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Sand of Colombian beaches as low cost thermoluminescent dosimeter for radiotherapy Title

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This paper describes the thermoluminescent characteristics of the sand coming from Colombian beaches (Coveñas and Moñitos), for its use as dosimeter in therapeutic dose. The selected samples, annealed at 400°C per 1 hour, were irradiated under different doses using a unity of ^{60}Co Theratron 780 C on air at room temperature. The reading was carried out in a Harshaw TLD 4500. The main dosimetric properties of the material (glow curve, response reproducibility, reuse, linearity, and thermal decay) have been in detail studied. The results show a lineal response to the dose from 50 cGy to 1000 cGy, in the material. The studied samples of sand can be used as thermoluminescent dosimeters for applications in different fields. The importance of this work is that the sand is a natural low cost substance, available in large quantities, and can be used in clinical physics in order to make assessments about the received dose by the patient during the medical treatment.

Keywords: Sand; dosimetry; thermoluminescence.

Este trabajo describe las características termoluminiscentes de arena proveniente de playas Colombianas (Coveñas y Moñitos), para su uso como dosímetro en dosis terapéuticas. Las muestras seleccionadas, tratadas térmicamente a 400°C por 1 hora, fueron irradiadas a diferentes dosis usando una unidad de ^{60}Co Theratron 780 C en aire a temperatura ambiente. La lectura se realizó en un Harshaw TLD 4500. Las principales propiedades dosimétricas del material (curva de brillo, reproducibilidad de la respuesta, reutilización, linealidad y decaimiento térmico) han sido estudiadas en detalle. Los resultados muestran que el material tiene una respuesta lineal con la dosis desde 50 cGy hasta 1000 cGy. Las muestras de arena estudiadas se pueden utilizar como dosímetros termoluminiscentes para aplicaciones en diferentes áreas. La importancia de este trabajo radica en que la arena es una sustancia natural disponible en grandes cantidades, bajo costo y puede usarse en física clínica para evaluar la dosis recibida por el paciente durante el tratamiento médico.

Descriptores: Arena; dosimetría; termoluminiscencia.

PACS: 78.55.Qr; 87.53.Bn; 78.60.Kn

1. Introduction

The irradiation processes in sites that use ionizing radiation requires a quality control program [1]. The verification of the absorbed dose is essential part of that program and consists in the comparison between the measured doses with the prescribed doses, either for its medical or industrial use.

Although different dosimetric techniques do exist, the thermoluminescent dosimetry (TLD) is one of the most used in order to develop this kind of studies, and is undoubtedly the main practical application of the thermoluminescence (TL). A great development and a high acceptance degree have been reached in the use of the dosimetry by TL, among the international scientific community since the first works carried out by Daniels, Boyd and Saunders [2].

On this basis, the TL has been now consolidated as a measurement method with high versatility, which is adapted undue hardship to the wide variety of scientific or technical activities, which require a precise determination of the absorbed dose irradiation. As well as the theoretical model is used, the dosimetric systems have been widely recorded in the literature [2-6].

All kind of materials have been proposed and proved, in order to be used in TLD, which wide variety goes since common glasses until sand of different beaches [6-12]. The

sand is a natural material easily available in large quantities. The sand is mainly composed of quartz and the feldspars, which exhibit thermoluminescence. Thank to these properties, some researchers have considered that the sand can be used as radiation TL dosimeter or by Electron Paramagnetic Resonance [11-15].

Vaijapurkar and Bhatnagar [11] described the TL characteristics of the sand from Rajasthan (India), for its use as a Gamma dosimeter available in radiotherapy treatments. In their work, they found a linear response.

After that, Vaijapurkar, Raman and Bhatnagar [12] studied the TL properties of the sand from Rajasthan for its use as high dose dosimeters, and it has been found a significant change in the glow curve's shape for doses above of 6 kGy.

Teixeira, Ferraz and Caldas used the EPR technique to study sand samples from different Brazilian beaches as high dose dosimeters [13]. The dosimetric properties recorded were: response reproducibility, reuse, batch uniformity, detection range, and dose response.

Teixeira and Caldas studied sand from Brazilian beaches to be used as high dose TL dosimeters in routine procedures with ionizing radiation [15]. The results indicate that the material can be repeatedly used for high dose TL dosimetry in various application fields of ionizing radiation.

This paper describes the main TL dosimetric characteristics of sand samples from Colombian beaches (Coveñas and Moñitos), with the object to explore the possibility to use this natural material as TL dosimeters in radiotherapy.

2. Materials and Methods

In order to remove the organic impurities, the sand samples with hydrochloric acid 1 N were washed. For it, for each 5 g of sand 3 ml HCl per 45 minutes were added to each sample at room temperature. The sand was removed every five minutes to make possible the interaction between the grain's superficial area and acid. The previous procedure was repeated three times. After that, the acid was retired by washing it with distilled water and finally the samples were dried at 75°C during 1 hour with the object to remove moisture (humidity) from them.

To determine the presence of elements, compounds and minerals in the samples, an elemental chemical analysis and a structural analysis were carried out. The elemental chemical analysis was made by X-ray fluorescence (XRF) in the Management and Technology Laboratory of the factory Cerro Matoso S.A., using Philips FRX 2400 equipment, at room temperature; 5 g of the sample were used and the results are the arithmetical average from several measures. The structural analysis was carried out by X-ray diffraction (XRD). At room temperature the spectrums were obtained in a diffractometer Philips PW 1710, using the copper line $K\alpha_1$ ($\lambda=1.54046\text{\AA}$) as incident radiation, under 40 kV of voltage in the tube and a stream of 30 mA, varying the sweep angle among $10^\circ < 2\theta < 70^\circ$ every 0.02 degrees/s.

Before the irradiation process, the magnetic particles in the samples were removed by appropriate magnets (~ 5000 gauss), to minimize the TL signals absorption, as suggested by Vijapurkar and Bhatnagar [11].

Then, it was applied a thermal treatment at 400°C during 1 hour, continued by a cooling phase on air until the room temperature was reached. In order to carry out these treatments a furnace Lindberg/Berg UP150® was used. This treatment object is to reduce the natural sign of the material before each irradiation and let be its reutilization as it's suggested by Teixeira, Ferraz and Caldas [13].

The irradiation were made on air at room temperature ($\sim 32^\circ\text{C}$) using an unity of ^{60}Co Theratron 780 C at 80 cm of distance from the source, inside of a radiation area of $10\times 10\text{ cm}^2$ in the irradiation plane. The samples of sand dust, with approximately 100 micras of grain size and each one with 30 mg of mass, were sited between two acrylic plates of 5 mm density, in order to reach electronic equilibrium conditions.

The reading was carried out in a TLD 4500 made by Bicron®, using a heating planchet from the equipment. During the process a preheating temperature of 50°C was used, which was obtained from de data acquisition, with a rate of 10°C/s until reach a maximum temperature of 330°C, continued by a heating at 330°C. To eliminate the infrared contri-

TABLE I. Chemical composition of sand samples (Wt %).

Sample	Compounds		
	SiO ₂	Al ₂ O ₃	CaO
Coveñas	83.49	4.40	3.56
Moñitos	87.29	4.60	0.85

bution due to the heating, and to reduce the humidity effects, all of the readings were carried out in a high purity N₂ atmosphere during the process.

3. Results

3.1. Elemental chemical analysis

The results for the FRX analysis are on Table I, where only the major presence compounds have been recorded. Let us notice that the samples show high SiO₂ concentrations, quartz major component and the feldspars, minerals that exhibit TL. Also low Ni, Fe, Cr₂O₃, MnO, Co₃O₄, P₂O₅ and other compounds (not shown in the Table) concentration were found.

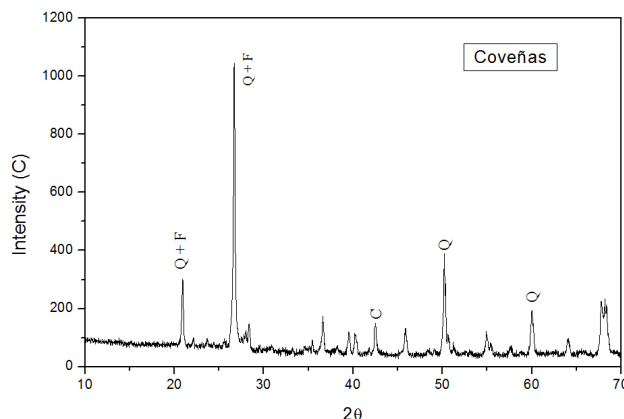


FIGURE 1. X-ray diffractogram for the Beach of Coveñas, in which the presence of Quartz (SiO₂) and the feldspar stand out, these, are minerals that exhibit thermoluminescence.

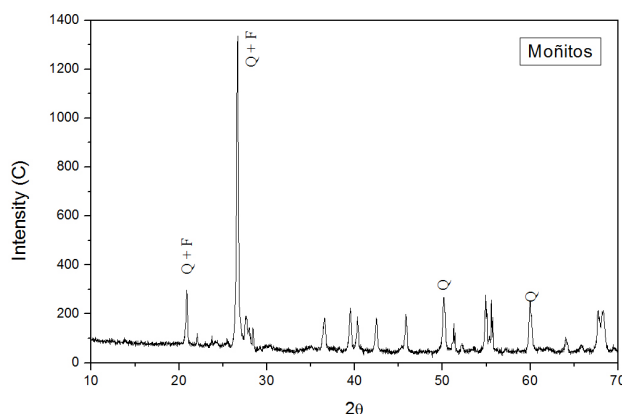


FIGURE 2. X-ray diffractogram for the Beach of Moñitos, in which the presence of Quartz (SiO₂) and the feldspar stand out.

3.2. Structural analysis

The diffraction patterns for the sand samples from Coveñas and Moñitos are respectively observed on Figs. 1 and 2. Both diffractograms clearly show a set of peaks or signals corresponding to a mineral mixture among the Quartz (Q) as its main compound and the feldspar (F), such set of peaks seems to be the second group in presence in these samples. Also other minerals such as the calcite (C), the enstatite, diopside, and augite were found, but these are not relevant for our work.

3.3. Natural response of the material

The emission spectrums of the sand samples from Coveñas and Moñitos are on Fig. 3 respectively, before the thermal treatment, after this thermal treatment (without irradiation), and its response after receive a 50 cGy dose. Even though the thermal treatments don't reach the total elimination of the background signal, its effect can be depreciated when it's compared with the response of the material after irradiation. It's also observed that the sample from Moñitos beach exhibit a minor response to the intensity of the TL signal, which is demonstrated during all the experiment.

3.4. Glow curve

The glow curves for the sand from Coveñas and Moñitos have been illustrated on Figs. 4 and 5 respectively, which were irradiated to different doses. Note that the signal intensity increases proportionally with dose and that the glow curves exhibit a single peak, which is located approximately at 145°C for Coveñas sand and 148°C for the sample of Moñitos, regardless of the dose

3.5. Reproducibility

In order to study the TL response reproducibility, a batch of samples from each beach was subjected to a single irradiation of 100 cGy and randomly divided into groups of five elements for analysis. The results showed an associated uncertainty with the mean value of the measurements of 4.8% for the Coveñas sample and 1.6% for the Moñitos sample. This result is acceptable if one considers that commercial dosimeters TLD-100 have an uncertainty of 1.1% under the same environmental conditions [17].

3.6. Response with the dose

To determine the response of the material with the doses, the samples were irradiated until 10 Gy. The results are seen in Figure 6, where the points represent the mean values of each set of measurements obtained after each irradiation. The error bars correspond to a typical deviation on the mean value of each group of measurements. The results show a linear behavior in the used dose range.

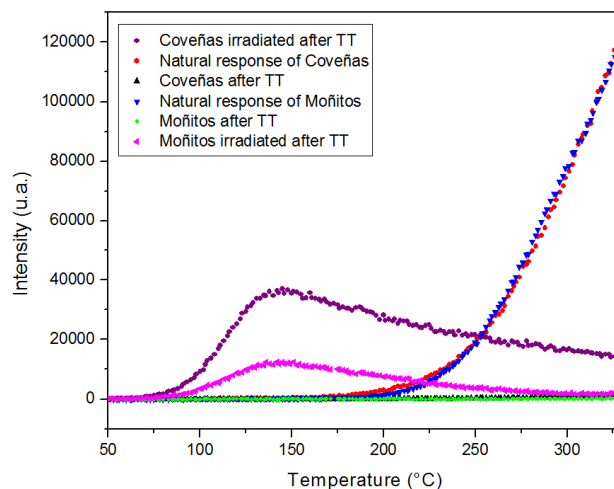


FIGURE 3. TL spectrums of natural sand from Coveñas and Moñitos without thermal treatment (TT), with a 400°C/1 hour TT, and after receive a 50 cGy irradiation.

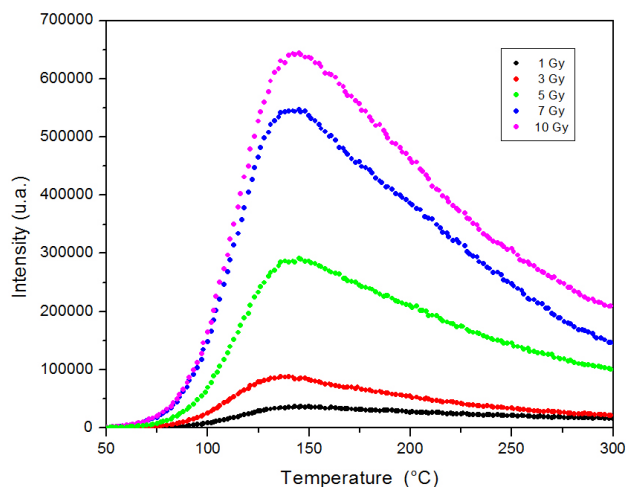


FIGURE 4. Thermoluminescence spectrums for a sample from Coveñas beach, irradiated at different doses.

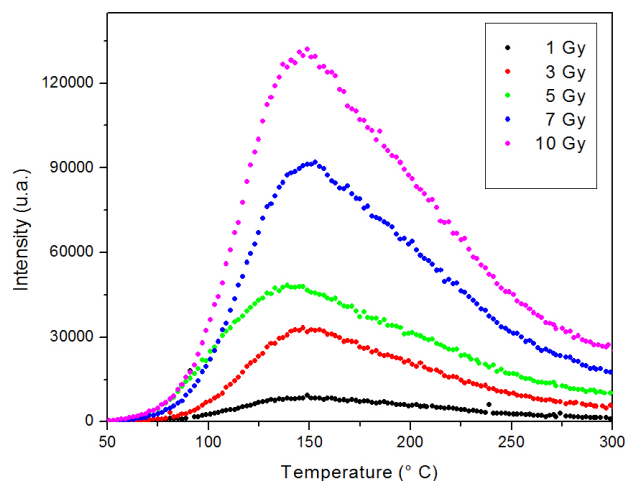


FIGURE 5. Thermoluminescence spectrums for a sample from Moñitos beach, irradiated at different doses.

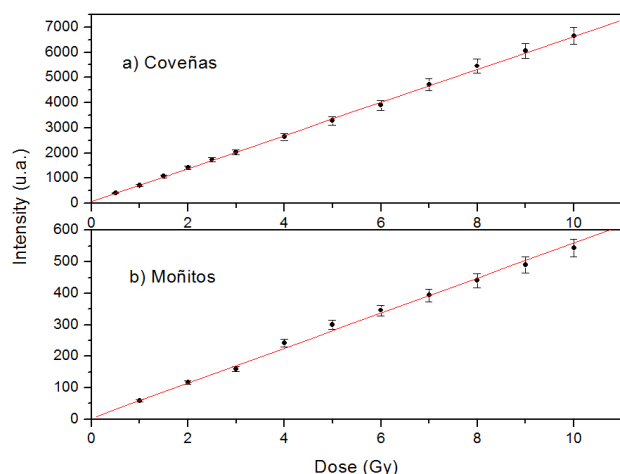


FIGURE 6. Linear response of the material with the dose until 10 Gy.

3.7. Signal decay

A group of sand samples was subjected to thermal treatment at 400°C for 1 h and then exposed to gamma radiation at a dose of 50 cGy to study the thermal weakening of the signal. The measurements were made during the next 30 days after irradiation, at which time the samples were stored at ambient conditions (high relative humidity and temperatures around 32°C). After 5 days, the Coveñas sand showed a signal decay of 36% approximately, tending to a constant value during the next 30 days. Meanwhile, the Moñitos sand only reached this decay after fifteen days of being irradiated.

3.8. Reuse

To reuse the sand samples, they were submitted to different thermal treatments with the object of eliminate the signal originated by the ionizing radiation. Although the signal could not be totally eliminated, it was observed a remarkable decrease in the intensity of the spectrum with the temperature increase and the heating time. Finally, it was chosen a 400°C/1 hour thermal treatment, given that successive thermal treatments can be harmful to the material reproducibility and sensitivity, especially when they are carried out under high temperature and by long time.

3.9. Detection limit

The minimum dose detected by the sand samples was 0.02 Gy, after being subjected to gamma radiation. It should be clarified that this is the minimum dose delivered by the Theratron 780 C which was verified using commercial dosimeters TLD-100 and used the calibration standards reported in the literature [16-17]. In all cases the samples showed a glow curve similar to that observed with the other doses.

4. Discussion

In the materials thermoluminescent the TL emissions are essentially processes related to the presence of impurities (dopants) that have been naturally or artificially introduced into the network of the host matrix [16]. These impurities generate specific energy levels or traps in the forbidden energy band situated between the valence and conduction bands, which don't exist in the pure material. These new levels can be occupied by charge carriers, electrons and/or holes, released during irradiation.

By comparing the glow curves of the sand from Moñitos and Coveñas, it appears that the first exhibits lower signal intensity, although they be irradiated at the same dose. This lower intensity is associated with a lower concentration of defects or traps (N) in the material, leading to a lower concentration of electrons (n) in traps and holes in recombination centers. Furthermore, by observing the amplitude of the TL curves of these sands can be assumed that they consist of a superposition of two or more TL signals. This response is consistent with the results obtained in the chemical and structural analysis where there was found a "blend of minerals" that could have different crystallographic phases with different "kinds" of defects. Hence, it is necessary to make use of computational "deconvolution" techniques in order to separate the TL independent signals and to determine "kinetic parameters" such as the order (b), activation energy (E), pre-exponential factor (S), and others, making use of different existent theories [5].

The glow curves differ enough of the results reported by Vaijapurkar and Bhatnagar [11], whom described the TL characteristics of the sand from Rajasthan (India), which exhibited two well defined TL peaks on its glow curve at 80°C and 220°C. On their way, Teixeira and Caldas [15] studied sand samples from Brazilian beaches in order to be used as high dose TL dosimeters and they found that TL glow curves showed two peaks approximately at 110°C and 170°C. In both works, the first peak was not considerate for its use in dosimetry due to the fast decay at room temperature, on the other hand the second TL peak showed an adequate response as a function of the absorbed dose.

The study of the dosimetric characteristics (reproducibility, dose response, and thermal decay) of the Colombian sands showed a linear behavior from 0.5 to 10 Gy, as had been reported by Vaijapurkar and Bhatnagar for a similar dose range [11].

The samples presented a higher decay at 30% during the first fifteen days, after being irradiated; other researchers' results on similar materials don't show a significant decay during the first thirty days [11]. The strong thermal weakening presented by the sands from Coveñas and Moñitos can be related to the storage conditions (relative humidity ~70% and temperature average $32 \pm 4^\circ\text{C}$), since similar results [17] were observed in commercial TLD-100 dosimeters, for which, fading in the TL response varies according to the literature, from 5 to 10 % per year [6].

In order to decrease this strong weakening, the sands were subjected to thermal treatment at 100°C by ten minutes, after this time the samples exhibited a glow curve similar to the first one but with a remarkable decrease on its intensity and a shift in the main peak of about 45°C. This behavior is easily explained using a phenomenological model, whereby a high number of traps near the conduction band are the first to be vacated, which generate the responsible photons for the low temperature spectrum part, deducing that only the off electrons in major energetic depth traps “survive” after the thermal treatment. Given this case, the samples presented a hardly decay in the response of 10% during the first 48 hours, tending to a constant value during the next thirty days.

5. Conclusions

The obtained results in the structural and chemical analysis show that beaches from Coveñas and Moñitos presented high quartz and feldspar concentrations, minerals which exhibit TL. The glow curves of the used sands (Coveñas and

Moñitos) exhibit a wide peak which intensity proportionally increases with the dose, showing a linear response in the used range. Besides, although the curve shape does not vary, its proportion and position depend on other factors from used reading cycle and applied thermal treatments before and/or after the irradiation.

Given that measures uncertainty showed acceptable values and the signal decay can be minimized using adequate post-irradiation thermal treatments, the results let us conclude that the studied sand samples can be used as TL dosimeters in the therapeutic doses range. The advantage from this kind of material is that the sand is a natural substance available in large quantities on these beaches.

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