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Sand for high-dose dosimetry using the EPR technique

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Abstract

The electronic paramagnetic resonance (EPR) technique was utilized to study sand samples from different Brazilian beaches for high-dose dosimetry. Sand also contains concentrations of heavy minerals. Sand samples were studied in relation to their main dosimetric properties: response reproducibility, reutilization, batch uniformity, detection range and dose response. The EPR signal grows significantly as a function of absorbed dose for $g = 1.999$. All studied sand samples can be used as EPR dosimeters for different applications in medical, agricultural and industrial areas.

Keywords: Sand; Dosimeter; EPR

1. Introduction

Advances in irradiation technology have made commercial processes such as sterilization, pasteurization, food preservation, and treatment of various materials possible (McLaughlin et al., 1989; Farrar, 1999).

Radiation processing at irradiation facilities requires a quality control program. The verification of absorbed doses is an essential part of such a control. Several kinds of dosimeters have been proposed, tested, and are presently in use for this purpose (McLaughlin et al., 1989). Dosimetry material easily found in nature, such as sand, or commercial products, such as glass, were proposed by some researchers (Vaijapurkar and Bhatnagar, 1993; Vaijapurkar et al., 1998; Quezada and Caldas, 1999; Caldas and Teixeira, 2002; Rodrigues and Caldas, 2002).

The main component of sand is quartz (SiO_2), and its electronic paramagnetic resonance (EPR) properties have received great attention in the literature; Griscom (1990) published a review article of this important insulating material. Marfunin (1979) and Ikeya (1993) identified the main paramagnetic defects in quartz.

In this work, EPR properties of sand from different Brazilian beaches were investigated in order to propose an efficient and low-cost dosimeter.

2. Materials and methods

Sand samples used in the present work were obtained from different Brazilian beaches, over 500 km from each other: Barra do Sahy Beach, São Paulo; Santinho Beach, Santa Catarina; and Ponta Negra Beach, Rio Grande do Norte, Brazil. Sand was crushed to grain sizes between 0.037 and 0.074 mm. The sand samples were washed with 1 N (1 M) HCL; after that, distilled water was used

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to remove the HCl and organic impurities (Vaijapurkar and Bhatnagar, 1993). The wet sand samples were dried in an electric oven, Formitex, at 75 °C for 1 h.

The magnetic particles were removed using a magnetic separator from S.G. Frantz, model Isodynamic, L-1, in order to minimize the absorption of TL signals, as suggested by Vaijapurkar and Bhatnagar (1993). Neutron activation analysis of the sand samples, performed by the Radiochemistry Department of IPEN, is shown in Table 1.

Irradiations were performed in air (room temperature) using a Gamma-Cell 220 system (^{60}Co, dose rate of 5.17 kGy/h) under electronic equilibrium conditions achieved by covering the samples with 6-mm-thick Lucite plates.

Thermal treatments (500 °C/1 h) were applied to the sand samples to see whether the material can be reutilized. EPR measurements were carried out using a Bruker EMX spectrometer with a rectangular cavity (ER4102 ST), at room temperature, at microwave frequency of 9.76 GHz (band X), microwave power of 0.202 mW, and frequency and field modulation amplitude of 100 kHz and 0.1 mT, respectively.

The sand samples, average mass of (150 ± 1) mg, were placed in a pure quartz tube and inserted in the cavity. Due to the thermal fading of the sand sample EPR spectra, all measurements in this work were taken exactly 1 h after irradiations.

3. Results and discussions

The EPR spectra obtained of the sand samples from the three different origins, natural and demagnetized, exposed to an absorbed dose of 10 kGy are shown in Figs. 1 and 2, respectively. These spectra present signals with a *g*-factor at 1.999 and 1.995. These signals are formed in quartz grains present mainly in sand: 97%–99% (Sem, 1991). These *g*-values show these are electron centers. Toyoda and Schwarcz (1997) observed an EPR signal in quartz grains at $g = 2.0005$, which is usually attributed to the E'_1 center. However, the signal

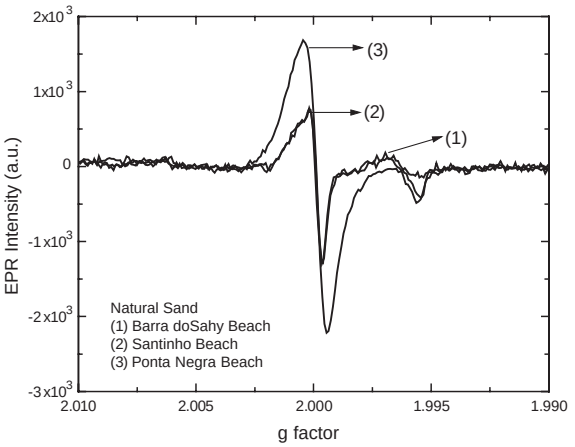

Fig. 1. EPR spectra of the different natural sand samples irradiated (^{60}Co) with 10 kGy.

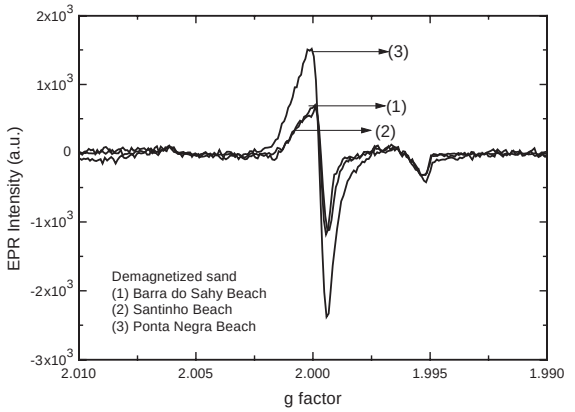

Fig. 2. EPR spectra of the different demagnetized sand samples irradiated (^{60}Co) with 10 kGy.

Table 1
Neutron activation analysis results of sand samples: concentration of the elements above 10 μg g^{-1}

Brazilian beaches						
	Barra do Sahy		Santinho		Ponta Negra	
Elements	A1	A2	B1	B2	C1	C2
Al (μg g^{-1})	7015 ± 86	5765 ± 86	4701 ± 51	3760 ± 51	8099 ± 81	5476 ± 70
Ca (μg g^{-1})	4075 ± 299	952 ± 70	2999 ± 220	267 ± 20	27666 ± 2033	2168 ± 159
Mn (μg g^{-1})	94 ± 3	12.8 ± 0.9	21.7 ± 1.3	5.5 ± 0.6	992 ± 29	2.4 ± 0.2
Na (μg g^{-1})	322 ± 66	141 ± 36	146 ± 15	66 ± 10	444 ± 24	342 ± 10
Th (μg g^{-1})	18.41 ± 0.08	1.73 ± 0.01	0.83 ± 0.02	0.42 ± 0.01	17.3 ± 0.07	7.03 ± 0.03
Ti (μg g^{-1})	1588 ± 113	286 ± 58	667 ± 57	134 ± 37	28615 ± 1445	461 ± 54

A1, B1 and C1 are natural sand samples, and A2, B2 and C2 are desmagnetized sand samples.

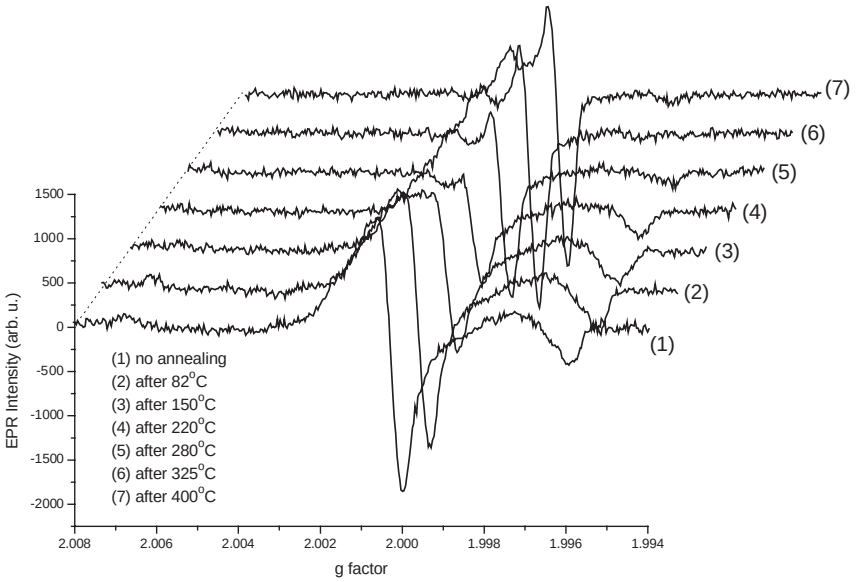

Fig. 3. EPR spectra of demagnetized sand from Ponta Negra Beach after irradiation (5 kGy) and consecutive isochronal annealing (15 min).

observed by them was slightly different from the E'_1 center. The name of the counterfeit E'_1 center was attributed to this signal, because it exhibits the same dependence of intensity on microwave power (it saturated around 0.4 mW). The counterfeit E'_1 center showed a different behavior to the E'_1 center after successive post-irradiation thermal treatments, namely the counterfeit E'_1 center is less stable than the E'_1 center. A thermal treatment of 170 °C for 15 min was enough to quench this center. After this thermal treatment, the observed spectrum was due only to the E'_1 center that is normally quenched only at 450 °C (Toyoda and Schwarcz, 1997).

In this work, the signal observed at $g = 1.999$ presents similar characteristics (mainly the thermal behavior) to those observed by Toyoda and Schwarcz (1997). Fig. 3 shows the spectra of a demagnetized sand sample from Ponta Negra Beach, after irradiation to 5 kGy followed by consecutive isochronous thermal treatments (15 min). The signal of the E'_1 center starts to appear about 150 °C; its intensity increased after the sample was submitted to the thermal treatments at 150 °C until 325 °C. On the other hand, the response to microwave power of the signal at $g = 1.999$ did not follow the behavior described by Toyoda and Schwarcz (1997). The EPR intensity saturated above 2 mW and started to decrease only after 5 mW, as can be seen in Fig. 4. However, the results presented in Fig. 3 suggest that the observed signal at $g = 1.999$ is probably due to the counterfeit E'_1 center.

The signal at $g = 1.995$ was not identified, and its intensity saturated easily around 1 kGy; therefore it is less interesting from the dosimetric point of view than the signal at $g = 1.999$.

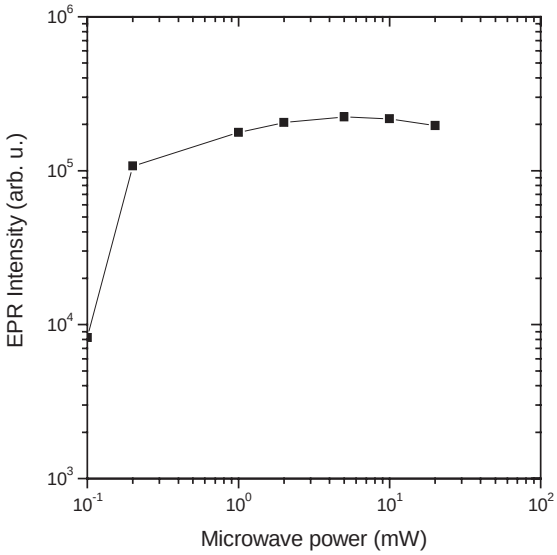

Fig. 4. EPR intensity of signal at $g = 1.999$ as a function of MW power for demagnetized sand from Ponta Negra Beach. The signal was obtained after 30 kGy of γ-radiation (^{60}Co).

The corresponding hole centers must exist, probably $[AlO_4/h]^0$ centers, due to high content of this impurity (see Table 1). Its EPR spectrum is observable only at low temperatures, around 77 K.

Fig. 5 shows the EPR spectra of the sand samples from Ponta Negra Beach, irradiated with different

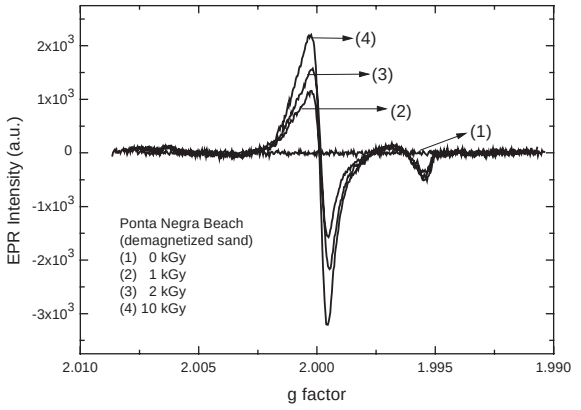

Fig. 5. EPR spectra of demagnetized sand samples from Ponta Negra Beach irradiated to different absorbed doses (^{60}Co).

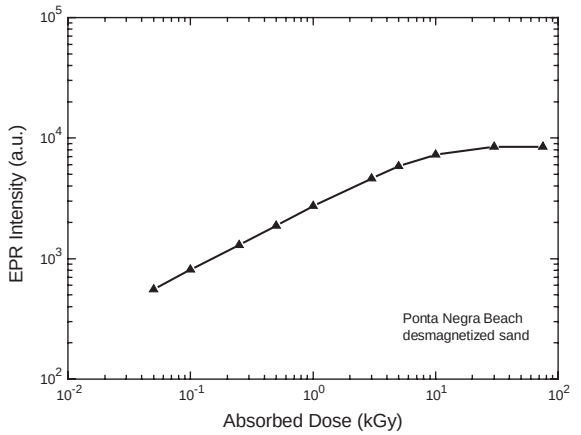

Fig. 6. Calibration curves of EPR signal at $g = 1.999$ for demagnetized sand samples from Ponta Negra Beach for ^{60}Co radiation.

absorbed doses of 1, 2 and 10 kGy. In this work, the main dosimetric characteristics of the signal at $g = 1.999$ of the samples from Ponta Negra Beach were studied, because these sand samples showed the best signal among all other sand samples.

3.1. Reproducibility

Groups of five sand samples (Ponta Negra Beach) were submitted five times to the same procedure of thermal treatment at 500 °C/1 h (defined for reutilization) and irradiation (5 kGy), in order to study the response reproducibility. The maximum standard deviations obtained were approximately 7.4% for the natural sand sample and 6.1% for the demagnetized sand sample.

3.2. Dose response

The sand samples (Ponta Negra Beach) were irradiated to various doses in the range between 50 and 75 kGy. Fig. 6 presents the calibration curve. The maximum standard deviation obtained of these measurements was 2.0%. The dose response showed (Fig. 6) sublinearity, and it is proportional to the square root of dose, but saturation effects are clearly evidenced about 50 kGy. This behavior is another evidence that the center at $g = 1.999$ is the counterfeit E'_1 center. Toyoda et al. (1996) observed that the saturation of the dose response of the E'_1 center occurs only at 100 kGy.

3.3. Fading

The thermal fading of the sand samples (Ponta Negra Beach) was studied by taking daily measurements up to 30 days; this effect cannot be neglected. A response decay occurs after irradiation of the sand samples (5 kGy). The response results in a reduction of the

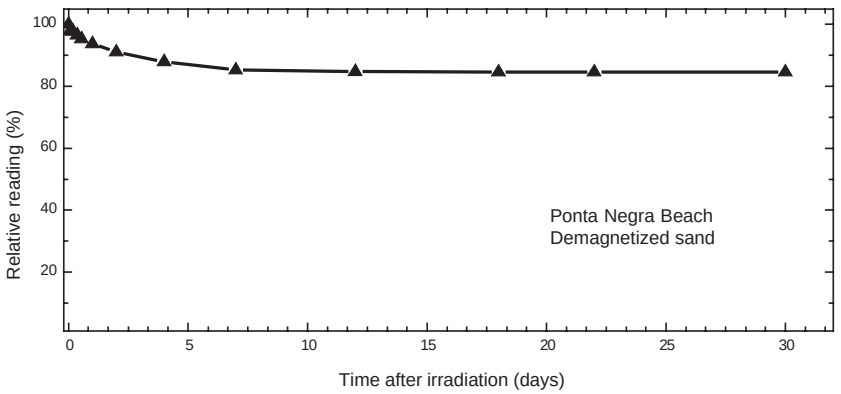

Fig. 7. EPR intensity of signal at $g = 1.999$ as a function of post-irradiation time for evaluation of the fading of demagnetized sand from Ponta Negra Beach.

EPR spectra of about 16% (natural sand) and 8% (demagnetized sand), 24 h after the irradiation; afterwards, it shows a slow decay with tendency to a constant value that occurs after about 7 days post-irradiation time, as can be seen in Fig. 7.

4. Conclusions

The dosimetric characteristics studied in this work show that sand samples may be used in high-dose dosimetry, taking into account room temperature fading. The basic advantage of sand samples is their very low cost. Sand samples may be suitable for dosimetry in the main radiation processes of seed stimulation, mutation breeding, industrial radiography, insect population control, pasteurization and water purification.

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