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Study of thermoluminescence of green quartz pellets for low dose dosimetry

Edy E. Cuevas-Arizaca[a,d,e,*], Máximo Rondón R[d], René R. Rocca[b], M.B. Gomes[c], B. Cortez[c], Carlos D. Gonzales-Lorenzo[a,e], J.H. Takara[a], T. Gundu Rao[a], N.F. Cano[b], J.F.D. Chubaci[a], L.L. Campos[c], S. Watanabe[a]

[a] Instituto de Física, Universidade de São Paulo, Rua do Matão 187, CEP 05508-090, São Paulo, SP, Brazil
[b] Departamento de ciências do Mar, Universidade Federal de São Paulo, Av. Alm. Saldanha da Gama 89, CEP 11030-400, Santos, SP, Brazil
[c] Instituto de Pesquisas Energéticas e Nucleares, IPEN-CNEN/SP, Av. Prof. Lineu Prestes, 2242, CEP 05508-000, São Paulo, SP, Brazil
[d] Universidad Católica de Santa Maria, Arequipa, Peru
[e] Universidad Nacional de San Agustín, Arequipa, Peru

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ABSTRACT

Green quartz is usually studied for low dose TL dosimetry of gamma and X radiation. The aim of the present work is the dosimetric characterization of natural green quartz in the dose range of 0.47 mGy up to 1000 mGy of gamma and X radiation. Green quartz pellets were produced by cold pressing green quartz powder. The pellets are very sensitive to gamma and X radiation with main prominent TL peak at 230 °C. The TL sensitivity of MTS-N (LiF:Mg,Ti) and green quartz pellets were also compared. The dose response curve presented linear behavior in the dose range studied. The glow curve was deconvoluted. Kinetic parameters such as trap depth, kinetic order (b) and frequency factor (s) are determined. The TL photon energy dependence was also evaluated.

1. Introduction

Natural or synthetic silicate minerals had great attention recently in the dosimetry application using thermoluminescence (TL) technique since most of them have excellent TL properties both for low and high radiation doses (Watanabe et al., 2015a, 2015b; Barbosa et al., 2014; Della Monaca et al., 2013; Teixeira and Caldas, 2012; Carvalho-Junior et al., 2012; Woda et al., 2011; Texeira et al., 2010). Examples are green and blue quartz minerals. There are other dosimetry systems such as those based on LiF or Al_2O_3 crystals, which are widely used (Matsushima et al., 2013; Almeida et al., 2018). For low doses only a few systems are available.

The quartz crystal is one of the most known natural materials and extensive researches have been done regarding its TL and OSL properties for applications in the dosimetry, geological and archaeological dating (Cano et al., 2013, 2015). Recently, Farias and Watanabe (2012) investigated the TL properties of various natural quartz, showing that some of them are promising for low-dose dosimetry.

Carvalho-Junior et al. (2012) investigated the dosimetric properties of green quartz pellets manufactured by pressing of a homogeneous mixture of quartz and flocculated PTFE at room temperature.

As far as we know, no work was published on dosimetric properties of pellets produced with green quartz by the sinterization method for low dose radiation detection up to the present. In the present work, we have produced and investigated the behavior of green quartz pellets subjected to low dose radiation. All samples were irradiated with X-ray and gamma radiation and their TL glow curves, dose response, fading and reproducibility were evaluated as well as the energy dependence response. We compared the TL sensitivity and behavior of MTS-N (LiF:Mg,Ti) and Green quartz pellets for gamma radiation.

The dose response of natural materials for dosimetry applications is a very important property. An ideal dosimeter should present linear behavior of the TL response in the dose range of interest. In practice, it is usual to have linear dependence response for low doses, supralinear for intermediate doses and saturation for high doses (McKeever, 1985). Such curve is still useful for dosimetry purpose.

2. Materials and methods

Green quartz minerals were purchased for the present work from LEGEP Minerals Ltd. Brazil. Fig. 1(a) show pictures of fragments of crystal. It was crushed and sieved in fine powder and pressed with a pressure of about 11 ton/cm^2 to obtain pellets. The pellets were sintered at 1200 °C for 60 min. Each pellet has a mass of about 50 mg, 6 mm diameter and 1 mm thickness. These pellets are shown in Fig. 1(b). Crystal grains with size smaller than 0.080 mm were used for

* Corresponding author. Instituto de Física, Universidade de São Paulo, Rua do Matão 187, CEP 05508-090, São Paulo, SP, Brazil.
E-mail addresses: ecuevas@usp.br, arizaca@if.usp.br (E.E. Cuevas-Arizaca), clorenzo@if.usp.br (C.D. Gonzales-Lorenzo), watanabe@if.usp.br (S. Watanabe).

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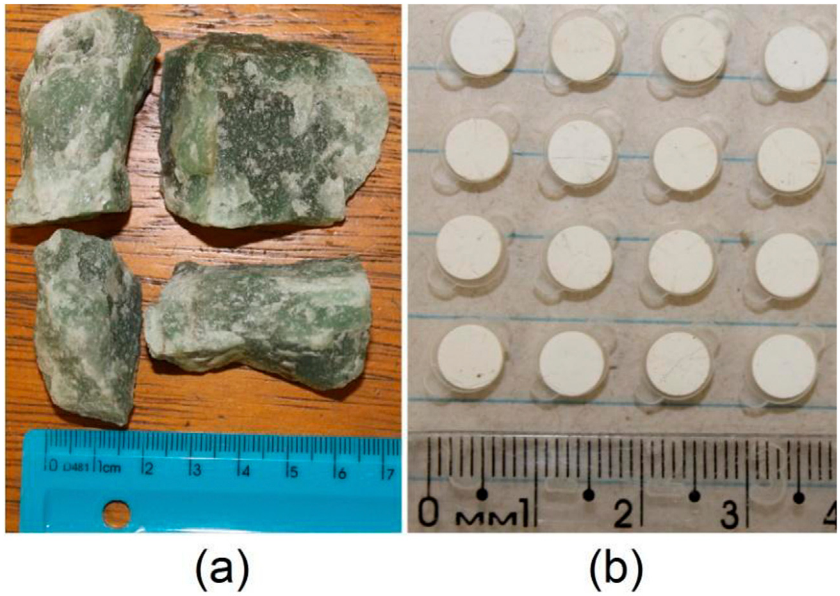

Fig. 1. (a) Green quartz minerals. b) Green quartz pellets. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

X-ray diffraction analysis (XRD).

Dosimeter pellets of 4.5 mm diameter and 0.9 mm thickness of Lithium fluoride doped with magnesium and titanium (LiF:Mg,Ti) that was produced by Thermo Luminescence Dosimetry LAB MIKROLAB with code MTS-N (Mg,Ti,Sintered, natural abundance) were used for comparation of TL sensitivity and response behavior.

X-ray diffraction measurements were performed utilizing a Rigaku Miniflex 300 diffractometer, with Cu K-alpha (λ = 0.15406 nm) radiation. Thermoluminescence glow curves were acquired from room temperature up to 400 °C, at a heating rate of 4 °C/s, with a Harshaw 4500 TL model reader.

The low dose and low energy irradiations were performed using an Industrial X-ray unit (Philips MG 225/450) with source-surface distance of 2.00 m. For high energy irradiations were used a Cs-137 gamma source was used with dose rate of 9.44 μGy/s with source-surface distance of 0.30 m and a Panoramic Co-60 gamma source with dose rate of 7.88 Gy/h with source-surface distance of 0.40 m. During the irradiation the pellets were placed on polymethyl methacrylate (PMMA) holders. The irradiations were performed at room temperature and under electronic equilibrium conditions.

3. Results and discussion

3.1. X-ray diffraction

The powder XRD pattern of green quartz is shown in Fig. 2, indicating that all intense reflections match with the powder diffraction data of quartz which is reported in ICDD Card no: 01-078-2315.

3.2. Thermoluminescence studies

In order to determinate the TL dose response of green quartz pellets for low doses, twenty pellets with TL response between ± 5% were selected from a group of sixty pellets, obtained from different crystal batches. The pellets were irradiated with doses between 0.47 mGy and 20.12 mGy for different X-ray energies, Fig. 3 (a) and (b). For each dose were performed 5 cycles of heat treat, irradiation and TL reading. Each presented result is the average of five measurements and standard deviation is better than ± 5%. We used the mean value of the TL intensity of the peak at 230 °C. The gamma radiation results are very similar to those for X-rays. These results are shown in Fig. 3 (c) and (d).

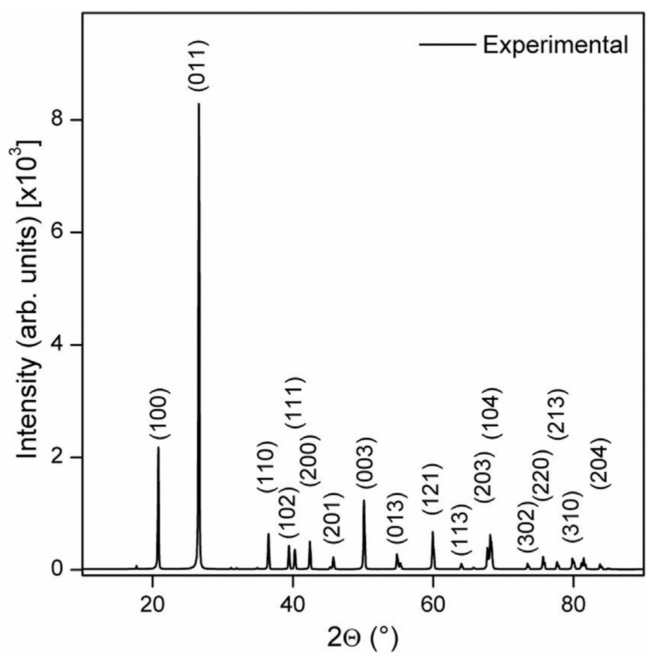

Fig. 2. X-ray diffraction of the Green quartz crystal.

Analyzing the dose response curves with log axes in the same scale, as shown in Fig. 4, it can be observed that the TL response of peak at 230 °C presents a linear behavior in the dose range from 0.47 mGy to 1000 mGy for X and gamma radiation. The linear fit parameters are presented in Fig. 4.

The TL glow curves presenting main TL peaks, around 232 °C for MTS-N (LiF:Mg,Ti) and 230 °C for green quartz pellets, for two different gamma doses are shown in Fig. 5. The peak shape for both materials is similar. The same figura shows the TL response as a function of gamma doce, as can be seem the behavior is lineal for both materials in the studied dose range. These plots were normalized to same mass and TL response for 1 Gy. The TL sensitivity of the MTS-N (LiF:Mg,Ti) dosimeters is 0.228 TL u.a./mg.Gy and the TL sensitivity of Green quartz

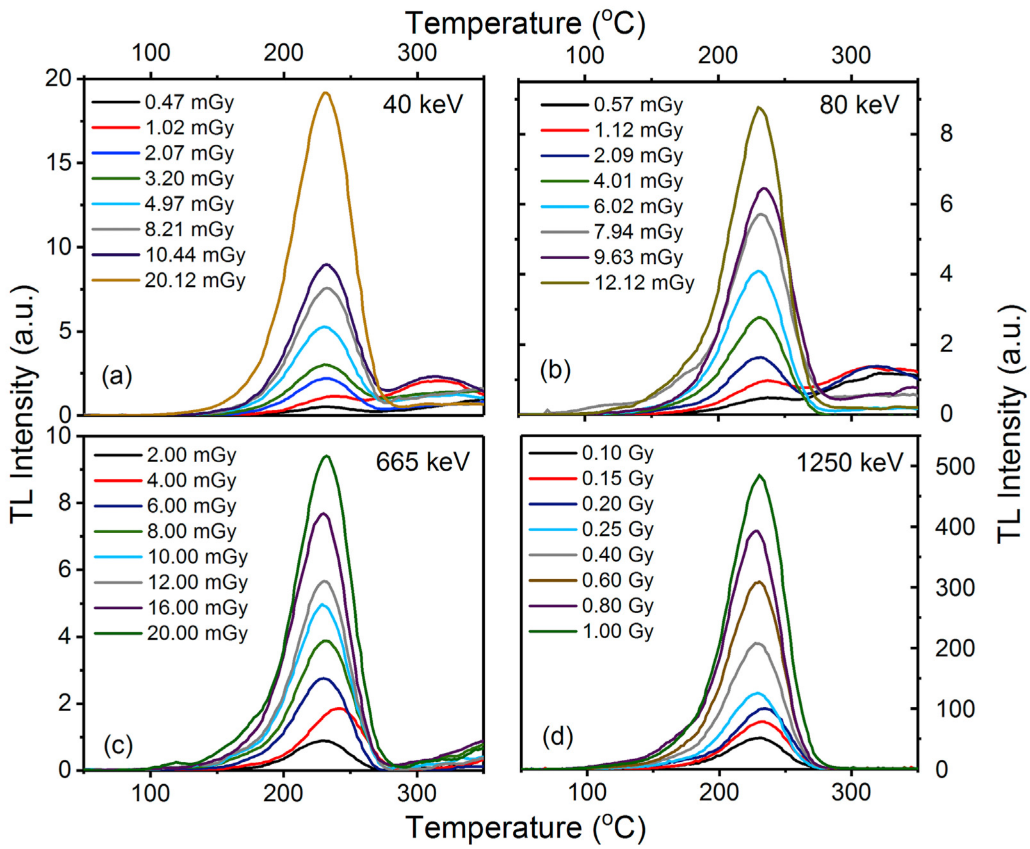

Fig. 3. TL glow curves of green quartz pellets irradiated at several gamma (Cs-137, Co-60) and X radiation doses (40, 80 keV). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

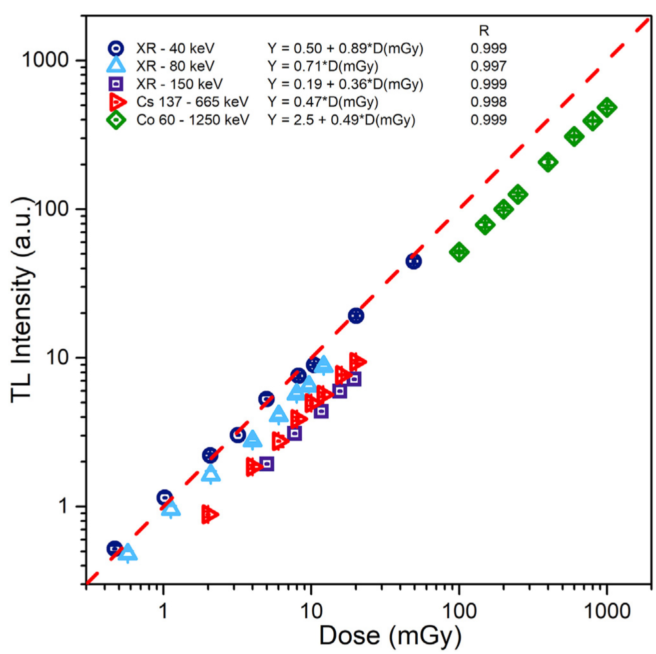

Fig. 4. TL intensity as function of gamma and X radiation dose of green quartz pellets. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

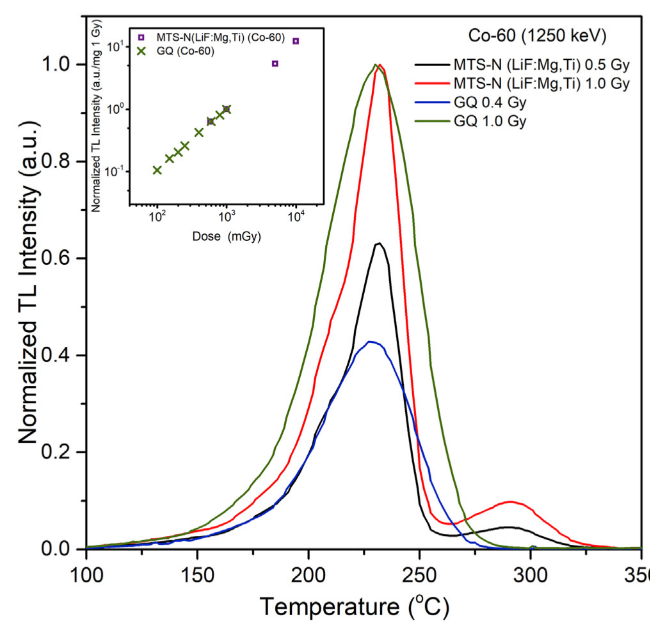

Fig. 5. The glow curve for MTS-N (LiF:Mg,Ti) (0.5 and 1.0 Gy) and Green quartz pellets (0.4 and 1.0 Gy) for gamma radiation. The TL Intensity was normalized with mass and TL response of 1 Gy. The upper inset shows the behavior of both dosimeters with dose. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

pellets is 0.032 TL u.a./mg.Gy. The mínimum detectable dose of MST-N (Li:Mg,Ti) is 70 μGy (Harvey et al., 2014) and of Green quartz is 109 μGy.

Fig. 6 shows the deconvolution of TL glow curve for green quartz

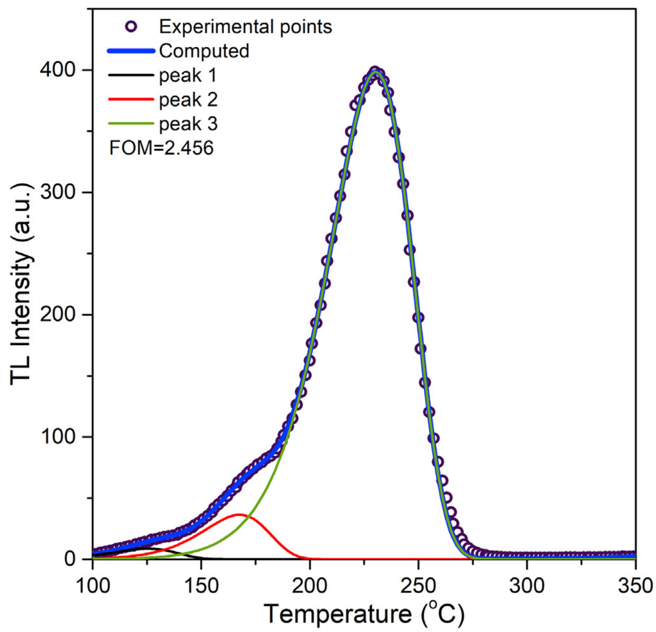

Fig. 6. The glow curve deconvolution of Green quartz pellets, irradiated with gamma radiation 0.8 Gy. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Table 1
Peak temperature (T_m) frequency factor (s) and trap energy (E) using CGCD method.

Kinetic Order	T_m (°C)	E (eV)	s (s^{-1})
1	125.60	0.89	4.61×10^{10}
1	167.60	1.07	4.39×10^{11}
1	230.20	1.10	2.08×10^{10}

pellets. The TL glow curve presents three TL peaks. The main and prominent at about 232 °C and the other two peaks are at 126 and 168 °C. Table 1 presents the TL parameters of the main dosimetric peak.

Another important factor is the storage condition of the dosimeters at room temperature and packaging to avoid exposure to visible light. This procedure is crucial for maintaining the stability of the electrons trapped in the matrix of the crystal. According to model of energy bands for thermoluminescence (McKeever, 1985), it is expected that the electrons trapped in a host matrix should not change depending on storage condition and duration time. This situation is denoted as fading of TL signal. TL signal should be stable at room temperature in a storage time to supply the compatibility between the light emitted resultant and the dose exposed (Toktamis et al., 2007; Topaksu et al., 2013). The fading of the TL peak of 230 °C of the samples exposed to gamma radiation (Co-60) was investigated.

The green quartz pellets were irradiated with dose of 200 mGy and stored protected from light. The TL was read out after 25, 60, 120, 330 and 1320 min. The results are shown in Fig. 7. It can be observed that there is decay of about 10% in the first 330 min, but after that, there is no decay. Therefore, we can say that green quartz pellets have small fading.

The reproducibility of the TL response of the dosimeters can be affected after repeated usage (Claffy et al., 1968); therefore, it is one of the most important factors for dosimetric materials. For analysis of the reproducibility, green quartz pellets were irradiated with dose of 200 mGy (Co-60): the cycle of heat treatment, irradiation, TL reading was repeated five times for the same pellets. Fig. 8 shows the reproducibility of TL peak at 230 °C.

The photon energy dependence for an energy range from 40 to

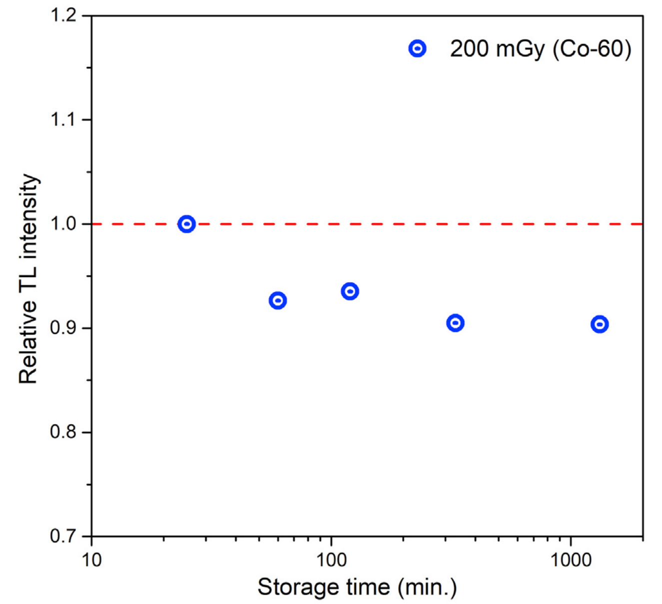

Fig. 7. Green quartz TL response as a function of storage time after irradiation at room temperature, protected from light. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

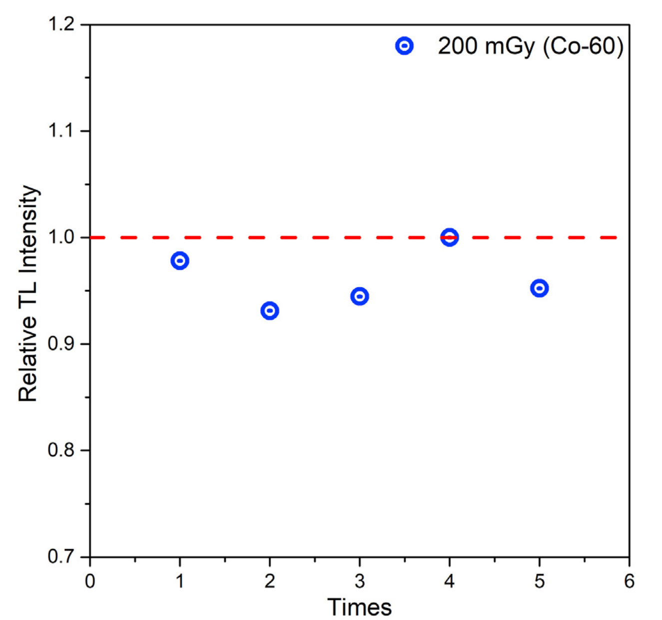

Fig. 8. Green quartz TL response reproducibility: dose 200 mGy of Co-60 gamma radiation. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

1250 keV is presented in Fig. 9. As can be observed, the green quartz pellets presented energy dependence, as expected, in the low energy range, where the predominant interaction is photoelectric.

4. Conclusions

In this work green quartz pellets have been produced for low dose dosimetry. The pellets were exposed to photons in the energy range from 40 to 1250 keV from X and gamma radiation. The glow curve present TL main peak at 230 °C. The dose response is linear in the dose ranging from 0.47 to 1000 mGy.

The sensitivity of Green quartz is less than MTS-N (LiF:Mg,Ti), but

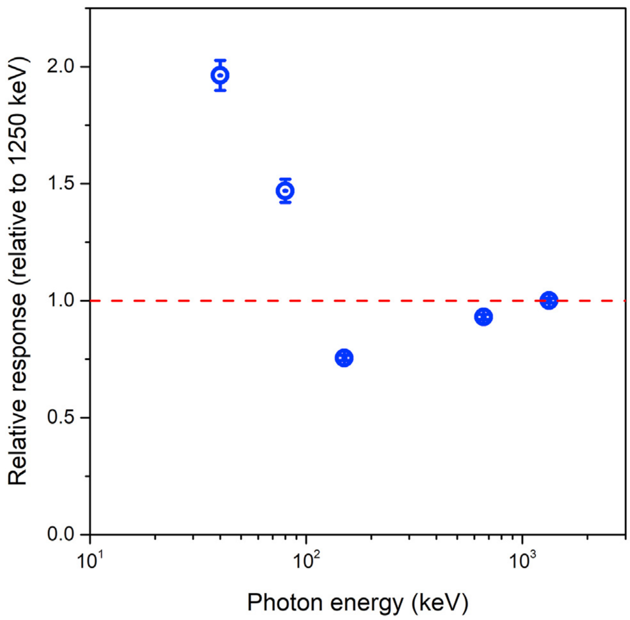

Fig. 9. Green quartz TL energy dependence response. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

both have a linear behavior with gamma radiation dose, from 0.1 up to 1000 mGy. Even with a lower sensitivity Green quartz is a natural mineral, that is cheaper and easy to find.

The deconvolution shows that the glow curve has three peaks with activation energies of, 0.89, 1.07 and 1.10 eV respectively.

The green quartz pellets present about 10% fading and exhibit small energy dependence.

CRediT authorship contribution statement

Edy E. Cuevas-Arizaca: Conceptualization, Methodology, Investigation. **Máximo Rondón R:** Funding acquisition, Methodology. **René R. Rocca:** Software, Data curation. **M.B. Gomes:** Methodology, Formal analysis. **B. Cortez:** Methodology, Formal analysis. **Carlos D. Gonzales-Lorenzo:** Conceptualization, Methodology. **J.H. Takara:** Software, Validation. **T. Gundu Rao:** Writing - original draft, Visualization. **N.F. Cano:** Methodology, Formal analysis. **J.F.D. Chubaci:** Writing - review & editing, Resources. **L.L. Campos:** Supervision, Project administration. **S. Watanabe:** Supervision, Project administration, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.radphyschem.2020.109142.

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