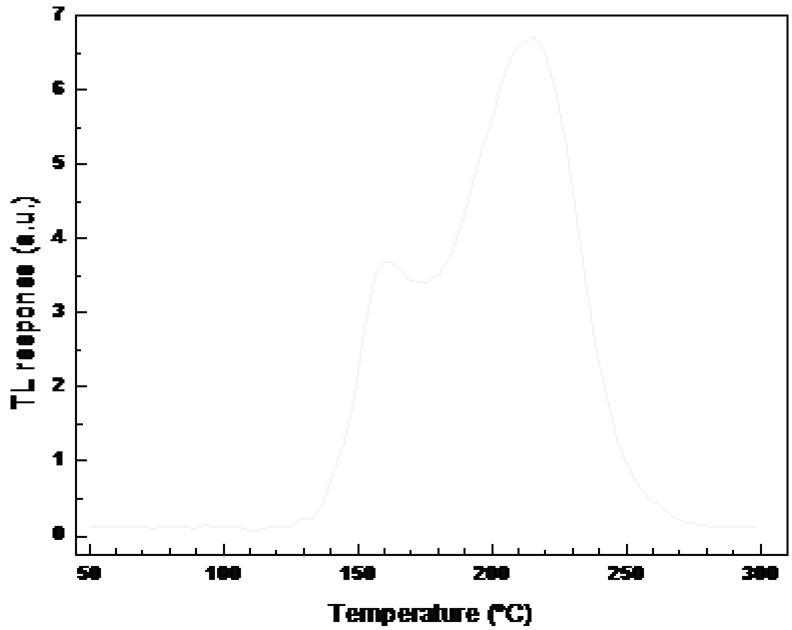

Figure 1.- TL glow curve of a citrine sample pellet irradiated with 10 kGy (^{60}Co).

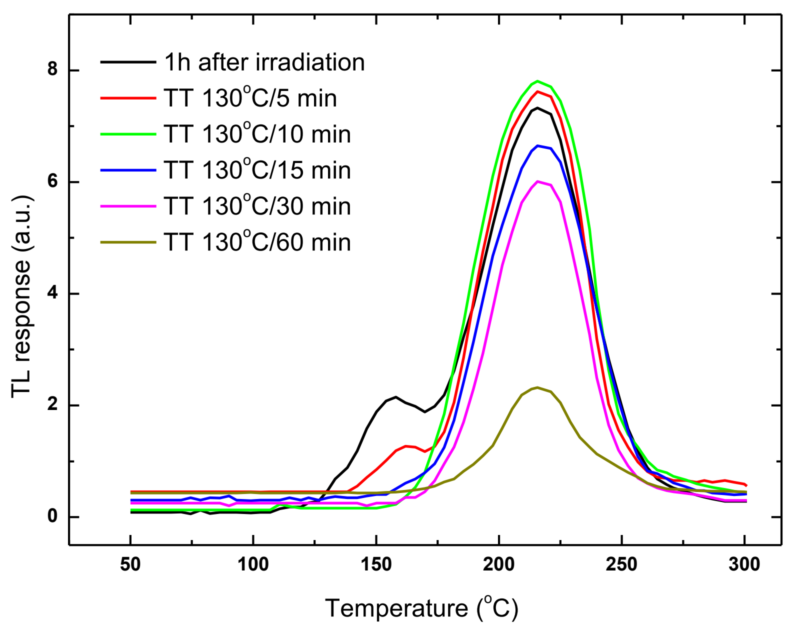

Figure 2.- TL glow curves of citrine pellets irradiated with 5 kGy (^{60}Co) and thermally treated at different time intervals after irradiation.

For the reproducibility study of the TL response, five samples of citrine pellets were submitted five times to the same procedure of thermal treatment at 300°C for 1h (defined for the reutilization), ^{60}Co irradiation (5 kGy), and thermal treatment at 130°C/10 min (to eliminate the first TL peak) and TL reading. The TL reproducibility response obtained was 3.25%.

The dose-response curve was obtained with citrine samples irradiated (^{60}Co) from 50 Gy to 100 kGy and thermally treated at 130°C/10min: Figure 3. Initially a linear behavior can be observed, and after 500 Gy sublinearity occurs. The maximum uncertainty of these measurements was 2.0%.

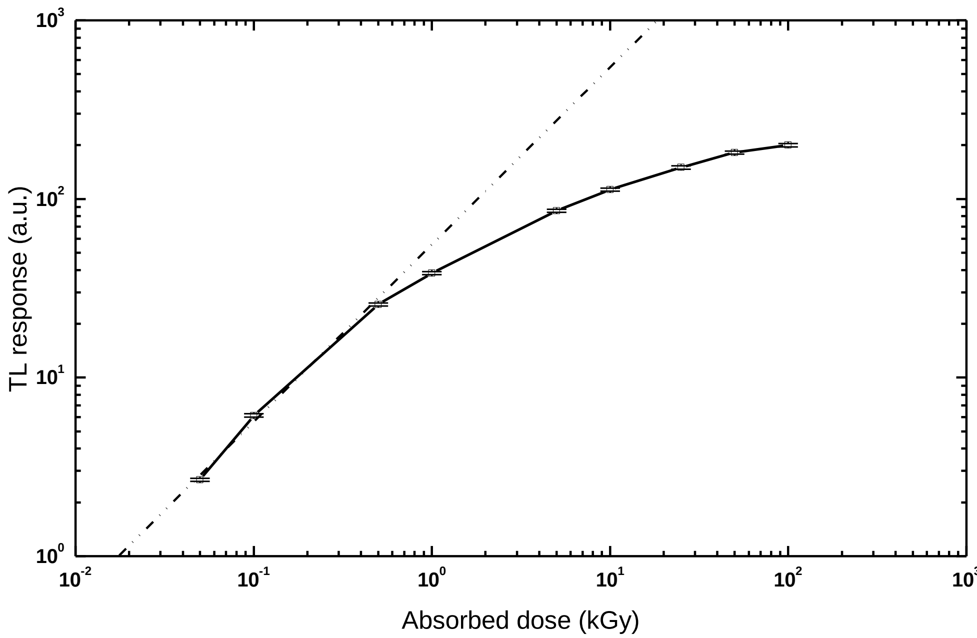

Figure 3.- Dose-response curve of citrine pellets for ^{60}Co radiation. TL measurements were taken after the thermal treatment of 130°C/10 min.

The lower detection limit of the citrine samples was determined by studying the variability of the TL signal obtained from samples treated at 300°C/1h but not irradiated. Taking three times the standard deviation of these measurements, the lower detection limit, was obtained as 150 mGy.

4.- DISCUSSION

TL emission curves of citrine samples were studied and two TL peaks at 160°C and 220ºC could be observed. In order to stabilize the TL response, the citrine samples were submitted to the post-irradiation thermal treatment at 130°C during 10 min to eliminate the 160°C TL peak.

The lower detection limit of 150 mGy is an advantage considering when the citrine samples are used for high-dose dosimetry. For the elimination of the residual signal of the citrine samples, a thermal treatment of 300 °C/1 h was needed for their reuse.

The reproducibility tests presented a value of 3.25% for citrine samples; this can be considered suitable for high-dose dosimetry, because no standardized control parameters were found that can contest these results.

The dose-response curve of the citrine samples showed linearity in a range between 50 Gy and 500 Gy, thereafter sublinearity occurs.

These results show that the citrine samples present possibility of application in high-dose dosimetry.

5.- CONCLUSIONS

The results of this study show that the citrine samples can be used in high-dose dosimetry using the TL technique. The results show that this kind of material could be useful for dosimetry in several applications such as inhibition of sprouting, disinfestation, delayed ripening and insect population control. The TL glow curve of citrine samples presented two peaks at ~ 160ºC and 220º C. To remove the first TL peak, a post-irradiation thermal treatment at 130 ºC during 10 min is enough. The citrine material has the advantage of being found in a great quantity in nature so that it presents low cost.

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REFERENCES

Athem G; Planck M. (2012). *A new dosimeter for diagnostic x-rays*. Applied Radiation and Isotopes **20**: 333-350.

Branco PM. (2008). *Dictionary of Mineralogy and Gemology*. Office of Texts, São Paulo. p.608 (In Portuguese).

Caldas LVE; Quezada VAC. (2002). *Influence of thermal treatments on the response decay of glass radiation detectors.* Radiation Protection Dosimetry **100:**4333-4336.

Caldas LVE; Teixeira MI. (2002). *Commercial glass for high doses using different dosimetric techniques*. Radiation Protection Dosimetry **101:** 149-152.

Caldas LVE; Teixeira MI. (2004). *Sintered sand pellets for high-dose dosimetry*. Nuclear Instruments and Methods in Physics Research Section B **218**: 194-197.

Favacho M. (2001). In: CASTAÑEDA, C. et al. (org.) *Gemstones of the Minas Gerais*. Belo Horizonte, Soc. Brazil. Geologia: 220-233.

McLaughlin WL; Boyd AW; Chadwick KH; McDonald JC; Miller A. (1989). *Dosimetry for Radiation Processing* (London: Taylor & Francis Ltd), ISNB 0-85066-740-2.

Melo AP; Teixeira MI; Caldas LVE. (2008). *TSEE response of silicates of the jade family in gamma radiation beams*. Radiation Measurements, **43**: 397-400.

Morrissey RF; Herring CM. (2002). *Radiation sterilization: past, present and future*. Radiation Physics and Chemistry **63:** 217-221.

Rodrigues Jr. AA; Caldas LVE. (2002). *Commercial plate window glass tested as routine dosimeter at a gamma irradiation facility*. Radiation Physics and Chemistry **63:** 765-767.

Rocha FDG; Oliveira ML; Cecatti SGP; Caldas LVE. (2003). *Properties of sintered amethyst pellets as temoluminescent dosimeters.* Applied Radiation and Isotopes **58:** 85-88.

Souza DN; Lima JF; Valério MEG; Caldas LVE. (2002). *Performance of pellets and composites of natural colourless topaz as radiation dosemeters.* Radiation Protection Dosimetry **100:** 413-416.

Schumann W. (2009). *Gemstones of the World*, Ed. Disal, New York, USA.

Teixeira MI; Ferraz GM; Caldas LVE. (2008). *Descalvado sand for high-dose dosimetrry.* Radiation Measurements, **43**: 1163-1165.

Teixeira MI; Caldas LVE. (2012). *Dosimetric characteristics of jasper samples for high dose dosimetry*. Applied Radiation and Isotopes **70**: 1417-1419.