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Applying the TSEE technique to Spectrolite and Opal pellets irradiated with high doses of gamma radiation

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HIGHLIGHTS

- Spectrolite and Opal pellets were studied using ^{60}Co source and the TSEE technique.
- The radiation dose interval of the materials was from 50 Gy to 10 kGy.
- The reproducibility for a dose of 1 kGy was 4.6% for Spectrolite and 4.7% for Opal.
- The dose curves of the TSEE response does not present linear regions.

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ABSTRACT

Spectrolite + Teflon and Opal + Teflon pellets were studied in this work in relation to their dosimetric properties, using the thermally stimulated exoelectron emission (TSEE) phenomenon. The purpose of this work was to study these materials to be used in high-dose dosimetry of ^{60}Co irradiators, which are employed for several industrial applications. The basic physical principle of this technique is the emission of low energy electrons from the surface of different crystals. For this reason, it is very employed in work with specially radiation sources of low penetrating power, as alpha and beta radiation, but also with gamma sources. Both materials had already their dosimetric chatacteristics verified in previous works after exposure to high doses of a ^{60}Co source and measurements by means of thermoluminescence (TL) and optically stimulated luminescence (OSL) techniques. The TSEE response was investigated in terms of the following tests: TSEE emission curves, reproducibility, minimum detectable doses and dose-response curves.

1. Introduction

Thermally stimulated exoelectron emission (TSEE) consists in a technique in which the emission of low energy electrons occurs mainly from the superficial region of the material; thus, it is well employed in studies with radiation sources of low penetrating power (alpha and beta radiation).

Over the years (it is possible to find reports of experimental procedures since the 70s), several researchers have published papers using the TSEE mechanism applied in different materials, as: LiF and BeO (Becker et al., 1970), pure LiF single crystal (Tomita et al., 1976) and α-Al_2O_3 (Molnár et al., 1999).

TSEE was already used for solid state dosimetry, with commercial and natural materials, as gemstones. The use of TSEE is also present in literature for gamma radiation (high penetrating power), in cases as for Topaz, Actinolite, Rhodonite and Tremolite; those results were achieved at the Calibration Laboratory (LCI) at IPEN, Brazil (Souza et al., 2001; Melo et al., 2008; Vila, 2012). The main results of these previous works will be related along of the item Results of this work. In all these cases, the authors show that the studied materials (Topaz, Actinolite, Rhodonite and Tremolite) can be used as radiation dosimeters; these materials are related to Spectrolite and Opal because all of them belong to the Silicate family. Studies with Quartz and Feldspar were already shown by a Danish group in relation to their TSEE response (Ankjærgaard et al., 2006).

The purpose of this work was to investigate the dosimetric characteristics of the Spectrolite + Teflon and Opal + Teflon pellets, and evaluate the possibilities of these samples to be used in high-

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dose dosimetry of gamma irradiators, with use of a ^{60}Co source and industrial applications in a range of absorbed dose from 50 Gy to 10 kGy. In this interval, some examples of applications can be: inhibition of sprouting (as onion, potato, garlic and ginger), reproductive sterilization for insect population control, extending shelf life of fruits (as papaya, banana and strawberry), delaying at the spoilage in foods as meat, chicken and fish; elimination of microorganisms and insects in spices, herbs and other seasonings, water purification, pasteurization of foods and recycling wastes (McLaughlin et al., 1989; IAEA, 2005).

These samples were already analyzed in relation to the TL and OSL luminescent techniques in previous studies (Antonio and Caldas, 2015; Antonio et al., 2016), after exposing them to high doses of gamma radiation beams of a ^{60}Co source. In these works, the two materials were studied in different proportions of powdered stone and Teflon: 1:1 and 2:1 for Spectrolite + Teflon, and 1:1, 1:2 and 2:1 for Opal + Teflon. The results obtained in the first paper revealed, by means of response characterization tests, that both proportions of Spectrolite + Teflon presented a potential use as high-dose dosimeters taking into account the fading effect and evaluating the signal always at the same time after irradiation. In the case of the second paper, the samples of Opal + Teflon in the concentration of 2:1 presented the most adequate results for application as high-dose dosimeters.

In the present work, both kinds of pellets were evaluated by means of the TSEE phenomenon, after irradiation with a ^{60}Co source, through their signal characterization tests. In the case of intermediate doses, the fading was not clearly observed, and this subject needs more investigation. Furthermore, it intends to verify the feasibility of their use as TSEE dosimeters.

2. Materials and methods

The Spectrolite + Teflon (concentration of 1:1) and Opal + Teflon (concentration of 2:1) pellets utilized in this work present 6.0 mm of diameter and 0.8 mm of thickness; their manufacture was already described in previous works (Antonio and Caldas, 2015; Antonio et al., 2016). The powdered samples of Spectrolite and Opal were mixed with Teflon to the samples acquire greater uniformity and mechanical strength (physical stability). Fig. 1 shows the Spectrolite (from Finland) and Opal (from Australia) stones in their natural forms (a and c), and their powder mixed with Teflon (b and d).

The samples were irradiated using a ^{60}Co source, Gamma-Cell 220 System, from the Radiation Technology Center, IPEN, using the irradiator Gamma-Cell 220 System, model 200, Atomic Energy of Canada LTD (47.64 TBq, 10/2014). The experiments were performed using a dose interval of 50 Gy to 10 kGy.

A homemade reader system, projected and developed at the Calibration Laboratory of IPEN, was used to measure the TSEE response, with a gas-flow proportional counter (Rocha et al., 2002). The measurement parameters were: continuous flow of P-10 gas (10% methane + 90% argon) and associated electronics, maximum temperature of heating of 250 °C, and a heating rate of 5 °C/s.

After the irradiation and evaluation of TSEE response steps, all the pellets were submitted to the thermal treatment of 400 °C during 1 h, for their reutilization.

3. Results

The dosimetric properties of both materials were verified using nine pellets of each kind, by means of the following characterization tests: TSEE emission curves, reproducibility (with an absorbed dose of 1 kGy), minimum detectable dose and dose-response curves (varying the dose from 50 Gy to 10 kGy). The results obtained are presented.

3.1. TSEE emission curves

The TSEE emission curves of the Spectrolite + Teflon and Opal + Teflon were obtained after irradiating the samples in an interval of absorbed dose of 50 Gy to 10 kGy (^{60}Co).

The TSEE emission curves obtained for different absorbed doses showed that the Spectrolite samples have only one dosimetric peak about 170 °C, and the Opal, also with only one peak, about 140 °C, according to Fig. 2.

TSEE emission curves were already obtained in other studies with silicate samples exposed to gamma radiation of a ^{60}Co source. Souza et al. (2000) found only one dosimetric peak for crystals of Topaz, at about 190 °C. In another study, Vila (2012) obtained two peaks (1 kGy) for Tremolite at about 185 °C and 240 °C.

Ankjærgaard et al. (2006) performed TSEE measurements of three different materials. For Quartz, they observed an emission signal peak about 170 °C, after exposing it to 4.4 kGy, and for Feldspar, it was possible to observe a peak about 180 °C, when the samples were irradiated with 2 kGy. In the case of NaCl, a first peak was revealed about 75 °C and a second one at 175 °C (absorbed dose of 1.1 Gy).

In relation to the present work, the most similar result to those presented in literature was for the Spectrolite + Teflon pellets, which dosimetric peak obtained was at 170 °C. Thus, this result is in line with those already obtained for other silicates (as Topaz,

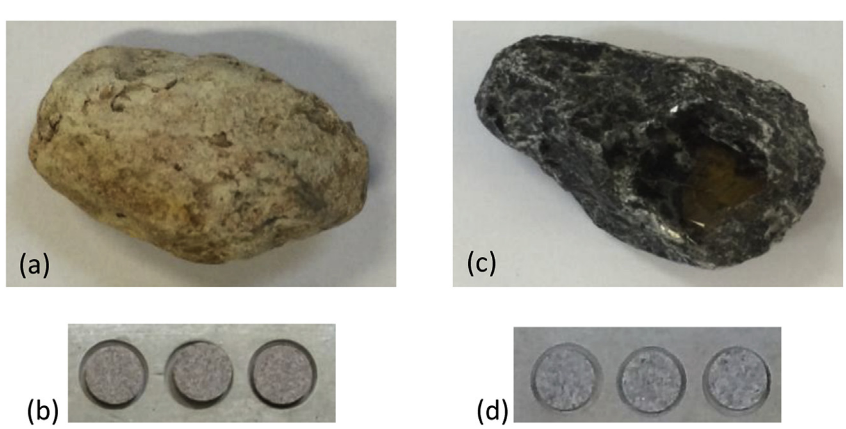

Fig. 1. Stones used in this work: Spectrolite (a) in natural form and (b) in pellets; Opal (c) in natural form and (d) in pellets.

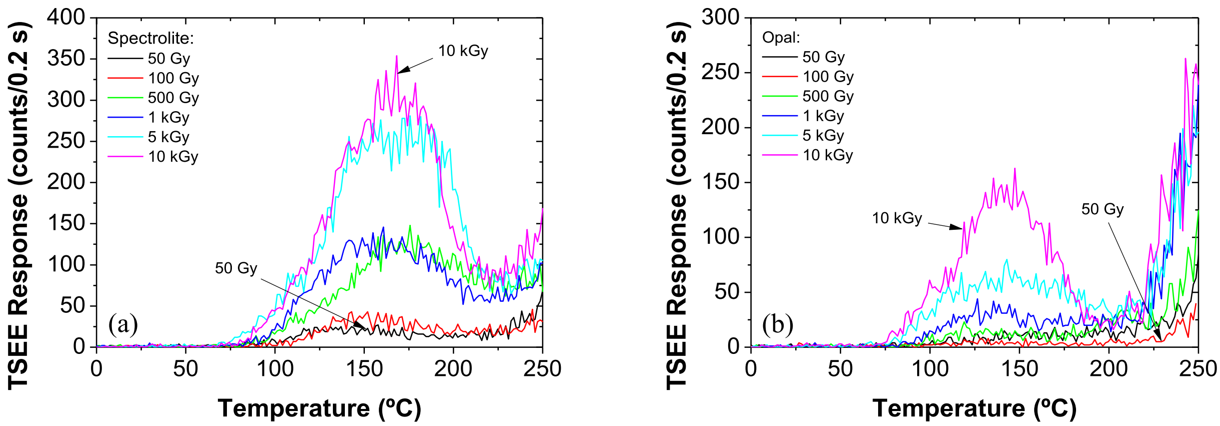

Fig. 2. TSEE emission curves for the: (a) Spectrolite + Teflon and (b) Opal + Teflon pellets (^{60}Co).

Tremolite, Quartz and Feldspar), and also for NaCl (although this material had been irradiated with a low absorbed dose: 1.1 Gy), while the peak for Opal is slightly below in comparison with previous results.

Antonio and Caldas (2015) obtained a TL glow curve for Spectrolite + Teflon (1:1) pellets after irradiation (^{60}Co) with an absorbed dose of 1 kGy, and a dosimetric peak at about 210 °C. Antonio et al. (2016) also obtained a TL emission curve for Opal + Teflon (2:1) after exposure of the pellets to a ^{60}Co source and an absorbed dose of 500 Gy, and observed a dosimetric peak at about 170 °C. Thus, a comparison between the results of this work and those obtained in the previous works is presented (Fig. 3), taking into account the same absorbed doses (1 kGy for Spectrolite and 500 Gy for Opal).

In both cases, the temperatures of the dosimetric peaks obtained in the present work for Spectrolite (170 °C) and Opal (140 °C) were lower using the TSEE technique than for the TL signal, as can be seen in Fig. 3.

3.2. Reproducibility of TSEE response

The series of irradiation, measurement and thermal treatment revealed that the pellets present a stability of response of 4.6% for the Spectrolite + Teflon samples and 4.7% for the Opal + Teflon samples. These results were reached taking into account the average of the group of measurements obtained for each pellet: the six values for Spectrolite + Teflon presented a mean value of 1130.3 counts, and the seven values for Opal + Teflon had a mean value of 1138.2 counts. Then, each measured value was divided by this average value, and so each measurement was normalized. This test was the only one performed using six pellets of Spectrolite and seven of Opal, irradiating the pellets with an absorbed dose of 1 kGy (^{60}Co). At Fig. 4 the response of these normalized measurements in relation to the average value can be seen for Spectrolite and Opal samples. The integrated TSEE signal interval was from 20 °C to 250 °C in this study, and in the dose-response curves too.

Reproducibility limits for several liquid and solid dosimeters were presented at a recommendation of IAEA (2013). These values varied from ±0.5% (alanine dosimeter, highly favorable for high-dose dosimetry) to ±3% (for some dosimeters).

Melo et al. (2008) observed reproducibility of 17.4% and 15.1%, for Actinolite and Rhodonite samples, respectively (for 10 Gy). Vila (2012) irradiated Tremolite samples with an absorbed dose of 1 kGy, and obtained a reproducibility of TSEE response of 8.6%.

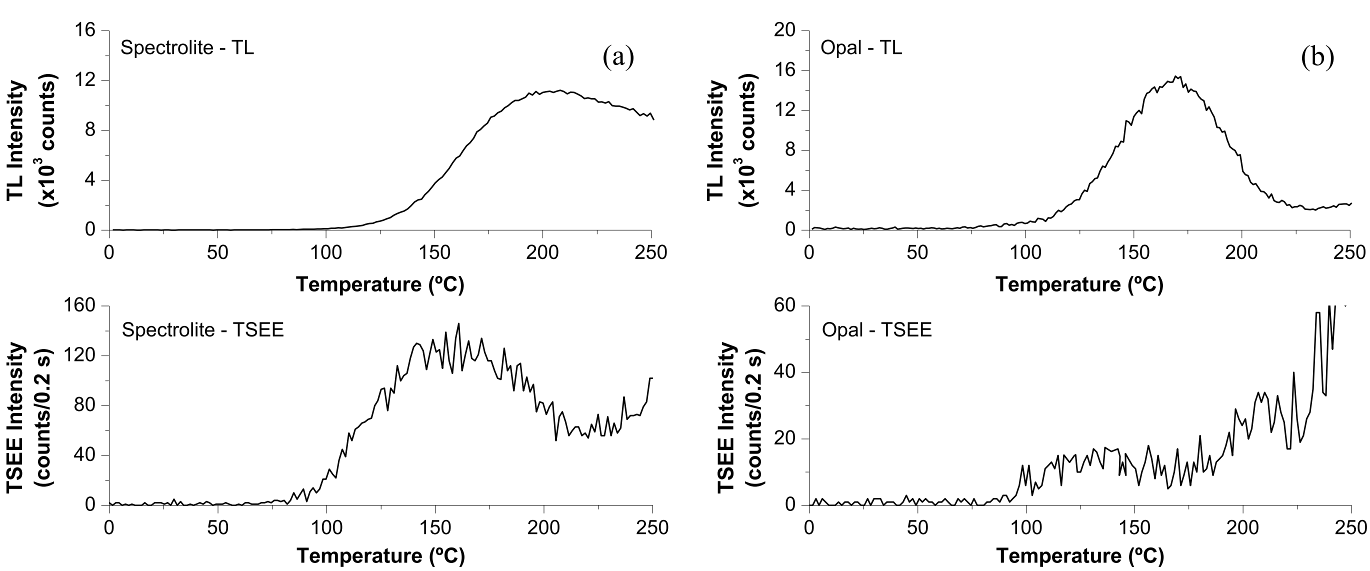

Fig. 3. TSEE emission curves obtained in this work and TL emission curves of previous works (Antonio and Caldas, 2015; Antonio et al., 2016) for: (a) Spectrolite + Teflon and (b) Opal + Teflon pellets (^{60}Co). The heating rates were: 10 °C/s (TL) and 5 °C/s (TSEE).

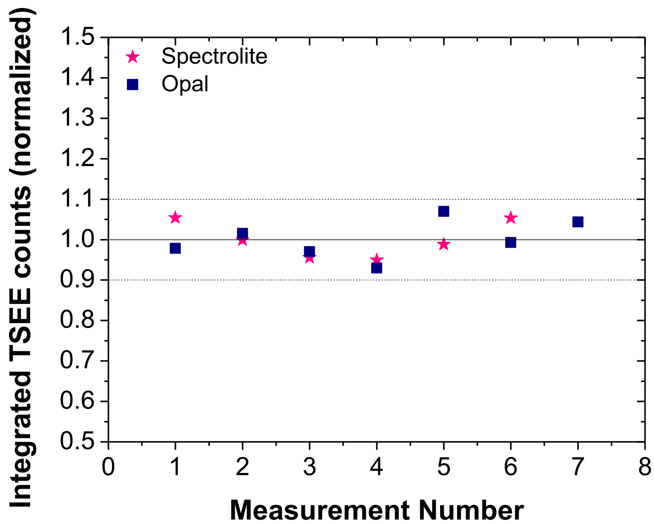

Fig. 4. Reproducibility of the TSEE responses for: (a) Spectrolite + Teflon and (b) Opal + Teflon pellets (1 kGy, ^{60}Co). The dotted lines represent limits of ±10%.

The results obtained for reproducibility of TSEE response for Spectrolite (4.6%) and Opal (4.7%) pellets are higher than those presented by IAEA (2013), but are lower than the results showed for other silicates, as Actinolite, Rhodonite and Tremolite. Taking into account the fact that the dosimeters presented by IAEA (2013) are commercial, it is possible to conclude that the reproducibility values obtained for Spectrolite + Teflon and Opal + Teflon pellets are acceptable for high-dose dosimetry.

3.3. Lower detection limit (LDL)

The lower detection limit (LDL), also called minimum detectable dose, represents the minimum value of absorbed dose that can be detected by a radiation dosimeter. The LDL relates the standard deviation of readings after thermal treatment and the application of a calibration factor obtained for the pellets after irradiation; in this case, the given absorbed dose to the samples in order to obtain this calibration factor was 1 kGy. The LDLs related to the TSEE response for the Spectrolite + Teflon and Opal + Teflon pellets were checked in this work using nine pellets.

The LDL, unlike of the other characterization tests, involves measurements of samples after resetting, and it was obtained according to Pagonis et al. (2006): by the variation of the TSEE signal of the samples after their resetting, and using the method of taking three times the standard deviation of the response of the pellets in this condition (after thermal treatment) (Pagonis et al., 2006). This limit was determined for both kinds of materials, and the results were: 15.4 Gy (Spectrolite) and 17.5 Gy (Opal).

The TSEE signal (used for the determination of LDL) after resetting the samples of Spectrolite + Teflon was 764.7 counts, and 717.1 counts for the Opal + Teflon samples. The uncertainty associated with the values was 113.9 counts for Spectrolite + Teflon pellets and 139.8 counts for Opal + Teflon pellets. The relative standard deviations of these measurements were 14.9% and 19.5% for Spectrolite and Opal, because the measurements of the samples after resetting were very low (and consequently increasing the relative standard deviations).

The limit obtained for Actinolite and Rhodonite by Melo et al. (2008) was 2.0 Gy for both materials. Vila (2012) performed the same study for Tremolite, and the LDL verified was 1.1 Gy.

The LDL obtained for the Spectrolite and Opal pellets was higher than of other silicates.

3.4. Dose-response curves

The TSEE responses for Spectrolite and Opal pellets were investigated varying the absorbed dose in eight steps, from 50 Gy to 10 kGy, a ^{60}Co source. Fig. 5 shows the response behavior observed for both materials.

Each point observed at Fig. 5 represents the mean value of the TSEE measurements obtained for all samples and for each given absorbed dose. The measured TSEE signal for the Spectrolite + Teflon pellets varied from 39.9 × 10^{3} counts to 88.1 × 10^{3} counts, and for Opal + Teflon it varied from 40.5 × 10^{3} counts to 66.4 × 10^{3} counts. The uncertainty of the datapoints is of the average values. The variation of the TSEE response in function of the absorbed dose exhibited an increasing behavior for both materials, with ranges of sublinearity over the whole interval of absorbed dose. Taking into account the TSEE signal of Fig. 2, for all absorbed doses, it is possible that the scattering occurred at the dose-response curves has relation with the signal variation.

Souza et al. (2000) verified that the Topaz presents a sublinear TSEE response in a range from 0.1 Gy to 100 Gy for Topaz. Melo et al. (2008) observed a sublinear behavior in the dose-response curve in an interval of 10 Gy–20 kGy (^{60}Co) for the Actinolite and Rhodonite samples. Vila (2012) found a linear behavior from 50 Gy to 500 Gy for Tremolite.

From the dose-response curves obtained for the Spectrolite and Opal samples, it is possible to observe that the response grows with the dose, although without presenting a linear behavior.

4. Discussion

Both materials analyzed in this work, Spectrolite + Teflon and Opal + Teflon, presented a significant TSEE response in the whole tested absorbed dose interval: from 50 Gy to 10 kGy, in all characterization tests performed.

The dosimetric peaks obtained for both kinds of pellets during the TSEE measurements (170 °C for Spectrolite and 140 °C for Opal) were lower than those demonstrated in previous works for the TL technique, of 210 °C for the first material and 170 °C for second one

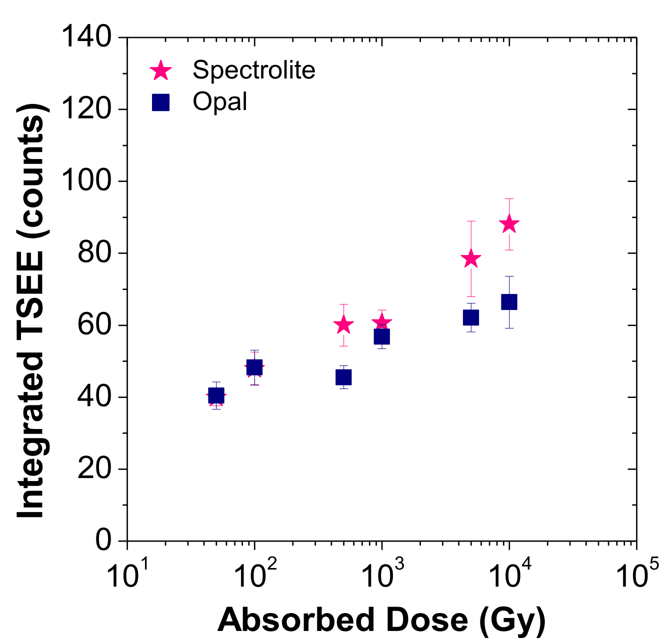

Fig. 5. Dose-response curves for Spectrolite + Teflon and Opal + Teflon pellets using the TSEE technique, after irradiation with a ^{60}Co source.

(Antonio and Caldas, 2015; Antonio et al., 2016).

The reproducibility of the TSEE response of Spectrolite + Teflon and Opal + Teflon pellets was respectively 4.6% and 4.7%. In the case of TL response, it was already determined for Spectrolite, of 4.2% (Antonio and Caldas, 2015), and for Opal, equal to 1.2% (Antonio et al., 2016). For the Spectrolite samples, the TSEE and TL presented similar results, but for the Opal samples they were different.

The lower detection limits (LDL) determined in this work were 15.4 Gy (Spectrolite) and 17.5 Gy (Opal), for the TSEE response. The LDL was determined for Spectrolite using the TL technique as 1.2 Gy (Antonio and Caldas, 2015), with one order of magnitude different of that obtained in this work. In the case of LDL for the TL response for Opal, the value was 19.5 Gy (Antonio et al., 2016), similar to the result in the present work.

The dose-response curves for TSEE signal were acquired in this work with ranges of sublinearity for both materials. For the same materials, but with the TL technique, the dose-response curves were determined for Spectrolite, and the response presented an initial linear behavior with tendency to saturation from 5 Gy to 10 kGy (Antonio and Caldas, 2015); in the case of Opal, the result was a supralinear behavior from 50 Gy to 5 kGy and a tendency to saturation from 5 kGy to 10 kGy (Antonio et al., 2016).

Analyzing the comparisons made between TSEE signal of this work and TL response of previous works, for Spectrolite + Teflon and Opal + Teflon pellets, it is possible to affirm that both techniques can be used for high-dose dosimetry. The TL technique presented more intense and less noisy signal than TSEE, but all the characterization tests demonstrated specific particularities for TSEE or TL response. Thus, it cannot be assured that the TSEE technique is better than the TL technique, but that both techniques may be used.

Throughout this work, comparisons were made between results obtained here and from other materials studied at the same conditions (high dose gamma radiation and natural materials). The study of new materials is always a possibility to replace commercial ones. Due to the high cost of the commercial and known materials, sometimes it is very difficult to acquire them. Then, in this work new possibilities to perform high-dose dosimetry in gamma irradiators using new materials of low cost and of easier access are presented.

5. Conclusions

The main objective of this work was to present two new dosimetric materials and their TSEE results with application in high-dose dosimetry for gamma irradiators.

The characterization tests of the Spectrolite + Teflon and Opal + Teflon pellets were performed using the TSEE technique in order to verify their signal when exposed to high doses of gamma radiation. The results obtained for the Spectrolite and Opal pellets allowed to observe that they present a good reproducibility of response when irradiated with high doses of a ^{60}Co source. The TSEE emission curves showed well defined dosimetric peaks for both materials. The dose-response curves for the Spectrolite and Opal pellets presented an increasing response behavior in function of dose, but no linear region for these materials as radiation dosimeters. The results were favorable and showed that the materials can be used to detect gamma radiation.

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References

Ankjærgaard, C., Murray, A.S., Denby, P.M., Bøtter-Jensen, L., 2006. Measurement of optically and thermally stimulated electron emission from natural materials. Radiat. Meas. 41, 780–786.

Antonio, P.L., Caldas, L.V.E., 2015. Finnish spectrolite as high-dose gamma detector. Radiat. Phys. Chem. 116, 120–124.

Antonio, P.L., Gronchi, C.C., Oliveira, R.A.P., Khoury, H.J., Caldas, L.V.E., 2016. TL and OSL dosimetric properties of Opal Gemstone for gamma radiation dosimetry. Radiat. Meas. 90, 219–223.

Becker, K., Cheka, J.S., Oberhofer, M., 1970. Thermally stimulated exoelectron emission, thermoluminescence and impurities in LiF and BeO. Health Phys. 19 (3), 391–403.

IAEA, International Atomic Energy Agency, 2005. Gamma Irradiators for Radiation Processing. International Atomic Energy Agency, Vienna.

IAEA, International Atomic Energy Agency, 2013. Guidelines for the Development, Validation and Routine Control of Industrial Radiation Processes. IAEA Radiation Technology Series n. 4. International Atomic Energy Agency, Vienna.

McLaughlin, W.L., Boyd, A.W., Chadwick, K.H., McDonald, J.C., Miller, A., 1989. Dosimetry for Radiation Processing. Taylor & Francis, London.

Melo, A.P., Teixeira, M.I., Caldas, L.V.E., 2008. TSEE response of silicates of the jade Family in gamma radiation beams. Radiat. Meas. 43, 397–400.

Molnár, G., Papin, E., Grosseau, P., Guilhot, B., Borossay, J., Benabdesselam, M., Iacconi, P., Lapraz, D., 1999. Thermally stimulated luminescence and exoelectron emission mechanism of the 430 K (D') dosimetric peak of α-Al_2O_3. Radiat. Prot. Dosim. 84 (1–4), 253–256.

Pagonis, V., Kitis, G., Furetta, C., 2006. Numerical and Practical Exercises in Thermoluminescence. Springer, N. Y., pp. 144–149

Rocha, F.D.G., Oliveira, M.L., Cecatti, S.G.P., Caldas, L.V.E., 2002. Dosimetric characterisation of Brazilian natural stones using the thermally stimulated exoelectron emission technique. Radiat. Prot. Dos. 100 (1–4), 417–420.

Souza, D.N., Valerio, M.E.G., Lima, J.F., Caldas, L.V.E., 2000. Dosimetric properties of natural brazilian topaz: a thermally stimulated exoelectronic emission and thermoluminescence study. Nucl. Instr. Meth. B 166–167, 209–214.

Souza, D.N., Valerio, M.E.F., Lima, J.F., Caldas, L.V.E., 2001. The use of pellets of Brazilian natural Topaz as radiation dosimeters. Radiat. Eff. Defect S. 156, 325–330.

Tomita, A., Hirai, N., Tsutsumi, K., 1976. Correlation between thermally stimulated exoelectron emission and thermoluminescence of pure LiF single crystal. Jpn. J. Appl. Phys. 15 (10), 1899–1908.

Vila, G.B., 2012. Characterization of Silicates and Calcium Carbonates Applied to High-dose Dosimetry. PhD Thesis. Nuclear and Energy Research Institute, IPEN-CNEN/SP - São Paulo University, São Paulo (In Portuguese).