

The Possibility to Use Natural Material as a Measure of Ionizing Radiation

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Abstract

The suitability of gabbro for the gamma (γ) radiation dosimetry using thermo-luminescence (TL) technique has been investigated. Its properties are systematically investigated utilizing measurements of natural and laboratory induced blue TL emission band, annealing effects, and storage effects. The emission of the natural blue band exhibited broad peak at $\sim 342^\circ\text{C}$. The TL- γ dose response has a linear behavior over the dose range 0.1Gy-10Gy followed by a supralinear one at high dose level 100Gy-700Gy. The fading in TL signal over two weeks of storage period at room temperature was not more than 41%. From our obtained results it is possible to conclude that the natural gabbro is a suitable material for accident and environmental dosimetric applications.

Keywords: Thermoluminescence; Gabbro; Gamma; Fading.

1. Introduction

Thermoluminescence (TL) dosimetry has come of age that it now represents a key technique in absorbed dose measurement [1]. Presently because of its place in environmental, personal and clinical applications, efforts are geared towards producing new and highly performing TL materials. Thermoluminescence spectra of feldspar in pegmatite form south china were experimentally investigated using a newly-constructed three-dimensional thermoluminescence spectrometer, under variant experimental conditions: the storage time period between sample irradiation and spectral measurements, and the dose rate used for sample irradiation. The experimental results show that (1) the feldspar TL spectra display in a pattern of wavelength bands, centered on eight different distinguishable wavelengths; (2) the thermoluminescence components of feldspar from pegmatite fade away heterogeneously depending on wavelength and thermal stimulated temperature; (3) the two different dose rates used for artificial irradiation result in distinguishable diverse thermoluminescence spectra. All these luminescence spectral characterization features suggest that more complicated ingredients influencing feldspar luminescence dating need to be ruled out than expected. Especially it is necessary to correct the normal luminescence dating results in regard to these influential factors when feldspars extracted from sediments but originating from rocks are used for luminescence dating. On the other hand the experimental results also suggest the feasibility of optimization of feldspar luminescence dating [2].

The thermoluminescence (TL) characteristics of quartz are highly dependent of its thermal history. Base on the enhancement of quartz crystals. However, little is known about the influence of the temperature of quartz crystallization on its TL characteristics. The TL sensitivity and dose-response curves of hydrothermal and metamorphic quartz were evaluated with crystallization temperatures from $209 \pm 15^\circ\text{C}$ to $633 \pm 27^\circ\text{C}$ determined through fluid inclusion and mineral chemistry analysis. The studied crystals present a cooling thermal history. Which allow the acquiring of their natural TL without influence of heating after crystallization. The TL curves of the studied samples present two main components formed by different peaks overlapped around 110°C and $200\text{-}400^\circ\text{C}$. The TL sensitivity in the $200\text{-}400^\circ\text{C}$ region increased linearly with the temperature of quartz crystallization. No relationship was observed between temperatures of quartz crystallization and saturation doses ($<100\text{ Gy}$). The elevated TL sensitivity of the high temperature quartz is attributed to the control exerted by the temperature of crystallization on the substitution of Si^{4+} by ions such as Al^{3+} and Ti^{4+} , which produce defects responsible for luminescence phenomena. The linear relationship observed between TL and crystallization temperature has potential use as a quartz geothermometer. The relative abundance of quartz in the earth crust and the easiness to measure TL are advantageous in relation to geothermometry methods based on chemistry of other mineral [3].

Topaz shows thermoluminescence (TL) on heating after exposure to ionizing radiation. A study was carried out to explore the possibility to design and develop a TLD (thermoluminescence dosimeter) from locally occurring topaz by neutron irradiation, X-ray diffraction and X-ray fluorescence. The crystals of topaz were cut into chips of dimensions compatible to the commercial TLD readers. The investigation features included glow curve, dose and energy responses, sensitivity, fading, reproducibility and mechanical stability. The TL glow curves revealed

a stable peak at about 250 °C, whose height rose linearly with increase of irradiation dose. The TL response versus dose (calibration curve) showed the linear behavior between 10^{-2} and 10^2 Gy. The dose response was independent of gamma energies of ^{60}Co and ^{137}Cs . The response topaz chips remained constant within 10% deviation from the initial value after 30 cycles of reuse. The rate fading of topaz was very fast just after irradiation and slowed after a few days. Mechanical stability of the chips remained constant during handling in all investigation experiments. The topaz from Yono Shigar mine may be recommended as a TLD for gamma dose within 10^{-2} - 10^2 Gy [4].

The Thermally Stimulated Luminescence (TSL) at room temperature X-ray irradiated natural biotite in form of micro-grain powder was studied under various rates. TSL peaks showed at temperatures 393.6 K, 403.5 K, 404.5 K, 406.9 K at their respective heating rates 2 K/s, 4 K/s, 6 K/s and 10 K/s. The effect of thermal quenching on thermoluminescence parameters such as peak maximum temperature, peak area, FWHM, geometrical symmetry factor, the activation energy were investigated. From the symmetry factor it is clear that the TL glow curve follows the first order kinetics for the lowest heating rate, but as the heating rate increases it defers from the first order. When activation energy is calculated by variable heating rate method it is observed that the method overestimated the value of activation energy and pre-exponential frequency factor significantly due to thermal quenching [5].

Thermoluminescence spectra of manganiferous carbonate has been studied from 30°C to 400°C over the wavelength range from 200 nm to 800 nm. The natural thermoluminescence shows continuous spectral distribution from 240 nm to 800 nm. Above 500 nm the emission appears in the form of line structure emission. The emission features are attributed to presence of Mn^{2+} impurities in the carbonate matrix. The spectral study was supplemented by kinetic analysis of the most prominent peaks and their kinetic features are reported [6].

Thermoluminescence (TL) of natural light-orange color calcite (CaCO_3) mineral in micro-grain powder form was studied at room temperature X-ray and UV irradiation under various irradiation times. TL was recorded in linear heating rate (2 K/s) from room temperature (300 k) to 523 k. Due to thermally assisted tunneling of electron and subsequent center-to-center recombination, a distinct peak of lower activation energy (0.60 eV) was observed at relatively higher temperature (~360 K) for X-ray irradiated sample. In UV excitation, there was an indication of photo-transfer phenomenon, where low TL intensity might have been observed; but due to simultaneous excitation of electrons from valence band to the trap level, TL intensity was found to increase with UV irradiation time. The results obtained within temperature range 300-523 K were explained by a band diagram [7].

The dosimetric characteristics of some natural hydrothermal quartz (NHQ) samples were investigated by using the thermoluminescence (TL) technique for the purpose of determining whether they are suitable as dosimetric materials or not and calculating the kinetic parameters. The obtained results showed that the trap depths and the frequency factor values are consistent with the literature. The studied samples have linear dose responses for the absorbed doses ranging between 6.689 Gy and 301 Gy. In conclusion, the examined quartz samples can be used as dosimetric materials in high dose applications [8].

In spite of numerous previous studies, no work was performed on igneous rocks from Saudi Arabia. Therefore, some TL characteristics of the blue emission band of natural gabbro are studied in the present work. Four types of measurements were used:

- (i) The glow curves of the sample before and after irradiation with γ -rays.
- (ii) The effect of pre-irradiation annealing.
- (iii) The TL gamma dose response.
- (iv) The fading effects.

2. Material and Methods

2.1 The material

The employed natural sample of gabbro powder (particle size $75\ \mu\text{m}$) used in this study was collected by the geology group in the Department of Geology, Faculty of Science, King Saud University. Gabbro is deep igneous rock (Which is shaped in the core of the earth), therefore, its granules are hard and rough in a way that could be seen by the naked eye.

It is formed of Plagioclase and Pyroxene, in addition to other metals sometimes such as Apatite, Magnetite and Garnet, which is gray in color.

It is found in KSA within among the base rocks, which are the oldest igneous rock from the Precambrian Era, i.e., more than 400 Million years, and it is found within a rocks called the Arabian Shield, and is originated from the city of Al-Qwaieiah about 200 Km far form Riyadh towards Mecca, and these rocks contains accompanied significant economic metals, which are used in industries such as constructions, roads paving, Cement mixes and others.

To minimize the statistical error five aliquots each of 7mg were used for each measurement.

2.2 The source of excitation

The sample was irradiated at room temperature with γ -rays from a calibrated ^{60}Co source. The time duration between irradiation and required TL readout was always kept constant at about 30 min.

2.3 TL recording system

Harshaw3500 TLD reader was the used apparatus in this study. This equipment is divided into a heating and a photon-counting unit. Light pulses were detected with the photomultiplier tube and a narrow band glass filter with a blue-violet transmittance band. A linear heating rate of 5°C s^{-1} was chosen; heating the sample from the room temperature up to 400°C . The incandescent background was measured to be subtracted from the TL data.

3. Results and Discussion

3.1 Natural sample glow curve

The TL glow curve of the blue emission band of natural gabbro exhibited a broad peak centered about 342°C (Figure 1a). We attributed this natural peak to irradiation from a number of sources which include self irradiation, irradiation from surrounding geological formations, ultraviolet and cosmic sources. The broadness of this peak could be as a result of the existence of closely spaced trapping centers for which individual glow peaks could not be resolved, which indicates a complex trapping system. Choudhury et al. [9] attributed this peak to ionic motion. As observed in Figure1(b-e), gabbro appears as a very sensitive material for both non- ionizing radiation and ionizing radiation (curves b-e). In addition, one can appreciate how the shape of both TL emissions is quite different depending on the type of the irradiation source used.

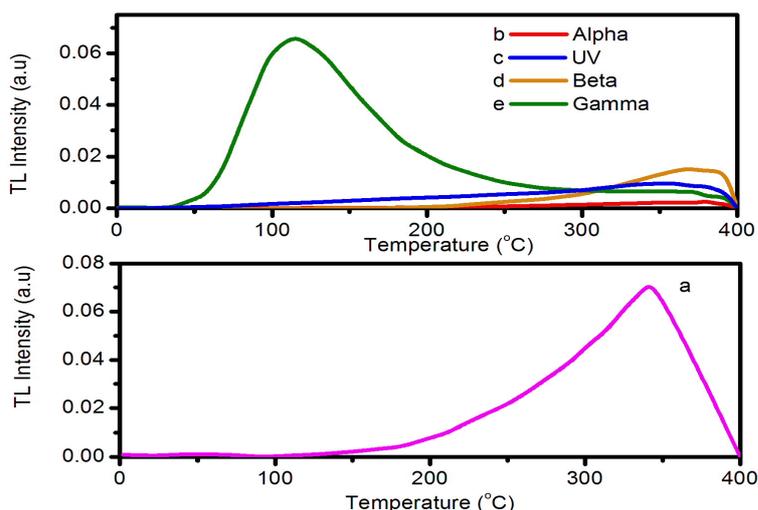


Figure (1) Thermoluminescence of blue emission band: (a) natural gabbro; curves (b-e) after exposed to alpha, ultraviolet, beta and gamma radiation

3.2. Pre-irradiation annealing

Before irradiation, gabbro requires a thermal treatment to clear the dosimetry traps. For this study a series of natural aliquots were used. Each was heated at a certain temperature in the range from 250°C to 700°C ($t = 15$ min) and when cooled down γ dose of 50Gy is given and TL was measured. After 30 min of irradiation the TL glow curves were read out. Important modifications in the shape of the glow curves are observed Figure (2).

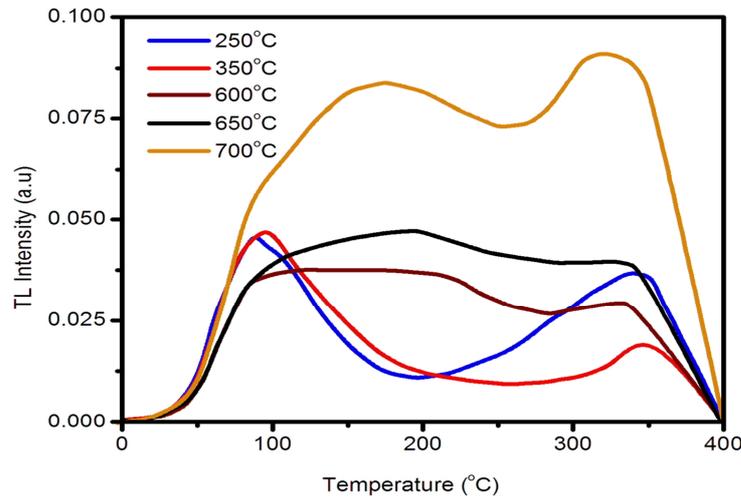


Figure (2) Glow curves of gabbro annealed at various temperatures

Treated aliquots showed two main TL peaks. The first one appeared at 89°C after annealing at 250°C and shifted to high temperature $\sim 175^\circ\text{C}$ after annealing at 700°C. The second one appeared at 353°C after annealing at 250°C and shifted to lower temperature at $\sim 324^\circ\text{C}$ after annealing at 700°C (Figure 3). A gradual decrease in TL was observed as the sample was heated from 250°C to 400°C. The TL signal is nearly constant over the temperature range 350°C to 550°C, after that it increases (Figure 4). The reduction of the TL intensity at temperatures 250°C - 400°C may be attributed to thermal quenching [10]. The sensitivity of gabbro annealed at 700°C is found to be 4 times that annealed at 400°C.

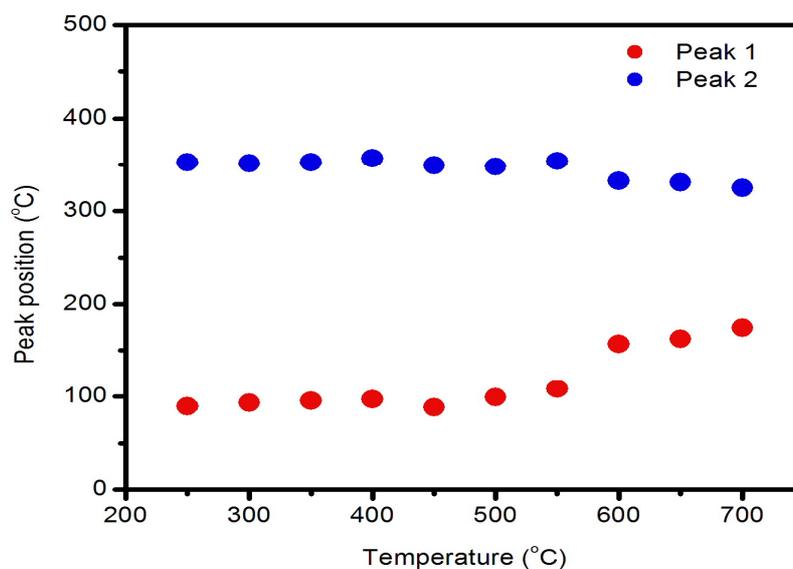


Figure (3) The effect of the annealing on the peak temperature.

This sensitizing effect of thermoluminescence may be due to heat- induced changes in the population of radiation induced defects or in the diffusion of impurities to the lattice, or interstitials sites at which they constitute effective trapping centers [11, 12].

3.3. The effect of γ irradiation

Figure 5 shows some selected glow curves for annealed (at 450 °C for 30 min) and subsequently irradiated gabbro at room temperature. One broad TL peak at ~140°C was obtained by the irradiated gabbro sample after exposed to a γ -dose of 10Gy. This means that (1) the traps related to the peak at 140 °C were filled at first because their traps were all emptied . (2) The capture cross section of the carriers in the traps associated to this peak is much higher than the corresponding value for other peaks. This peak shifted to a higher temperature at 192°C with increase of irradiation dose of 1.5kGy. A new peak was observed at 360°C after exposure of 50Gy then shifted to 275°C at higher doses.

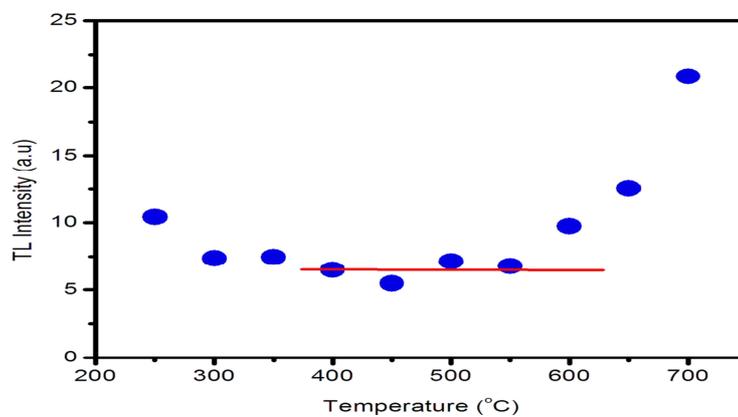


Figure (4) TL intensity of gabbro after annealing at different temperatures

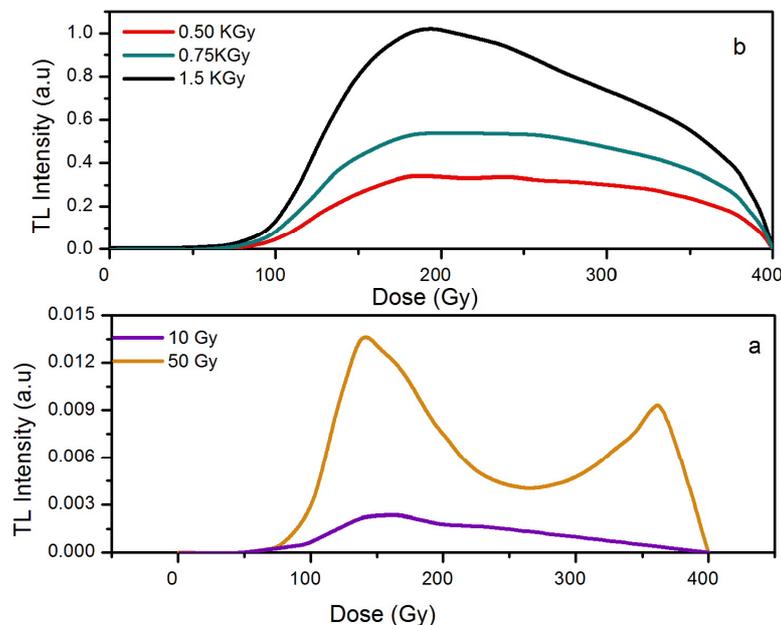


Figure (5) TL glow curves for gabbro at various γ - doses:
 (a) low doses and (b) high doses

3.4. Growth curve

Figure (6) shows the response of the TL as a function of γ dose. The TL- γ dose response has a linear behavior $I = 0.0125D + 0.9101$ over the dose range 0.1Gy-10Gy; followed by a supralinear $I = 0.0032D^{1.5472}$ at high dose level 100Gy-700Gy where I refer to the intensity of the TL signal and D is the given dose in Gy. Using the fitted equations, the obtained TL intensity of the natural un-irradiated sample corresponds to a γ -dose of ~ 149.61 Gy. By invoking the defect interaction model [13] in which the defects interact with each other during the heating stage via the process of charge transfer one can explain the above supralinear behavior. If there are no competitors, no supra-linearity will result. While the linear response at low doses may be obtained by a spatial association between the trap and the recombination site.

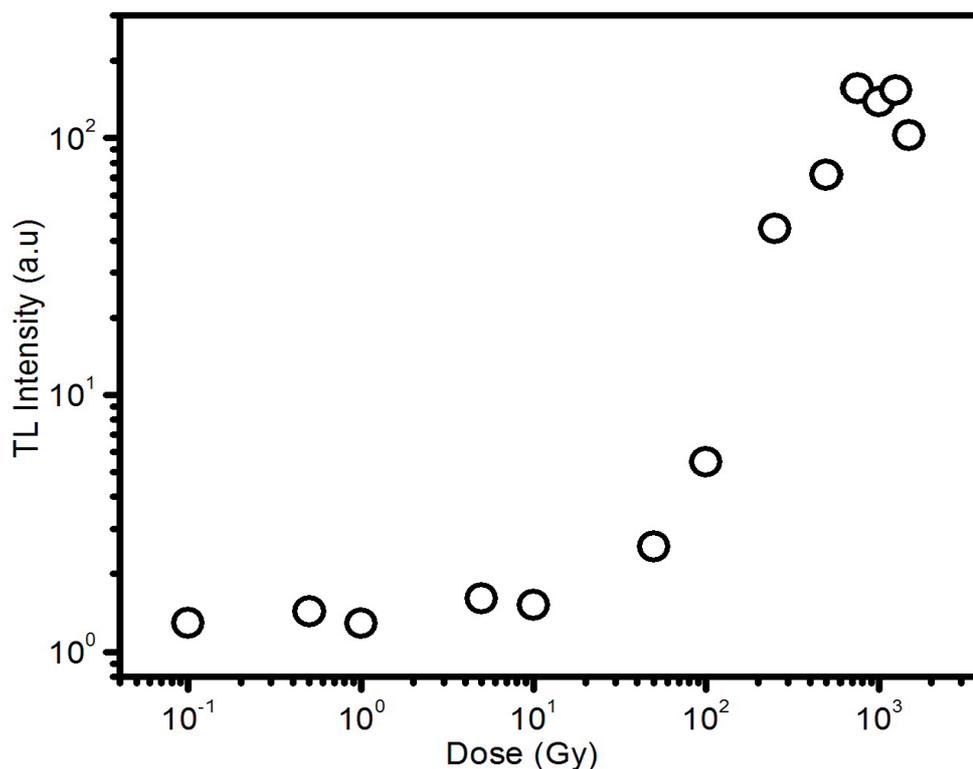


Figure (6) Gamma dose vs. TL response of gabbro.

3.5 Storage effects

The TL fading of laboratory induced TL signal (50Gy of ^{60}Co gamma dose) given to annealed aliquots (at 450°C in air for $t = 30$ min) is studied over a period of 14 d. The large amount of annealed powder gabbro was irradiated (50Gy) and stored for a period of 14 d in dark at room temperature before the TL reading, the small amount of it used as the control, being stored first, then irradiated with a dose identical to that above and read out immediately on the same day to avoid variations from instrumental drift. Glow curves of the blue-band, often the delay period ($t=14$ d) and without a delay period ($t = 0$ d) are shown in Figure (7).

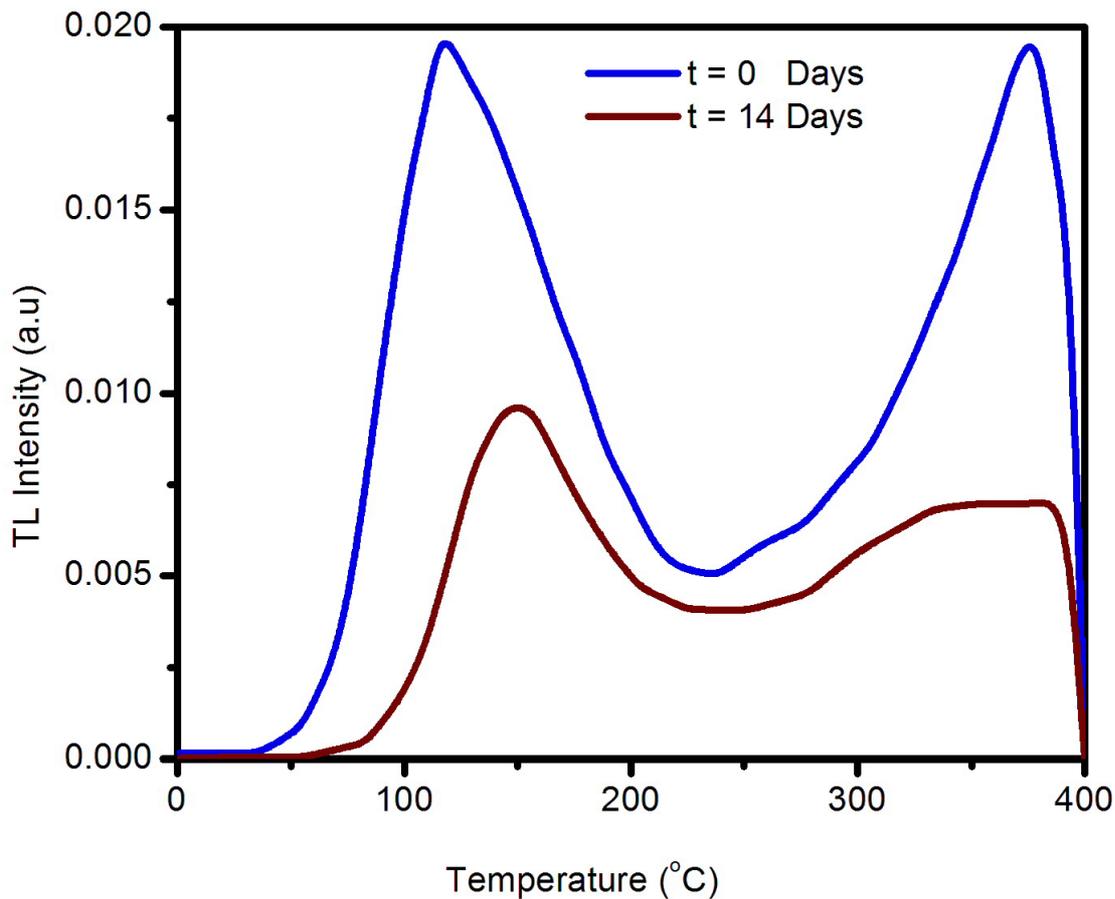


Figure (7) The effect of room temperature dark storage on the laboratory - induced glow curves of gabbro.

The stable TL signal was found to be (65%) after a delay period of 1d and to settle down a constant value of 41% of the initial value after 7 days of storage Figure (8).

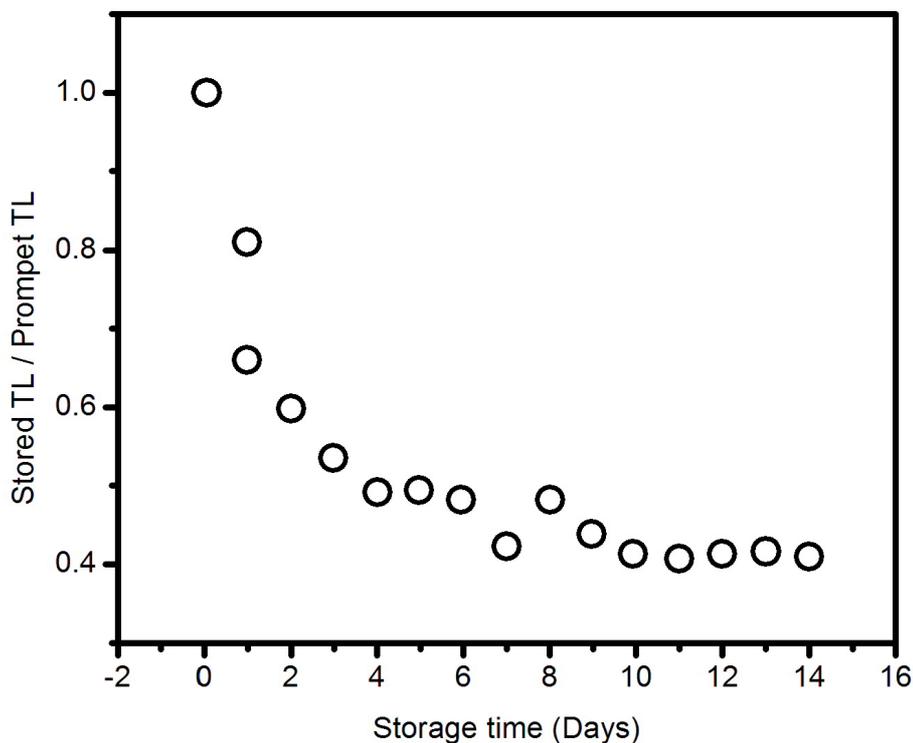


Figure (8) TL fading of gabbro at room temperature.

Conclusion

The TL characteristics of gabbro blue emission band have been studied in the present paper considering its usage as a γ radiation dosimeter in accident and industrial applications, the most notable features are:-

- (1) Very small weight can be used for TL measurements (7 mg).
- (2) The blue emission in the range of photomultiplier sensitivity.
- (3) A simple glow curve.
- (4) The natural blue TL emission band is strong and corresponds to γ irradiated sample with a dose of ~149.61Gy.
- (5) The dose response curve is linear over the dose range 0.1 – 10 Gy.
- (6) Water tight.

Acknowledgements

I would like to give a big thanks for everyone who help me with this research.

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