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THERMOLUMINESCENCE PROPERTIES OF RED EMISSION BAND OF SANDSTONE

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ABSTRACT

The red and green emission bands in thermoluminescence (TL) of natural sandstone has been recorded in the range from room temperature to 400°C.The natural glow curves have peaks at 250°C and 285°C in the red and green bands respectively with a heating rate 5°C/s. The laboratory irradiated sample revealed peaks at 115°C, 135oC, 155oC, 195oC, 250oC, and 315°C in the red band and 95°C, 135°C, 285oC, and 315°C in the green band. The TL gamma-rays dose response curves are linear for both bands over the two ranges (1Gy-100Gy) and (500Gy -10^{4}Gy).TL fading measurements showed a remnant signal of 0.853 and 0.627 in the red and green bands respectively after 30 days of storage in dark at room temperature.

Keywords: Thermoluminescence; Sandstone; Glow Curve; Gamma Rays; Fading.

1. Introduction

Features sensed in the earlier data were, that the presence of the red signals was most obvious in quartz obtained from volcanic material. This complexity is explained by the variety of defects in sandstone that are either intrinsic (e.g., Si and O vacancies) or related to impurity atoms (e.g., Al or Ti). The concentration of impurity-related defects is dependent on the conditions of mineral formation or subsequent alteration. Experimental data have shown that the luminescence properties of sandstone are highly variable with geological source and vary even at a grain-to-grain level within sediment

The study of the TL characteristics of quartz extracted from burnt sandstones has allowed the approximate determination of the temperature to which they had been exposed and enabled their suitability for dating to be judged [1].Red thermoluminescence(RTL)of natural quartz grains offers many desirable properties for retrospective dosimetry, quaternary chronology and archeological dating. The RTL phenomenon was initially reported for quartz grains from dune sand [2].Subsequently RTL of quartz grains has been observed in volcanic products , burnt archaeological samples and Chinese loess ([3]and references therein).The thermal background was reduced in combination with a light guide , cluster heater, optical filters , and photomultiplier tube cooling to $-20^{o}C$ in the used system [4].

The lifetimes of trapped electrons and holes related to RTL emissions were confirmed to be longer than 1 Ma at ambient temperature which is long enough for archaeological dating [5].In luminescence dating and dosimetry studies, quartz RTL provide desirable properties for equivalent dose determination [6].Owing to the high thermal background, RTL measurements have been limited to relatively old samples, which accumulate doses of >50Gy [7].

The sensitivity of ferruginous sandstone samples as a function of the duration of annealing is not monotonic .For that reasons it seems necessary to consider the equivalent thermal history rather than an equivalent temperature [8]. Rendell et al. [9] reported on spectral changes induced in the TL of quartz as a result of a variety of thermal treatments. The rate of cooling changed the relative intensities of the component emission bands in all samples. In the volcanic quartz material, the ratio of red to blue emission is particularly sensitive to the cooling rate.

Red TL (RTL) of quartz grains has given higher activation energies than blue –TL (BTL) and a good response to absorbed doses [10].

Scholefield and Prescott [11] observed a parallelism between the blue and red emissions, in the sense that red TL peaks similar to the blue tended to appear at slightly higher temperatures. They concluded that the blue and red emissions probably involved a common set of electron traps feeding separate luminescence centers.

The red emissions of quartz has been associated with high amounts of aluminum impurity centers [12], Oxygen vacancy and sodium impurity [13] and Europium and / or Samarium light rare earth elements [14].

The suitability of sandstone from Egypt for the gamma radiation dosimetry using blue – TL(BTL) technique is investigated [15].Its properties are systematically studied utilizing measurements of natural and laboratory –induced blue TL emission band ,trap depths and storage effects . In this paper a preliminary study of the TL emission properties of the red (RTL) band of the same sample to explore the glow curves characteristics, gamma dose response, storage effects and comparison with green (GTL) and the previously measured blue band of the same sample.

2- Material and method

The employed natural sample of sandstone powder (particle diameter ≤ 45 used in this study was collected from Egypt. Information about the preparation and chemical composition of the sample is shown in Soliman and Salama [15].

To minimize the statistical error, five aliquots (5mg each) were used for each measurement.

The sample was irradiated at room temperature with γ-rays from a calibrated ^{60}Co source with doses in the range from 1Gy to 10^4Gy at King Saud University, Saudi Arabia .The time duration between irradiation and required TL readout was kept constant at 30min.

TL measurements were carried out using 3500 TLD reader equipped with two kinds of filter assemblies are employed to determine separately red or green TL intensity respectively and installed directly between the photomultiplier and the heater device, one is a narrow band green filter and a red glass filter (sharp-cut filter, R-60) combined with an infrared cut off filter (IRA-05).A linear rate of 5oC s^{-1} was chosen in a liquid nitrogen environment.

Glow and black –body curves were successively measured for each aliquot, then the black-body curve was subtracted from the glow curve.

TL fading was studied for 30 days at a γ -dose of 50 Gy at room temperature (RT).The exposed sample, as well as the control, was stored in the dark at RT and normal humidity conditions.

3. Experimental results

3.1 Natural sample glow curves

For investigation of the characteristic RTL and GTL glow curves of the natural sandstone sample was heated from room temperature up to 400°C and the TL intensity recorded. After-words a second readout was performed to record the background signal of the reader and sample .An average TL glow curve of the natural sample without any pre-annealing and irradiation treatments can be seen in Fig.1. One can easily see in the figure that the natural glow curves has one broad peak around 250°C and at 285°C in the red and green bands respectively and with intensity ratio of 1:10.In the previous studies the blue thermoluminescence of sandstone from Egypt exhibited a broad peak centered at 315°C [15]. The election source traps are different in the case of blue, red and green emissions and the electrons evicted from their traps recombine preferentially with the holes trapped in one or the other recombination center .When directly compared the TL intensity ratio is 28 : 10 : 1 for blue, green and red respectively . The previous studies of the natural sandstone indicated different glow curve peaks. The ferruginous sandstone from France [10] showed one peak at 110°C, TL glow curve from India showed one peak at ~240°C [16], while as the burnt sandstone glow curve exhibited two peaks at ~270°C and 350°C [1].Other sandstone from Brazil showed two peaks at 325°C and 375°C [17].

3.2 The effect of γ -irradiation

Before the irradiation, the sample was oven annealed (at 400°C for 30 min) in order to homogenize the impurities. As far as the detection bands are concerned there is a visible difference in the two detection bands .Figure 2 (a & b) shows typical glow curves for sandstone aliquots exposed to γ -ray dose of 50 Gy. At low doses starting at 1 Gy ,the glow curves of both bands show only one peak at 315°C.This peak is still appeared at high doses about 10^4 Gy .The irradiation added many modifications to glow curve of sandstone .At doses higher than 50 Gy ,TL peaks at 115°C, 135°C ,155°C ,195°C,285 °C and 315°C were appeared in red band and at 95°C ,135°C,285 °C and 315°C were appeared in green band.

The dissimilarity between natural sample glow curves (Fig.1) and the laboratory induced (Fig.2) can be ascribed to thermal treatment and ionizing radiation which induced new types of defects. Previously published results have shown that the blue emission band showed peaks at 88°C ,95°C , 210°C , 220°C ,and 305°C [15].This blue emission, lies lower in temperature than dose our results in green and red bands as in Fig.[2] .Sandstone sample from Japan showed that γ -irradiation induced two TL peaks at 150 °C and 350°C [18].

3.3 TL growth curves

The response of each integral TL, covering a temperature range from room temperature to 400°C, is examined for the absorbed doses by using the filter techniques. Figure 3(a &b) shows the dose response of red and green bands of sandstone over the dose range 1Gy-10^4 Gy .The TL dose response of red TL band follows linear relations $I=18.497+0.0041\ D$ and $I=11.678+0.203D$ with the correlation coefficients $R^2=0.987$ and 0.997 over the dose ranges 1Gy – 100Gy and 500Gy-10^4Gy respectively. Also the TL dose response of green TL band follows linear relations $I=28.31+7.618D$ and $I= 1880+0.704D$ with the correlation coefficients $R^2=0.998$ and 0.992 over the dose ranges 1 Gy – 100Gy and 500 Gy -10^4Gy respectively; where I refers to the intensity of the TL signal and D is the given dose. The evolution of the induced TL of blue band [15] with the dose over the range 1 Gy – 50 Gy showed a linear fitting (R^2 = 0.999) with an equation of the type $I = 135.62+5.82\ D$; followed by a sub-linear relation $I=74.3\ D^{0.61}$.

3.4 Thermoluminescence Fading

It is important to know the stabilities of the traps connected with the peaks ; since these reflect the storage capacities of the traps .To determine the stabilities ,the thermoluminescence measurements were performed for a period of 30 days (d) in the room environment (20°C -25 °C).Prior to storage the investigated material was given a standard annealing treatment at 400°C for 30 min .The large amount of annealed powder sandstone was irradiated (50 Gy) and stored in dark before TL reading . Another amount of the annealed sample used as the control being stored for a period of 30 days in the same conditions, then irradiated with the same dose and read out immediately on the same day to avoid variations from instrumental drift. Glow curves of the red and green bands are shown in Fig.4 (a&b) after a delay period of t= 14d ,and without any delay period (t=0).The differences in the glow curves with t=0 d and t=14 d result from the migration of the low temperature trapping centers to the high complex centers .Figure 5 (a&b) shows the time dependent TL fading in the period 0d – 30 d .It is shown in TL signal felt to 85 % and 63 % of its original value in red and green bands respectively.

4. Conclusion

The results indicate the complexity of the luminescence mechanisms in sandstone. The natural glow curves of red, green and blue [15] bands show different natural electron traps responsible for the three emissions.

The laboratory induced glow curves of the red and green bands involved same electron traps at 135 °C and 315 °C and other different traps .The results may be explained by that there are more than one zone in the crystal containing populations of different holes responsible for the three emissions .In each zone type ,there is different concentrations of electron traps of similar and different types .If the zones are large composed to the size of crystal cell , than it may be possible that the evicted electrons preferably recombine in the zone from which they originate.

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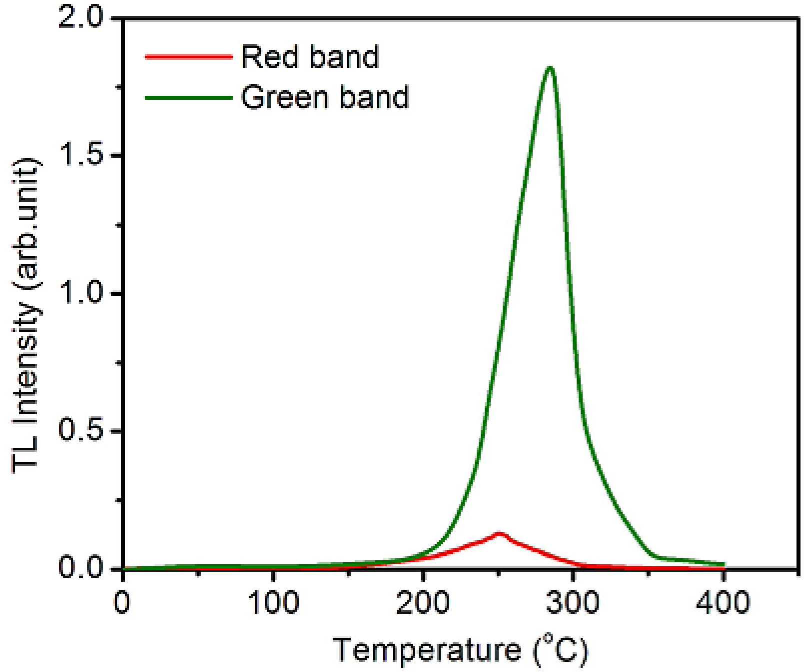
2.0
1.5
1.0
0.5
0.0
Red band
Green band
TL Intensity (arb.unit)
0
100
200
300
400
Temperature (°C)

Fig.1

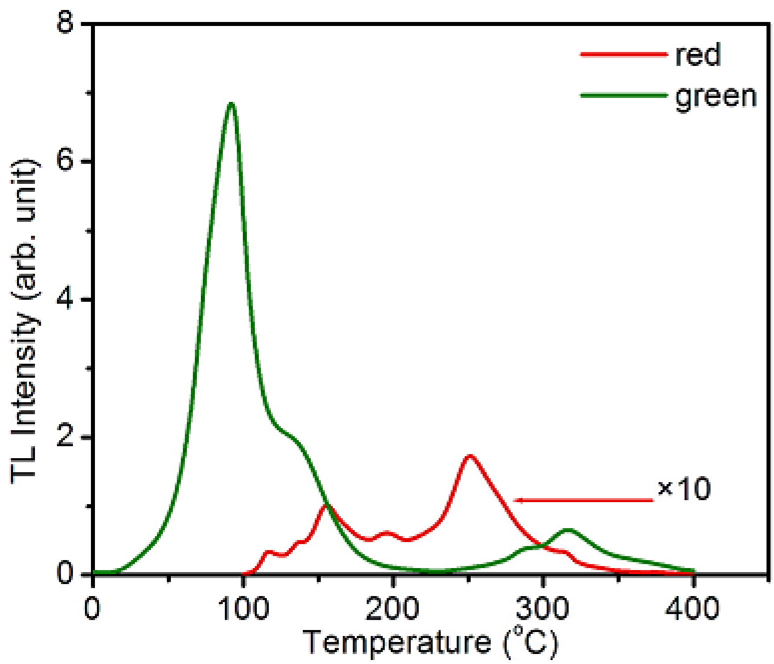
8
6
4
2
0
red
green
×10
TL Intensity (arb. unit)
0
100
200
300
400
Temperature (°C)

Fig.2

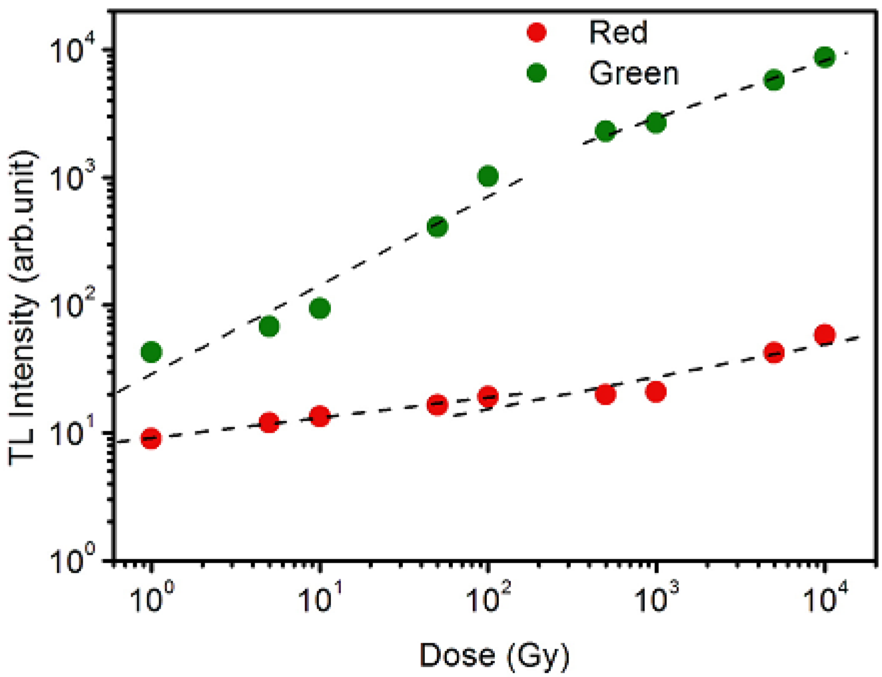
Red
Green
TL Intensity (arb.unit)
Dose (Gy)

Fig.3

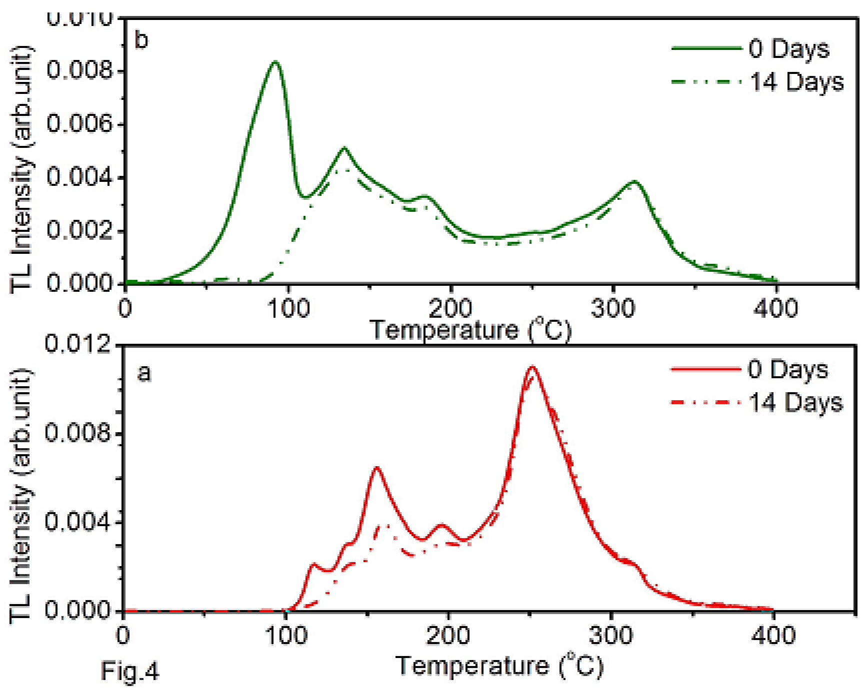
b
0 Days
14 Days
TL Intensity (arb.unit)
Temperature (°C)
a
0 Days
14 Days
TL Intensity (arb.unit)
Temperature (°C)

Fig.4

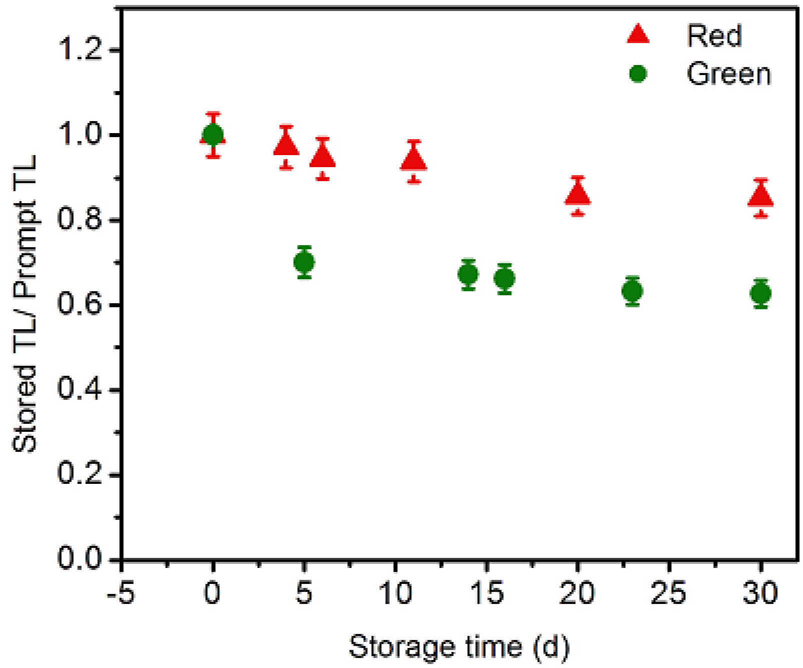
Red
Green
Stored TL/ Prompt TL
Storage time (d)
1.2
1.0
0.8
0.6
0.4
0.2
0.0
-5
0
5
10
15
20
25
30

Fig 5