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THERMOLUMINESCENCE OF BEACH SANDS RELEVANT TO RADIATION MEASUREMENTS

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ABSTRACT

In this study the thermoluminescence(TL) properties of natural beach sand sample collected from Saudi Arabia were investigated for the purpose of determining whether it is suitable as a dosimetric material or not. The experiments carried out can be outlined as the effects of pre-irradiation annealing procedures on TL sensitivity and calculating the kinetic parameters. The dose response has a linear behavior over the dose range 0.1Gy-100Gy followed by sub-linearity at higher dose level. The variation in TL signal over two weeks of storage period at room temperature was ~27%. From our results it is possible to conclude that beach sand over the dose range of 0.1Gy -100Gy is a suitable material for dosimetric applications.

1.Introduction

Sand is a natural material easily available in large quantities. Two important constituents of sand are quartz and feldspar which are well known to show thermoluminescence. Sand also contains varying concentrations of heavy minerals. It can be used as a dosimeter during nuclear emergencies like reactor accidents for monitoring a large number of people [1].

The thermoluminescence (TL) properties of typical sand from India have been studied for possible use as a high gamma dosimeter [1]. Earlier studies showed good dose response linearity in the used range [2]. The dependences of the TL spectra on radiation dose were examined for both laboratory dose and natural dose, which differ by 9 orders of magnitude in dose rate, no effect due to dose rate was observed.

The dose responses of both blue and red emission bands of quartz were examined in the range 0Gy-1600Gy and were found to be highly nonlinear and different. It is shown that the pre-dose effect in the high temperature thermoluminescence peak is associated with the blue emission and not the red emission [3]. The search using X-ray diffractometry and instrumental neutron activation analysis proved that the cause of distinctly different colorations was attributable to the impurity atoms; while structural defects were yielding the blue TL color images [4]. The study of TL characteristics of quartz extracted from sandstones has allowed the approximate determination of the temperature to which they had been exposed [5].Rendell et al.[6] reported on spectral changes induced in the TL of quartz as a result of a variety of thermal treatments. As a result in volcanic quartz material, the ratio of red to blue emission is particularly sensitive to the cooling rate. The blue thermoluminescence (BTL) sensitivities from natural and synthetic quartz samples showed a negative relationship with the Al contents above a few tens of ppm [7]. Scholefield and Prescott [8] study concluded that the blue and red emissions probably involved a common set of electron traps feeding separate luminescence centers, not necessarily in the same grains.

There are some applications of sand as a dosimeter presented in literature. Benny et al [9] reported the application of sand as an in-situ dosimeter for estimating gamma dose observed by sludge during its sterilization. Sand samples have also been used for dating [10]. Investigation of sand as a TL dosimeter for radiotherapy was undertaken by Pitalua et al.[11]. Sand near a nuclear power plant can be used for dose estimation in the case of nuclear emergencies. Sand samples can also be used as ESR dosimeters for different applications in medical, agricultural and industrial areas [12].

Although there has been a lot of work done related to application areas of sand samples in literature, there are no TL studies on sand samples from the Kingdom of Saudi Arabia .The aim of the present work is to carry out some TL studies relevant to gamma measurements on the beach sand sample such as, effect of thermal treatments, glow curves, gamma dose response, batch homogeneity and storage effects using some techniques in Topakasu et al. [13].

2. Material and methods

The sand sample used in our study is poorly graded sand, with silt taken from Jeddah, the west coast of Saudi Arabia. The sample was sieved and fractions of 75μm were used for TL measurements. The sample was washed with 1N hydrochloric acid; after that distilled water was used to remove the HCl and inorganic impurities. The sample was then allowed to dry at 50^{o}C. Magnetic particles were removed using Franz Magnetic Separator. After washing, drying and removal of magnetic particles, the sand sample was encapsulated in plastic capsules and kept in dark until measurements were achieved.

The results of the chemical composition of the sample obtained by the atomic absorption spectrometry are shown in Table 1.

Table 1: The chemical composition of beach sand.

Element/Compound	Ratio (wt. %)
SiO_2	89.3
Al_2O_3	0.348
Fe_2O_3	0.018
CoO	0.204
CuO	5.070
PbO	3.656

Irradiations were performed at room temperature (25^{o}C-30^{o}C) in air using a ^{60}Co gamma ray source. TL measurements were monitored using a Harshaw 3500 TLD reader. Light pulses were detected by the photomultiplier tube provided with a narrow band blue filter plus Schoot BG39 glass filters of blue- violet transmittance band. A linear heating rate of 5^{o}Cs^{-1} was chosen; heating the sample from room temperature up to 400^{o}C. The incandescent background was measured then subtracted from the data. To minimize the statistical error, five aliquots each of 7 mg were used for each measurement.

All irradiations and measurements were performed in King Saud University, Saudi Arabia.

3. Experimental results and discussion

3.1 TL glow curves of beach sand before and after annealing process

For investigation of the characteristic TL glow curves, the natural sand sample was heated from room temperature up to 400°C and the TL intensity recorded. Then a second readout was performed to record the background signal of the reader and the sample. An average TL glow curve of the natural sand aliquots without any pre-annealing or irradiation treatments can be seen in Fig.(1).

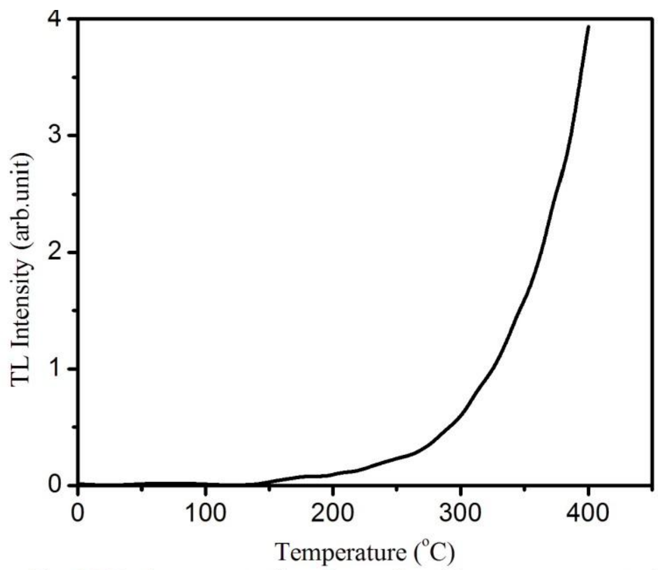

Fig. 1 TL glow curve of sand sample without any pre-annealing or irradiation treatment (The back ground is already subtracted).

3.2 Thermal treatment

The natural sand aliquots were pre-heated at different temperatures ranging from 250°C-700°C, for 15 min, cooled in air and subsequently irradiated up to γ-dose of 50Gy. After 1 h of irradiation the TL glow curves were read out. Important modifications in the shape of the glow curve are observed

(Fig. 2).

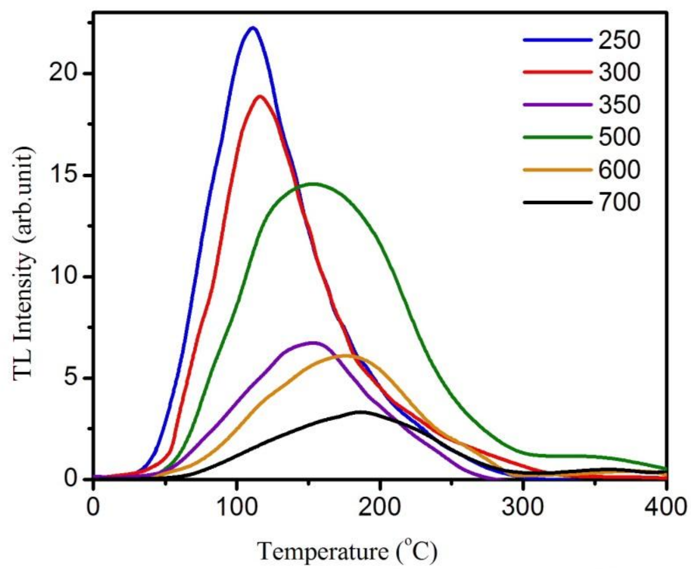

Fig. 2 TL glow curves of sand sample pre-annealed at different temperatures changing from 250°C up to 700°C.

Treated aliquots show one main TL peak starting at~130°C after annealing at 250°C and gradually shifted to 190°C after annealing at 700°C (Fig. 3).

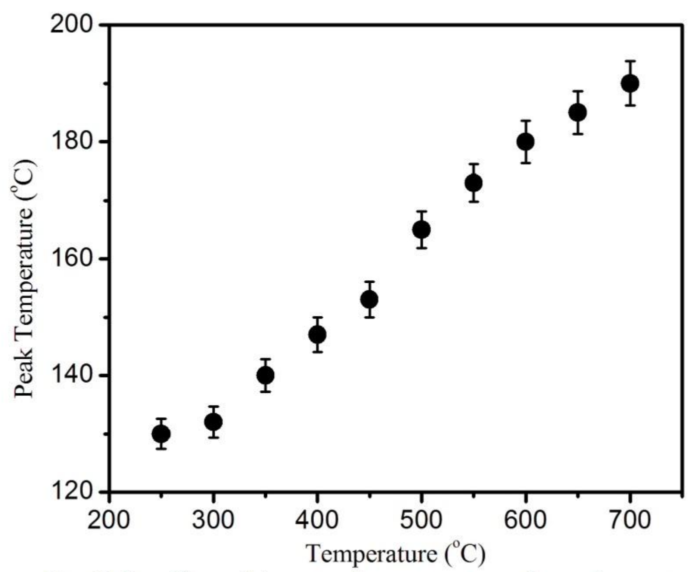

Fig. 3 The effect of the annealing process, on the peak temperature of the TL glow curves of sand.

Figure (4) shows the normalized TL sensitivity (i. e. the TL output per unit test dose) measured in the range from room temperature up to 400 ^{o}C as a function of annealing temperature. The main effect of this thermal treatment is an increase of the TL signal with increasing temperature. The sensitivity of sand sample annealed at 500 ^{o}C is found to be ~ 1.5 times that annealed at 250 ^{o}C. This sensitizing effect of thermoluoninescence may be due to heat- induced changes in the population of radiation induced defects or in the diffusion of impurities to the lattice, or interstitial sites at which they constitute effective trapping centers [14.15]. The reduction of TL intensity at temperatures ≥ 550 ^{o}C can be attributed to thermal quenching [16]. As can be seen in Fig.(4), the changes of TL sensitivity become smaller around 400 ^{o}C. Eventually in this study, the most convenient annealing temperature was 400 ^{o}C.

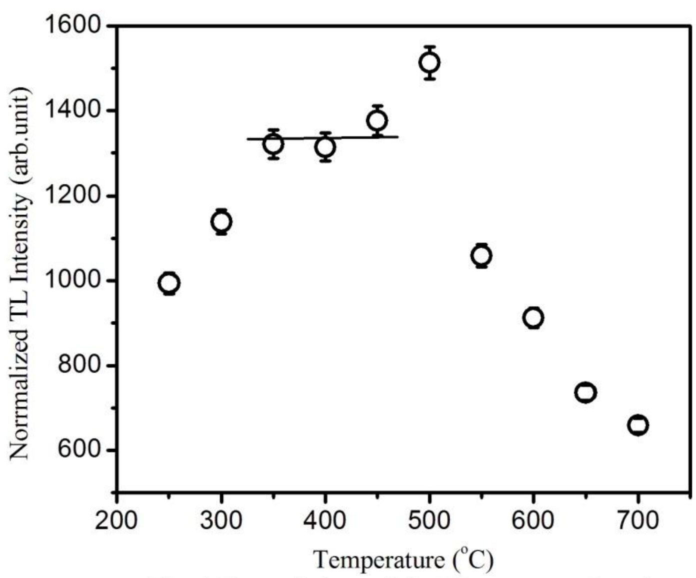

Fig. 4 The variations of the TL response of sand with the pre-heating temperature.

3.3 Gamma irradiations

In order to examine the TL glow curve of beach sand aliquots after annealing and irradiation process, the aliquots were annealed with the annealing receipt of 400°C (15min + cooling to room temperature). Afterwards they were irradiated with different doses in the range 0.1 Gy-1000 Gy and the TL glow curves were recorded 30 min after irradiations. As it is seen in Fig.(5 a), the TL glow curve has one peak ∼ 145°C after annealing and irradiation to a gamma dose of 10Gy then shifted to slightly higher temperature at ∼148°C with increase of dose to 50 Gy. In the high dose range 0.25 kGy-1 kGy (Fig. 5 b) two peaks appeared; the first one at 153°C, which shifted to a higher temperature at 165°C, the second peak appeared at 293°C, and shifted to a lower temperature at 285°C. The previously investigated beach sand [11] exhibited the same low temperature peaks at ∼145°C and 148°C.

3.4 Dose response

The plot of the TL signal versus dose, yields the so called 'growth curve'. The signal was estimated as an integrated TL signal over the temperature range from 50°C to 400°C. Gamma irradiations were obtained by carrying out laboratory irradiations with a ^{60}Co source, in the range from 0.1 Gy to 1000Gy.

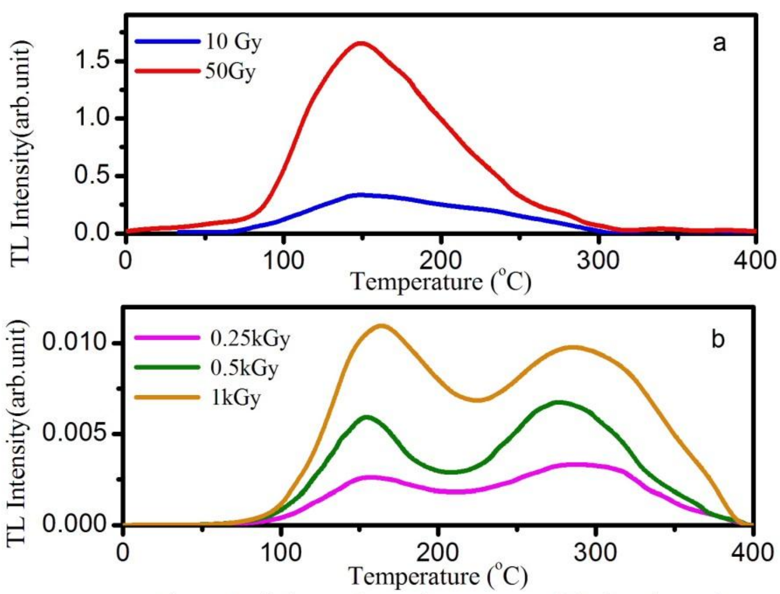

Fig.5: Variations of TL glow curves of the beach sand with the irradiation gamma dose.

After 30 min of irradiations the TL signal was read out. Figure (6) shows the dose response of beach sand over the dose range 0.1 Gy - 1000Gy. The TL response follows linear relation $Y=0.0001X+ 0.013$ followed by sub linearity $Y=0.0047\ X^{0.8808}$ with the correlation coefficients $R^2=0.921$and $R^2=0.988$ over the dose ranges 0.1Gy- 100Gy and 200Gy – 1000Gy respectively. Where Y corresponds to the TL intensity per gm and X is the irradiation dose in Gy.

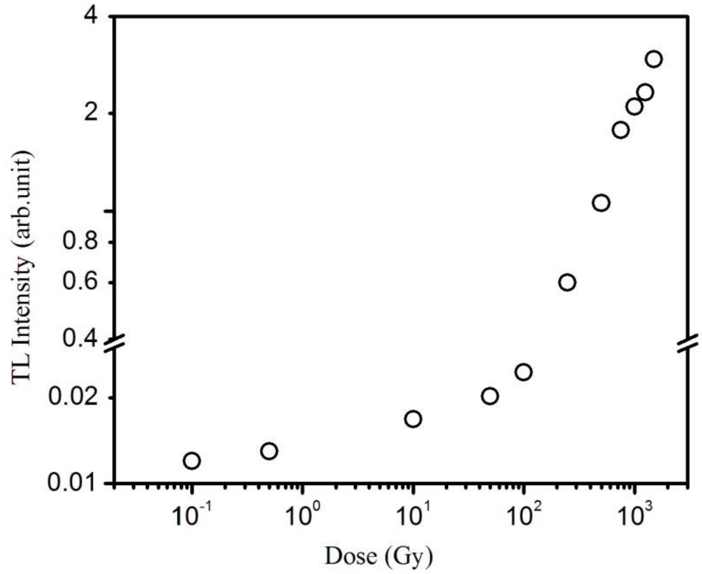

Fig.6: The variability of TL intensity with increament of gamma radiation dose.

The TL intensity of the natural sample (measured from Fig.(1) and estimated from the dose response curve (Fig.6)) corresponds to a laboratory irradiated dose with a ɣ -dose of 1.06 Gy. The minimum dose detected by the investigated material was 0.1Gy, after being subjected to gamma radiation. The previously studied samples [11] exhibited a linear dose response over the range 1 Gy– 10 Gy.

3.5 Kinetic analysis

Calculations of trap parameters related to the TL peaks of beach sand were performed by the 'initial rise' method. This method is known to be based on the assumption that the initial rise of a glow peak is independent of the kinetic order in the TL response, so that it may be represented by the following equation:

$$I \, \alpha \, \exp\left(-\frac{E}{kT}\right) \qquad (1)$$

where I is the TL intensity, E (eV) the activation energy, k (eV/k) the Boltzmann constant and T (K) the absolute temperature. A plot of ln (I) vs. 1/T over the initial part of the peak gives a straight line with slope – E/k. The frequency factor S is given by

$$S = \left(\frac{\beta}{k}\right)\left(\frac{E}{T_m^2}\right)\exp\left(\frac{E}{kT_m}\right) \qquad (2)$$

where β is the heating rate and T_m is the glow peak maximum. In this procedure, the beach sand has discrete sets of traps for both the natural and laboratory – induced glow curves (Table 2).

Table 2: Determined trapping parameters of the main peaks of the beach sand.

T_m(°C) Peak temperature	E(eV) Activation energy	S(s^{-1}) Frequency factor
145	0.1584	143958.735
148	0.44047	1.25E+15
153	0.23717	3.97E+07
155	0.28676	1.53E+09
165	0.28974	4.56E+08
285	0.69441	1.00E+12
290	0.68297	3.69E+11
293	0.62378	2.38E+10

3.6 Storage effects

TL fading of the beach sand studied is as follows: Large amounts of aliquots (annealed at 400°C for min) were irradiated (50Gy) and stored for a period of 14 days (d) in dark, at room temperature before the TL reading was taken. Small amounts of them were used as control samples, first being stored and then irradiated with a dose identical to the above mentioned, and read out immediately on the same day to avoid variations from instrumental drift. Glow curves of the blue band are shown in Fig. 7 after a delay period of t= 14d and without any delay period (t= 0d). The 'plateau test' is also shown in Fig. 7; in which the ratio between the two glow curves; at t = 14 d and the prompt at t = 0 is compared. This ratio is seen to rise from zero to maximum value 0.134 near 140°C.

In Fig. (8) the TL intensity is observed to settle down to a 94% of the initial value after one day of storage. Total fading is obtained as 73% of the TL left after 9 days of storage at room temperature, and remained constant over 14 days. Using this remnant (73%) of TL, the calculated natural dose in Section 3.4 (~1.06Gy) is corrected to 1.452Gy.The previously studied beach sand sample [11] after 5 days of storage at ambient conditions showed a signal decay of 36% approximately , tending to a constant value during the next 30 days.

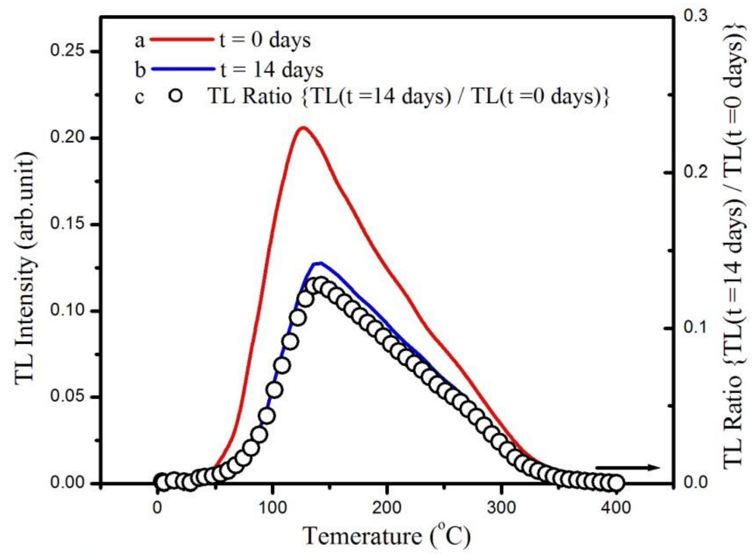

Fig.7: The effect of room temperature dark storage on the laboratory - induced glow curves of beach sand (a) Immediately after radiation ,(b) After 14 days of storage and (c)TL Ratio {TL(t =14 days) / TL(t =0 days)}.

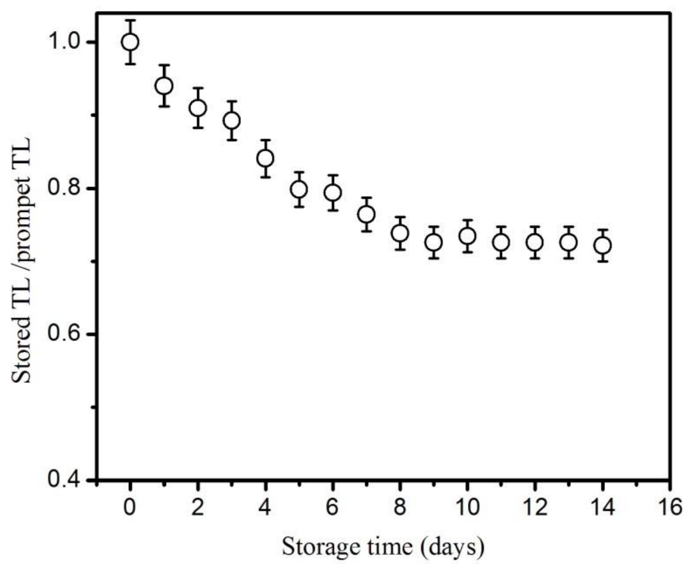

Fig.8: TL fading of the beach sand.

3.7 Batch homogeneity

It is recommended that the evaluated value of the batch homogeneity for any dosimeter will not differ from the evaluated value of any other in the same batch by not more than 30% [17]. Using the following definition of the batch homogeneity (f),

$$f = \frac{TL_{\mathbf{max}} - TL_{\mathbf{min}}}{TL_{\mathbf{min}}} \leq 30\%$$

The average value of f for beach sand has been estimated to be 23.2±1.35% over the low dose range 0.1Gy-100 Gy and 28.3±1.08% over the high dose range 500Gy-1000Gy, which is lower than recommended upper limit.

4. Discussion

By comparing the glow curves of the investigated material with the others [11], it appears that they have the same peaks at 145°C and 148°C when irradiated with low doses. This low intensity is associated with a lower concentration of defects of traps in the material, leading to a lower concentration of electrons in traps and holes in the recombination centers. Initial rise method showed trap depth values in the range 0.158 eV – 0.694 eV and frequency factor values in the range 143958.7 s^{-1} – 1.25E+15 s^{-1}.

The changes of peaks positions with increase of absorbed doses signify that either a multilevel or continuous distribution of trap depths is associated with the main TL peaks [18], which means that these peaks are likely to be a second order kinetics [19]. The sample presented a constant level of TL decay at 73% during the 9 days of storage at room temperature.

5. Conclusion

Beach sand showed a strong natural blue TL signal during the first cycle of heating to 400°C with an estimated natural dose of ~1.452Gy. The annealing study showed one main TL peak that started at 130°C after treatment at 250°C and gradually shifted to 190°C after treatment at 700°C. The TL sensitivity of aliquots annealed at 500°C is found to be 1.5 times of that annealed at 250°C. The TL characteristics of the investigated material such as: small weight (~7mg), simple glow curve, strong natural signal, linear TL dose response, and a good batch of homogeneity, offers beach sand the possibility of being used in radiation dosimetry over the dose range of 0.1Gy – 100Gy. A future study of post radiation heat treatment may reduce TL fading effects and increase sensitivity to radiation.

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