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Running Title*: Sand for high doses*

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INFLUENCE OF THERMAL TREATMENTS ON THE RESPONSE OF SAND RADIATION DETECTORS FOR HIGH DOSE DOSIMETRY

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ABSTRACT

The dosimetric properties of sand from Brazilian beaches have shown to be useful for high dose dosimetry. The thermoluminescent (TL) and electron paramagnetic resonance (EPR) techniques were utilized, and the sand samples were recently studied in relation to their main dosimetric properties. The EPR signal at g = 1.999 grows significantly in function of the absorbed dose, and the TL peaks appear at 110 °C and 170 °C. However, these sand samples present a post-irradiation thermal decay at room temperature, that is a problem for dosimetric procedures. In this work sand samples have been studied in relation to different thermal treatments. Post-irradiation treatments were performed at 50 °C up to 230 °C.

INTRODUCTION

The processes of irradiation of high doses have been developed for application in the areas of industry, medicine and agriculture [1].

Sand is a material found in the nature in great amounts; in the decade of nineties this material presented interest for its dosimetric properties. Sand is mainly constituted of quartz and feldspar, presenting the phenomenon of termoluminescece (TL). Sand contains also some other elements in smaller amounts [2,3]. The group of silicates represents one type of materials of greatest amount and importance in the world.

Vaujapurkar and Bhatnagar (1993) studied sand proceeding from Rajasthan, India, that presented two TL peaks at 80 °C and 220 °C. The first peak is not useful for dosimetry due to it its fast thermal decay at ambient temperature, while the second TL peak presents an adequate response in function of absorbed dose.

One of the compounds of sand, quartz (SiO_4), has been tested using the electron paramagnetic resonance technique (EPR); its importance as an insulating material, with paramagnetic defects, has already been demonstrated [4,5,6].

Teixeira and Caldas[7] and Teixeira et al.[8], utilizing TL and EPR techniques, studied the dosimetric properties of sand samples from different Brazilian beaches: response to gamma radiation of ^{60}Co, reproducibility, lower detection limits and fading. In these papers sand samples from Barra do Sahy Beach, São Paulo; Santinho Beach, Santa Catarina; and Ponta Negra Beach, Rio Grande do Norte were investigated.

The sand samples from Ponta Negra Beach were chosen, among all other sand samples, for this study, because they showed the best TL and EPR signals.

In this work effort has been given to the fading characteristic of the material. The effects of pre- and post-irradiation thermal treatments on TL and EPR response have been studied in order to improve the response sensitivity of sand samples, and to determine the most adequate temperature to minimize the post-irradiation fading of the samples.

MATERIALS AND METHODS

Sand samples originating from the Ponta Negra Beach, Rio Grande do Norte State, Brazil, were studied.

Sand was bolted to uniform grains size between 0.037 mm and 0.074mm in diameter. To eliminate organic impurities, the sand samples were washed with a solution of hydrochloric acid 1M (1 molar), and then rinsed with distilled water to remove the HCl. The sand samples were dried in an electric oven (Formitex) at 75 ºC during 1hour. Magnetic particles (mainly iron) were removed from some sand samples, using a magnetic separator (S.G.Frantz Com. Ind., Isodynamic, model L-1).

To facilitate the sample handling, sintered sand pellets were prepared at the Laboratory for Production of Dosimetric Materials, IPEN, using Teflon as binder. Sintering of the pellets was achieved with a thermal treatment of 300ºC for 1hour followed by another thermal treatment of 400ºC for 1.5hour. The sand pellets were divided into two batches: natural sand samples and demagnetized sand samples, with diameter of 6 mm and thickness of 0.8 mm, for use with the TL technique.

Thermal treatments at 300ºC/1h (TL pellets) and 500ºC/1h (EPR samples) were applied to the sand samples for reutilization. Post-irradiation thermal treatments were performed at 50ºC up to 230ºC during 15 min, with the objective to empty the instable electron traps, in order to show an acceptable stability in function of room temperature storage time, as recommended by Cameron [10].

The irradiations were performed using a Gamma-Cell 220 system (^{60}Co, 4.34 kGy/h, May 2004) of the Center for Radiation Technology, IPEN. The irradiations were made at ambient temperature, and the samples were fixed between 3 mm thick polymethyl meth-acrylate plates (Lucite), to guarantee the occurrence of electronic equilibrium during the irradiations.

The evaluation of the sintered sand pellets was carried out with a thermoluminescent reader (Harshaw Nuclear Systems, model 2000 A/B) using a heating rate of 10°C/s, an infrared filter and a S-11 type photomultiplier tube (2" diameter). All TL measurements were integrated between 50ºC and 300ºC.

The electron paramagnetic resonance (EPR) measurements were carried out using a Bruker EMX spectrometer with a rectangular cavity (ER4102 ST), at room temperature, with microwave frequency of 9.76 GHz (band X), microwave power of 0.2 mW, and with field modulation frequency and amplitude of 100 kHz and 0.1mT, respectively. This EPR spectrometer belongs to the Multi-user Group of the Institute of Physics.

The sand powder samples, with an average mass of (150 ± 1) mg, were measured inside of a pure quartz tube.

Due to the thermal fading of the TL and the EPR responses of the sand samples, the first measures in this work were taken exactly one hour after the irradiations.

RESULTS AND DISCUSSION

The thermoluminescent glow curves of the sand pellets irradiated (^{60}Co) with 10 kGy present two peaks at about 110ºC and 170ºC, as shown in Figure 1.

Figure 2 presents the EPR spectra of the natural and demagnetized sand samples after irradiation with 10 kGy. The EPR spectra present signals at g = 1.999 and g = 1.995. The signal at g = 1.995 is less interesting of the dosimetric point of view than the signal in g = 1.999 because its intensity saturates easily around 1 kGy (8). The signal at g = 1.999 is probably resulting from the counterfeit E'_1 centres (8, 9).

Thermal Fading

The thermal fading of the sand samples was determined by taking daily measurements up to 30 days after the irradiation using integrated TL area. After the first 24 h post-irradiation time, the natural and demagnetized sand pellets present a reduction of their TL response of about 20%; afterwards follows a slow decay, with a tendency to a constant value after about 15 days post-irradiation time, using a TL reader (Figure 3).

The EPR response presents a fading of about 16% (natural sand) and 8% (demagnetized sand), after 24 hours post-irradiation; however, it shows a decay with tendency to a constant value that occurs after about only 7 days post-irradiation time (with total reductions of 25% and 15% for natural and demagnetized sand samples, respectively), as shown also in Figure 4. During the storage time the sand samples were kept at room temperature of (23 ± 2) °C and humidity of (60 ± 6)%.

Post-irradiation thermal treatments

The sand samples were thermally treated during 15 minutes at different temperatures after an irradiation of 5 kGy (^{60}Co). The results are shown in Figure 5 for the TL and EPR measurements.

In Figure 5 the fading, in function of the temperature of thermal treatment, for each TL peak is compared with the fading of the EPR signal at g = 1.999 (counterfeit E'1 centre). The natural and demagnetized samples present similar behaviour.

After the thermal treatment around 100 °C the TL peak at 110°C disappear. The TL peak at 170°C disappears after the thermal treatment at 200°C, together with the EPR signal at g = 1.999, suggesting that the corresponding traps to the 170°C TL peak may be related to the counterfeit E'1 centre.

CONCLUSIONS

The effect of post-irradiation thermal treatments of the sand samples showed that the intensity of the 170°C TL peak and the EPR signal of the counterfeit E'_1 center are quenching at the same temperature, 200°C.

In order to stabilize TL response of the sand samples, we consider that the sand samples should be submitted to post-irradiation thermal treatments at 100 °C during 15 minutes to eliminate of the 110°C TL peak. This TL peak is the responsible for the greater part of fading shown in Figure 3.

The removal of magnetic particles is highly recommended, because the TL and EPR signals of the demagnetized sand samples are more intense than the natural sand samples. Furthermore, the dose response, in function of room temperature storage time, of the demagnetized sand sample is more stable than the natural one.

The present study shows that the sand samples (their main dosimetric properties were demonstrated elsewhere [7,8]) may be useful for high dose dosimetry in areas such as industry, medicine and agriculture, due to their reduced size, easy handling and very low cost.

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FIGURE CAPTIONS

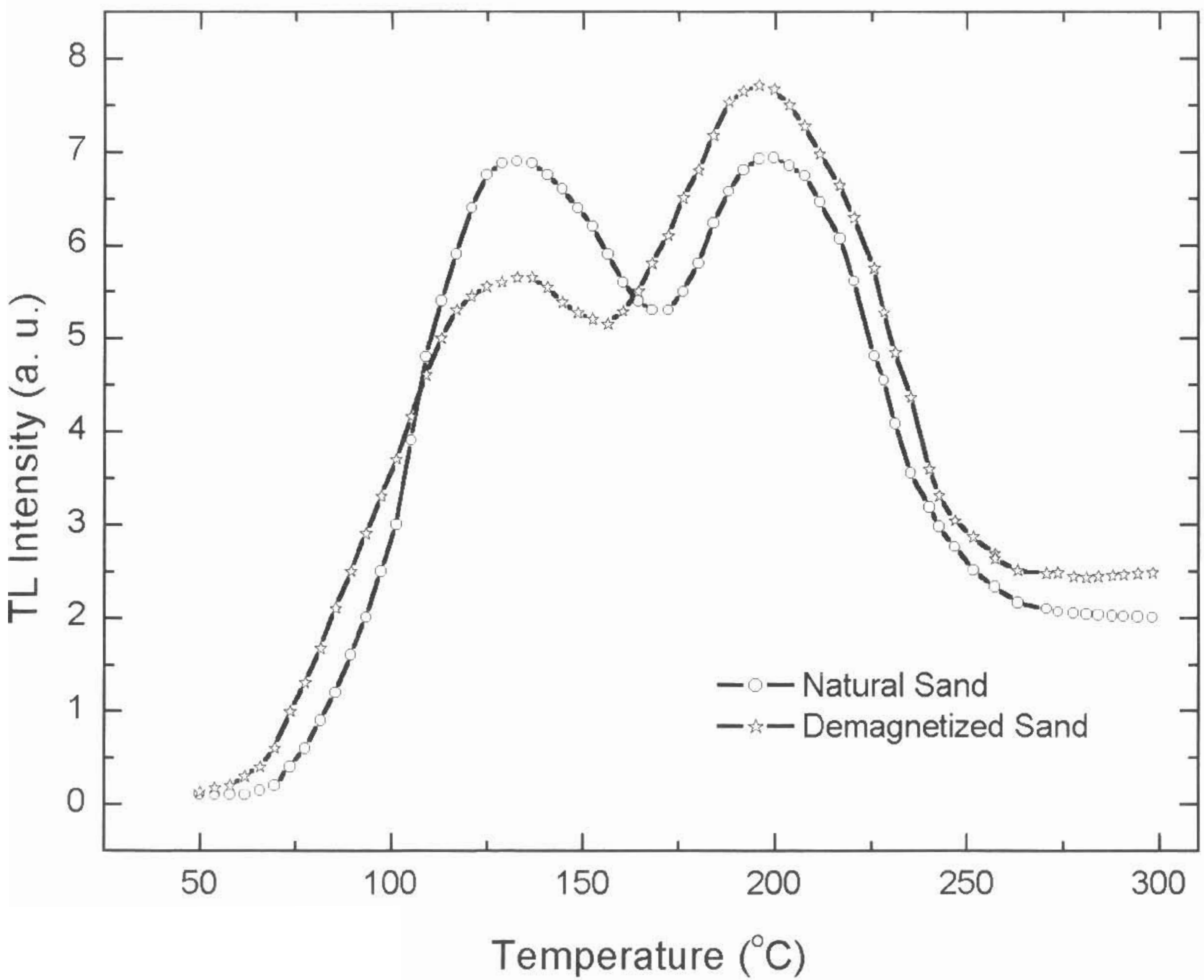
Figure 1: TL glow curve obtained of the natural and demagnetized sand samples irradiated with 10 kGy (^{60}Co).

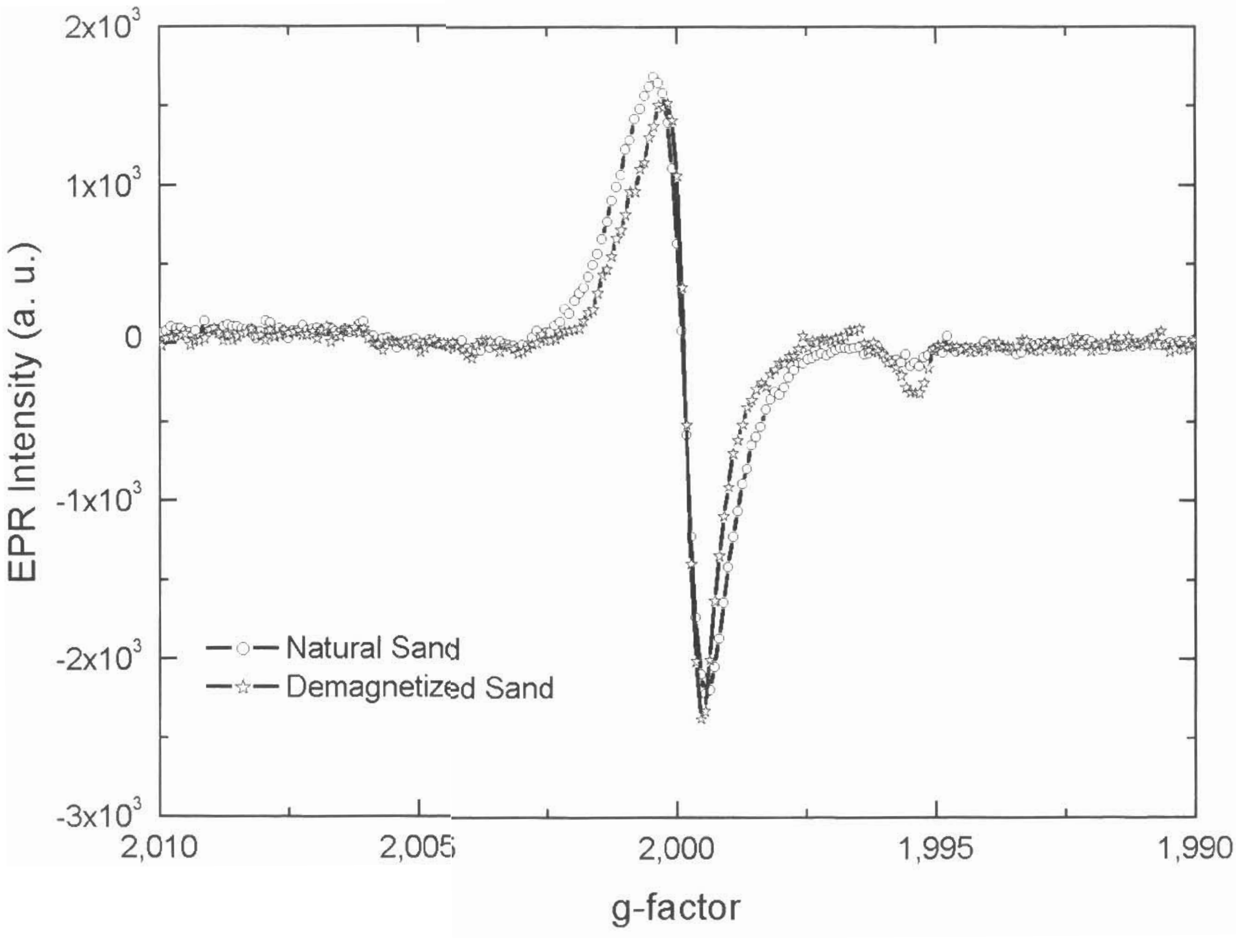
Figure 2: EPR spectra obtained of the natural and demagnetized sand samples irradiated with 10 kGy (^{60}Co).

Figure 3: (A) Thermal fading at room temperature up to 30 days of natural and demagnetized sand pellets, after an irradiation of 5 kGy (^{60}Co), using the integrated TL intensity (50°C – 300°C). (B) Thermal fading at room temperature up to 24 h post-irradiation time.

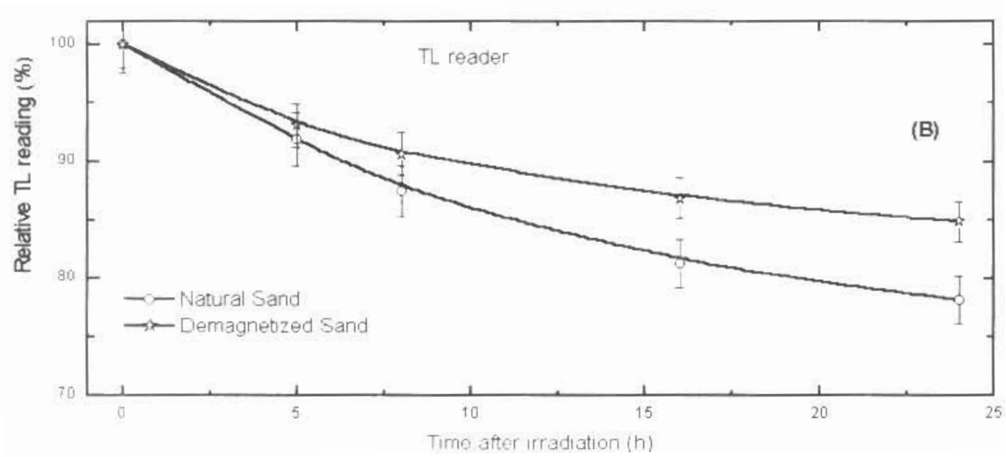
Figure 4: (A) Thermal fading at room temperature up to 30 days of natural and demagnetized sand pellets, after an irradiation of 5 kGy (^{60}Co), using the EPR signal at g = 1.999. (B) Thermal fading at room temperature up to 24 h post-irradiation time.

Figure 5: Effect of post-irradiation thermal treatments (15 min) at different temperatures, for natural and demagnetized sand samples irradiated with 5 kGy (^{60}Co); using the 110°C and 170°C TL peaks and the counterfeit E_1' EPR signal.

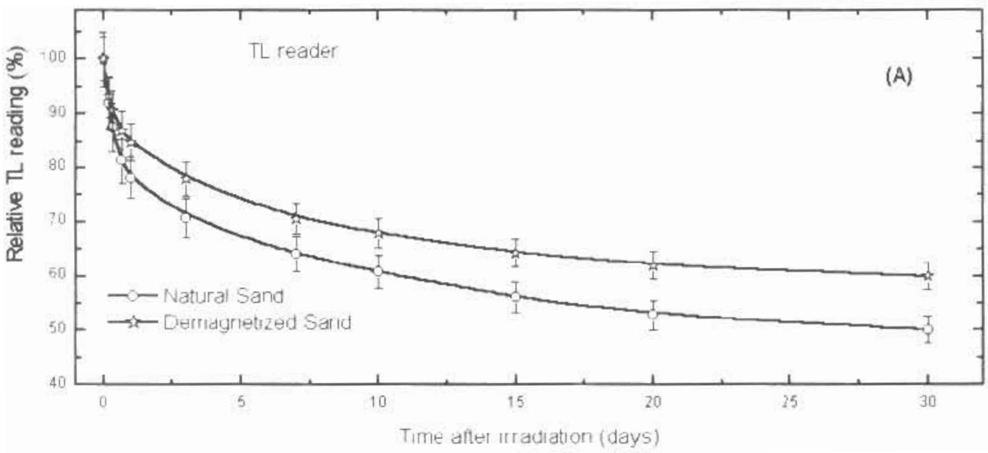

TL Intensity (a. u.)
0
1
2
3
4
5
6
7
8
50
100
150
200
250
300
Temperature (°C)
Natural Sand
Demagnetized Sand



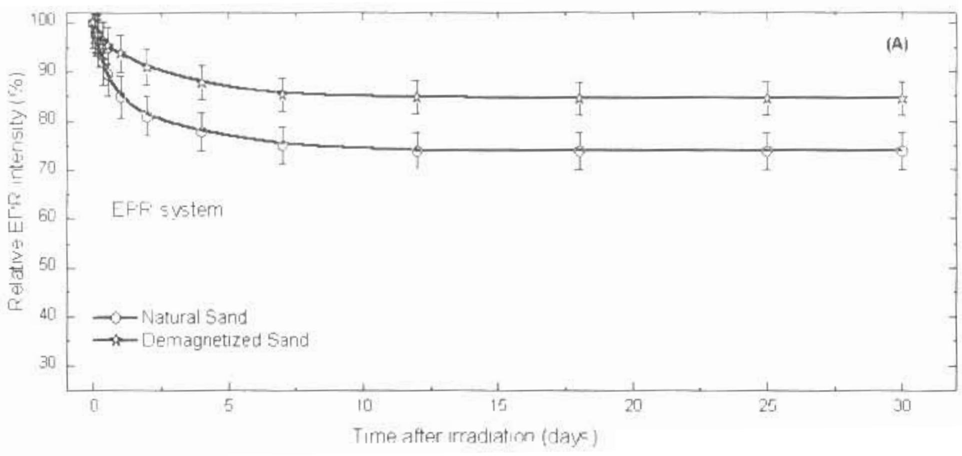
EPR Intensity (a. u.)
2x10^3
1x10^3
0
-1x10^3
-2x10^3
-3x10^3
2,010
2,005
2,000
1,995
1,990
g-factor
Natural Sand
Demagnetized Sand

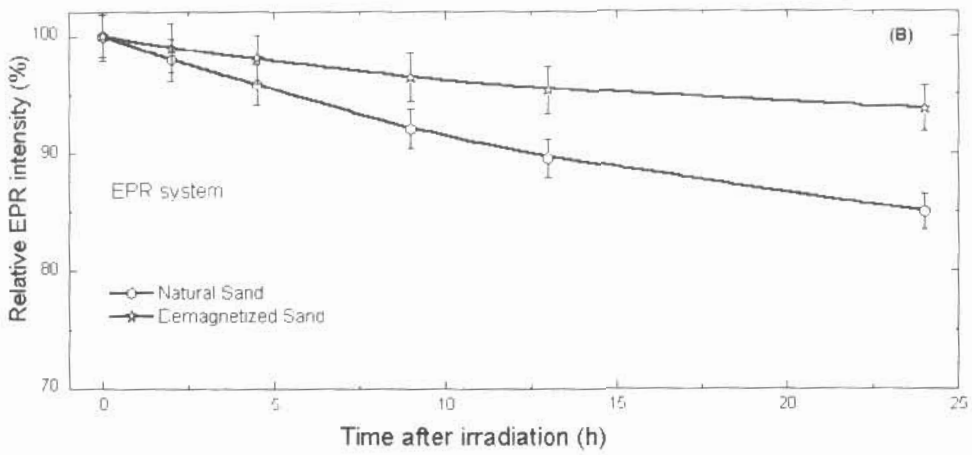


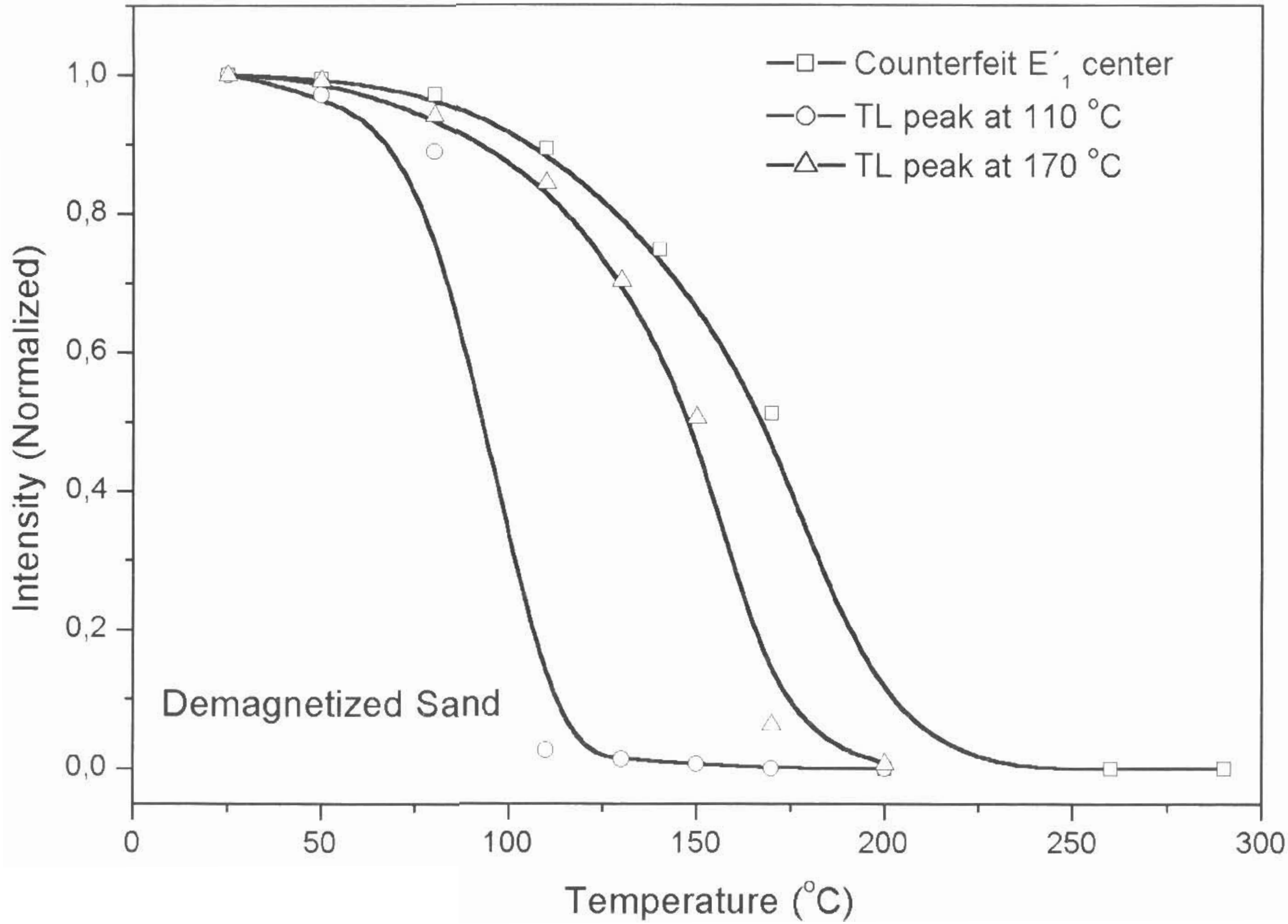
TL reader
(B)
Relative TL reading (%)
Time after irradiation (h)
Natural Sand
Demagnetized Sand
100
90
80
70
0
5
10
15
20
25



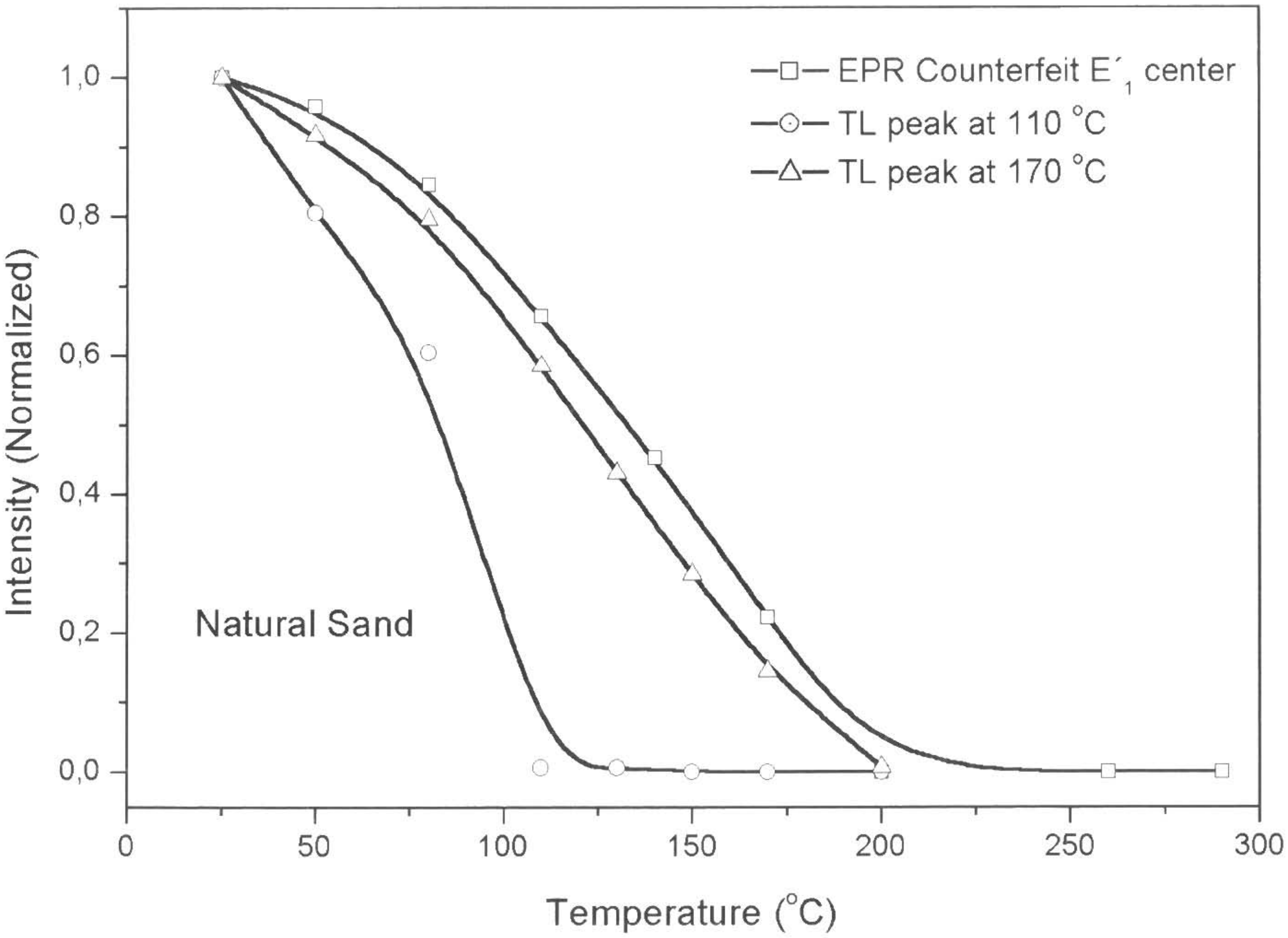
TL reader
(A)
Relative TL reading (%)
Time after irradiation (days)
Natural Sand
Demagnetized Sand

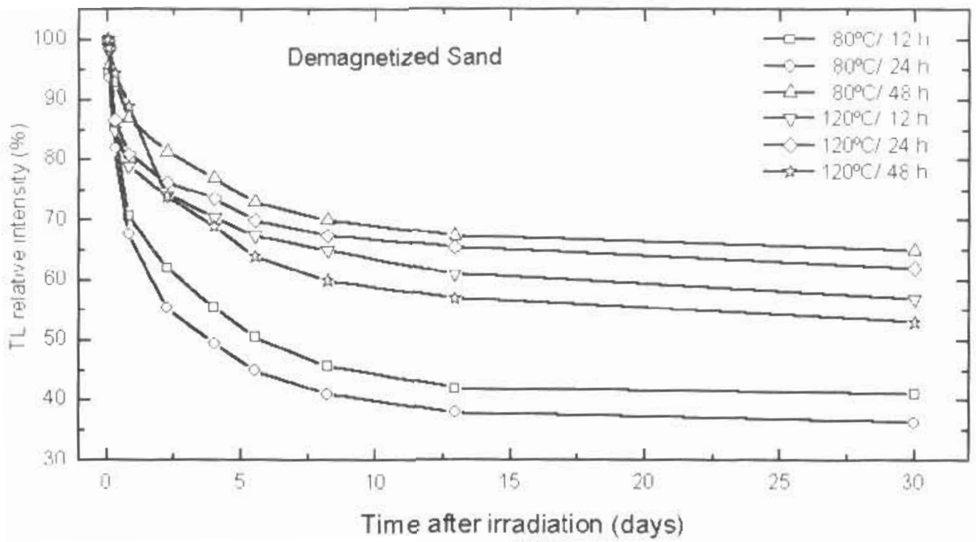

(A)
EPR system
Natural Sand
Demagnetized Sand
Relative EPR intensity (%)
Time after irradiation (days)

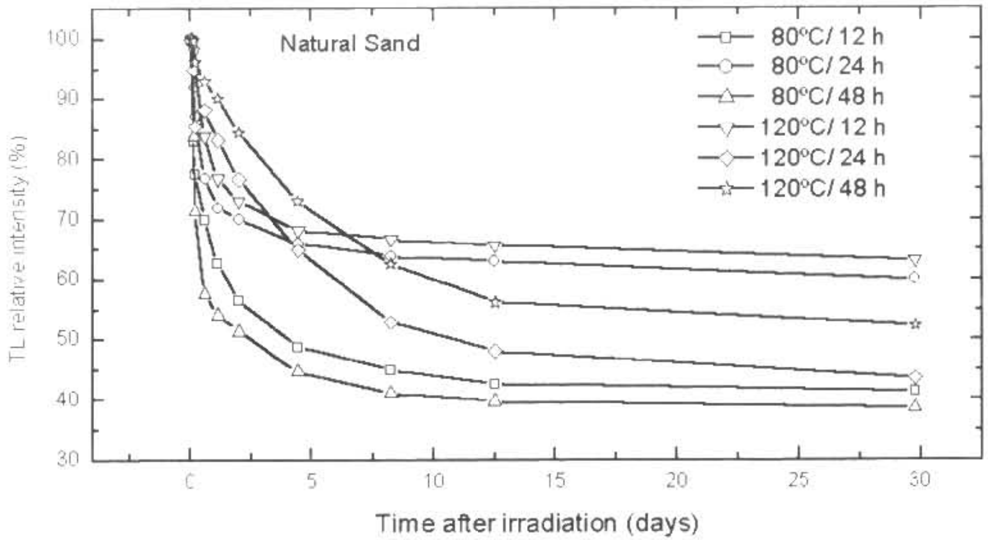

(B)
EPR system
Natural Sand
Demagnetized Sand
Relative EPR intensity (%)
Time after irradiation (h)
100
90
80
70
0
5
10
15
20
25



Counterfeit E´₁ center
TL peak at 110 °C
TL peak at 170 °C
Demagnetized Sand
Intensity (Normalized)
Temperature (°C)
1,0
0,8
0,6
0,4
0,2
0,0
0
50
100
150
200
250
300


EPR Counterfeit E´1 center
TL peak at 110 °C
TL peak at 170 °C
Natural Sand
Intensity (Normalized)
Temperature (°C)
1,0
0,8
0,6
0,4
0,2
0,0
0
50
100
150
200
250
300


Demagnetized Sand
80ºC/ 12 h
80ºC/ 24 h
80ºC/ 48 h
120ºC/ 12 h
120ºC/ 24 h
120ºC/ 48 h
TL relative intensity (%)
Time after irradiation (days)


Natural Sand
80ºC/ 12 h
80ºC/ 24 h
80ºC/ 48 h
120ºC/ 12 h
120ºC/ 24 h
120ºC/ 48 h
TL relative intensity (%)
Time after irradiation (days)
100
90
80
70
60
50
40
30
0
5
10
15
20
25
30