

Phototransferred Thermoluminescence from Obsidian Using Ultraviolet Radiation

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Abstract

The behavior of TL and PTTL under UV stimulation in gamma-irradiated obsidian was investigated. The noticeable change of the shape of the main TL peak after thermal or optical stimulation clearly shows that this peak is related to at least two traps. The PTTL curves recorded after preheating up to a given increased end temperatures are described by two processes. The first one is associated to the decay of the traps responsible for the main TL peak. The second is related with the phototransfer from the deep to the main trap. Fading studies over a period of 14 days show that the PTTL faded by 5% when exposed to 50Gy. The results further predict the potential of obsidian as a phosphor in TL dating, accident and industrial dosimetry.

Keywords: Obsidian, Phototransfer, Phosphor, Fading.

1. Introduction

Photo-transferred thermoluminescence (PTTL) is the phenomenon of optically stimulated transfer of electrons from deep traps into shallow traps, yielding subsequent thermoluminescence (TL) peaks depending on light source intensity, wavelength and illumination time. It is observed in many natural and artificial materials and it was proposed as useful tool for dosimetry and dating applications (Bailiff et al. (1977)). A part from the qualitative interpretation of the PTTL, coherent with the general theory describing other related luminescence phenomena, such as TL and optically stimulated luminescence (OSL), for many materials an exhaustive explanation of the PTTL processes is nowadays not yet available.

The study of PTTL is one of the sources of information in understanding the luminescence processes in the material of interest. The precise nature of the processes which take place in irradiated natural material as it is exposed to light of different wavelengths is not known, and the experimental data may be interpreted in several ways. Some studies of the OSL, PTTL and TL of Arkansas quartz by McKeever and Morris (1994) led McKeever (1994) to explain the decay of luminescence signals in terms of a loss of radiative recombination centers. In his model there are minimum of two recombination centers, only one of which is observed in the luminescence measurements because of the choice of optical detection region. The numerical ratio of these centers can be chosen such that there are a relatively small number of the luminescence centers available, compared with the non-luminescence centers. Using a computer simulation (McKeever and Morris (1994)) qualitative similarity was demonstrated in the form of the decay curve from their numerical model and that obtained from their sample. In their model, electrons are optically stimulated from a deep, thermally disconnected trap and some recombine at luminescence centers, thus reducing the number of these centers subsequently available for recombination with electrons released from shallower traps. Some electrons do not recombine after optical stimulation, but are transferred to shallower traps from which they can be thermally ejected to give PTTL. According to this model PTTL will decay with time under optical stimulation because of the reduction in the number of available luminescence centers; optical stimulation from the shallow trap giving rise to PTTL (or TL) is not allowed in their model.

In some cases it is necessary to reassess dose and check measurement correctness. This is especially required in personal dosimetry to ensure reliable and accurate results. Applying UV radiation to already read TL pellets causes relocation of electrons from deep traps to the dosimeter traps, which effect is called the phototransferred thermoluminescence (Alexander and McKeever (1998)).

In literature the photoluminescence of a variety of natural materials are reported. For example in analytical grade quartz the dependence of the PTTL intensity on the illumination time suggested that recombination processes involved a non-radiative recombination center (Bertucci et al. (2011)). The dependence of TL signals of natural quartz on the heating temperature and UV illumination enables us to propose plausible phototransfer mechanisms at low temperatures ~ -100 °C (Santos and de Lima (2001)). Bruce et al (1999) studied bleaching and phototransfer properties of limestone. Various optical and dosimetric properties of zircon were investigated using TL and PTTL (Kristianpoller et al. (2006)). The sensitivities of TL and PTTL were studied in natural fluorite (Kharita et al. (1995)).

In spite of numerous previous PTTL studies, no work was performed on obsidian. Obsidian is a natural

glass and is formed from volcanic lava which cooled too fast for significant crystallization to happen. It is used in semiprecious, weapons, surgical instrument, decorative specimens, gemstones, crystal healing and other applications. Therefore the aim of the present work is to obtain insight into some TL characteristics of the blue emission band of natural obsidian and PTTL mechanisms in gamma-irradiated aliquots. Four types of TL measurements were used:

- (i) the glow curves of the sample before and after irradiation with UV and γ -rays;
- (ii) the TL γ dose response;
- (iii) the effect of UV phototransfer; and
- (iv) the fading effects, as monitored for a period of 14 days.

2. Experimental Conditions

The chiefly investigated sample was natural obsidian (particle size 100 μm) provided by Astronomy and Geophysics Research Institute, King Abdul-Aziz City for Science and Technology (KACST) in Riyadh and originated from Harrat Khaybar area, in the western region of Saudi Arabia. It is composed of SiO_2 (75%), MgO (20%) other impurities (5%) and is low in water. For studying the effect of the pre-irradiation annealing, aliquots of obsidian were heated in quartz bowls at ambient air at a certain temperature in the range from 100 $^{\circ}\text{C}$ to 700 $^{\circ}\text{C}$. The temperature was controlled within 5 $^{\circ}\text{C}$ from the desired setting. At the end of the annealing time, the aliquots were allowed to cool rapidly, within a few minutes, to room temperature and were kept in dark until the TL measurements were performed. The UV source used in the work was a low-pressure mercury lamp (mainly emitting a wavelength of 254 nm at a power density of $1\mu\text{W}/\text{cm}^2$). The γ -irradiation was performed with a ^{60}Co source. Irradiations and TL measurements were performed in King Saud University, Saudi Arabia.

TL measurements were recorded with a Harshaw 3500 TLD apparatus with light pulses were detected by the photomultiplier tube provided with a narrow band blue filter plus Schott BG39 glass filters of blue- violet transmittance band. A linear heating rate of 5 $^{\circ}\text{C s}^{-1}$ was chosen. Each TL reading is an average of five readings. All TL readout data were acquired using a time temperature profile running linearly from room temperature to 400 $^{\circ}\text{C}$. The second reading with the same heating profile is considered to be the black body radiation and was subtracted from the first one.

3. Results and Discussion

3.1 Natural Sample TL Glow Curve

The glow curves of the natural sample (without any treatment in the laboratory) recording in the blue band, exhibited two peaks at 280 $^{\circ}\text{C}$ and 375 $^{\circ}\text{C}$ (Figure1). These peaks are attributed to irradiation from a number of sources which include self-irradiation, irradiation from surrounding geological formations prior to sampling and from cosmic sources. The broadness of this peak could be a result of the existence of closely spaced trapping centers for which individual glow peaks could not be resolved. This indicates a complex trapping system in the investigated material. On annealing the sample at 400 $^{\circ}\text{C}$ for 30 min, and cooling in air, TL readouts showed no peaks at all. This indicates a total cleaning of any stored TL signal. Three natural TL peaks were investigated at $\sim 200^{\circ}\text{C}$, $\sim 280^{\circ}\text{C}$ and $\sim 400^{\circ}\text{C}$ in obsidian from Northwest America (Huntley and Bailey (1978)). Obsidian from New Zealand showed one natural TL peak at $\sim 280^{\circ}\text{C}$ (Foss and Fankhauser (1978)).

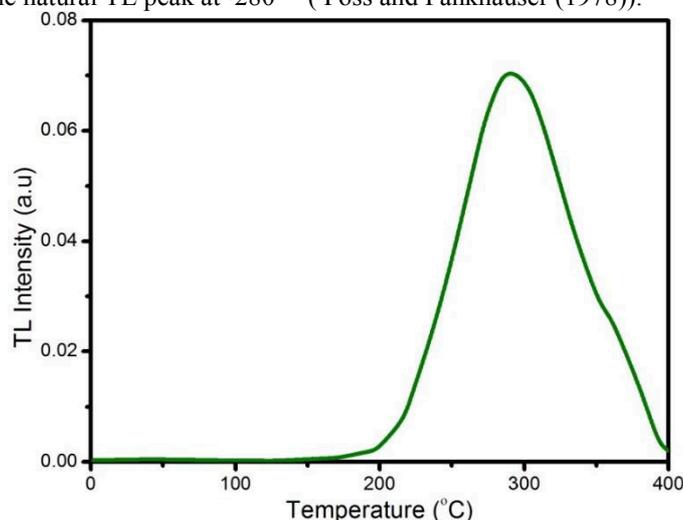


Figure. 1 The glow curve of natural obsidian recorded at a heating rate of 5 $^{\circ}\text{C s}^{-1}$.

3.2 Bleaching Effect

The dependence of the TL response of obsidian, using UV irradiation on the exposure time was studied from 10 to 80 min and is shown in Figure 2.

The observed results show that the TL intensity increases with the exposure time until 30 min. The behavior exhibited by obsidian can be explained by Alexander and McKeever (1998) using rate equation analysis. The explanation lies in the competition between radiative and non-radiative recombination centers in which the PTTL is seen to decrease at a certain illumination time due to the removal of holes, by recombination, from the luminescence centers. An initial increase, followed by a decrease, followed by a constant PTTL level is predicted from this model.

Typical glow curves of obsidian aliquots exposed to UV radiation at room temperature (RT) during different time intervals are shown in Figure3. At low doses starting at 10 min, the glow curves show two peaks at 310°C and 375°C . This means that:

- (1) The traps related to the high temperature peaks were filled at first because their traps were emptied.
- (2) The capture cross section of the carriers in the traps associated to these peaks is much higher than the corresponding value for the other peaks. The effect of bleaching is appeared as an absence of low temperature at 280°C and shift to higher temperature.

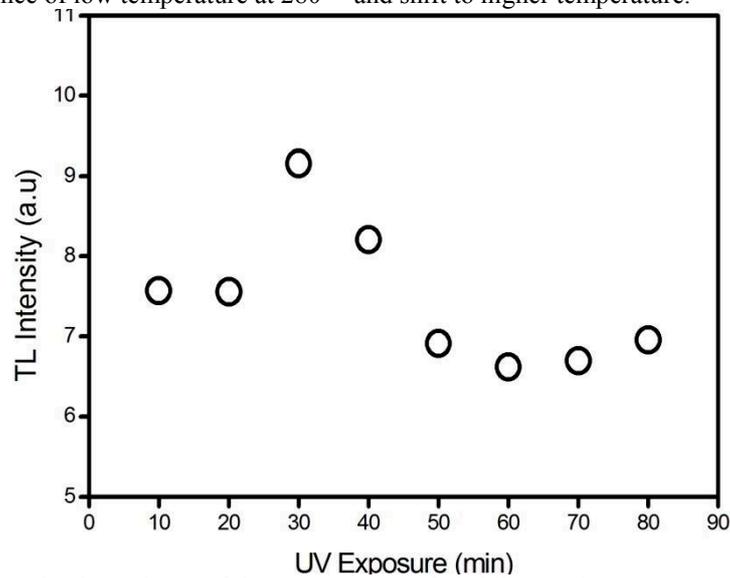


Figure 2. The dependence of the TL response of obsidian on the UV exposure time.

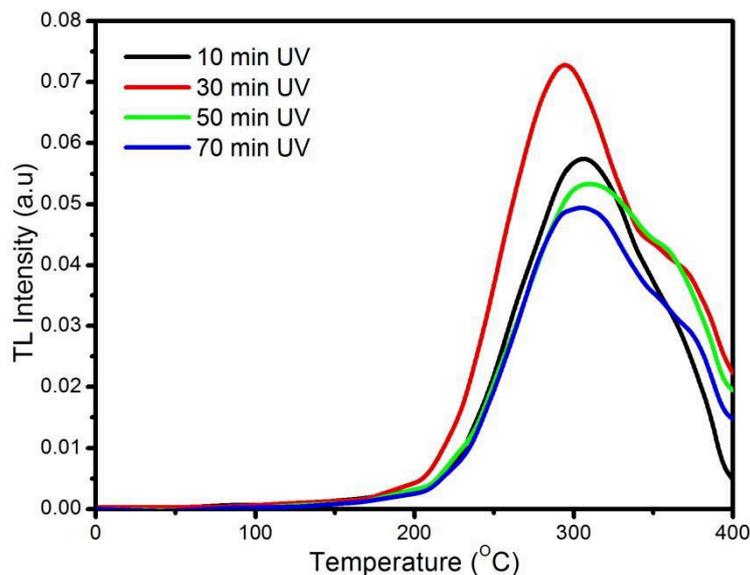


Figure 3. TL glow curves of obsidian exposed to UV light at 0°C during different time intervals.

3.3 TL Gamma Dose Response

The investigated material was pre-heated at different temperatures ranging from 100°C to 700°C for 30 min, cooled in air and subsequently irradiated up to a γ -dose of 50Gy. The main effect of this thermal treatment is decrease of the TL signal with increasing temperature. The sensitivity of obsidian annealed at 300°C for 30 min. is found to be minimum, which means emptying of the acceptor traps (Figure 4). Figure 5 reveals the presence of the main peak at 310°C after irradiation with a dose of 0.1Gy. With a further increase in radiation dose, this peak shifted to higher temperature.

3.4 Thermoluminescence Phototransfer

In order to determine without ambiguity the donor traps, the following procedure was employed:

- (1) Obsidian sample is first γ irradiated at RT.
- (2) Aliquots are then annealed up to various values of temperatures (100°C - 700°C): during this operation, the various traps are selectively emptied.
- (3) After return to RT and exposure to UV radiation (30 min at 0°C) the PTTL emission is registered.

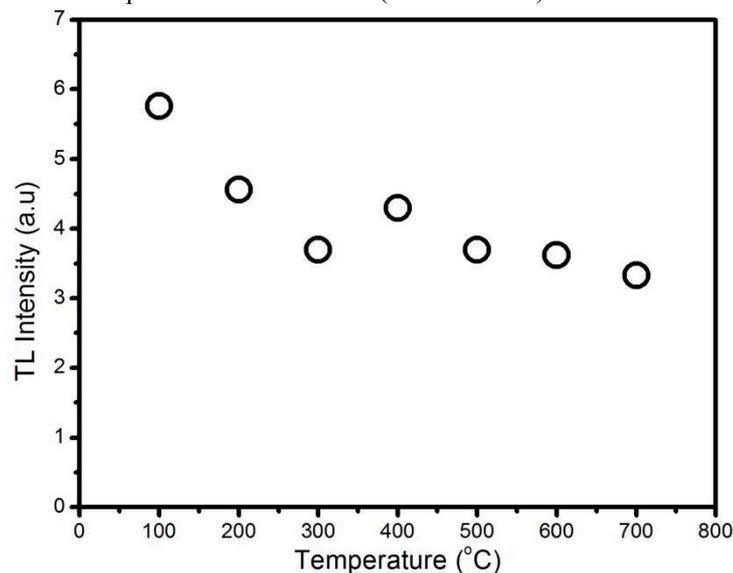


Figure 4. The variation of the γ response of obsidian with the pre-heating temperature.

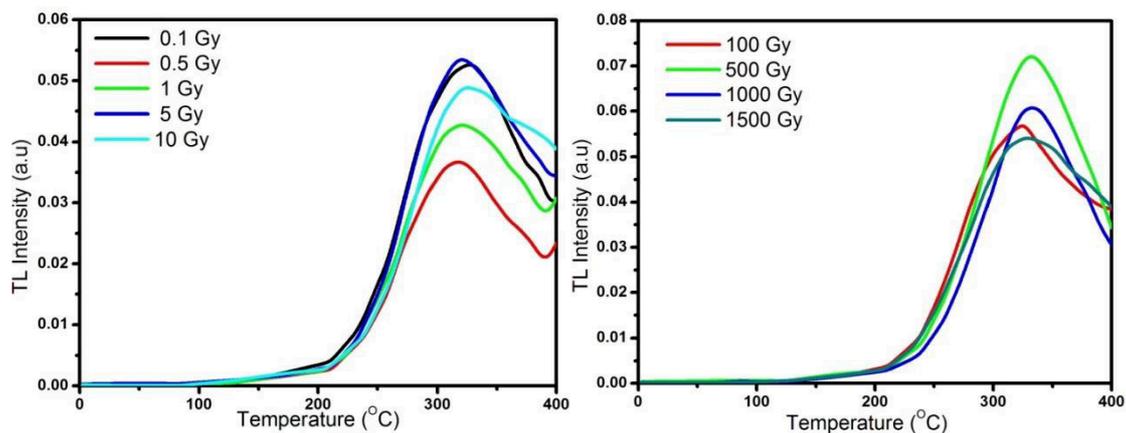


Figure 5. Variation of glow curves of obsidian with the gamma radiation dose.

When the sample is cooled and exposed to UV light, some of the centers can be depopulated and the holes are transferred to a shallower hole center that generates, during the heating up of the sample, a low temperature peak at 75°C is observed (Figure6).

For higher preheating temperature the main peak at 310°C is decreased in intensity. The noticeable change of the shape of the main TL peak after thermal or optical stimulation clearly shows that this peak is related to at least two traps at 75°C and 310°C and the PTTL curves changed drastically.

At the beginning of excitation the PTLL peak intensity (75°C) is low; afterward it increases with

annealing temperatures up to 300°C. As follows from the data presented in Figure 7, the PTTL destroys the main TL peak (preheating $\leq 300^\circ\text{C}$). Since the rate of destruction of the main TL peak is higher than the rate of its restoration the effect of UV will depend on the initial intensity of this peak. If the main peak is destroyed fully (curve preheating at 300°C, the readout will restore it due to phototransfer (Correcher et al.(2004)). A similar mechanism was suggested by Al-Khalifa et al. (1987) to explain the phototransfer of charges.

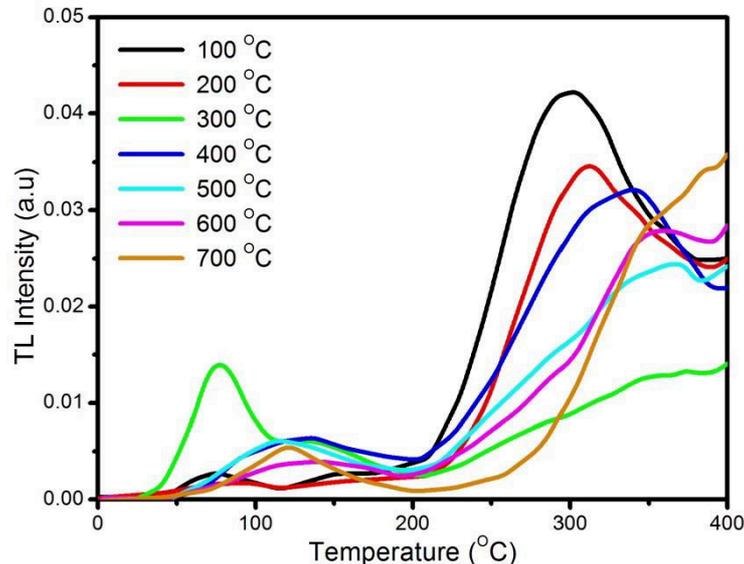


Figure 6. Typical phototransfer thermoluminescence glow curves of obsidian.

Figure 8 represents PTTL from obsidian as a function of gamma dose. The PTTL intensities were obtained by integrating the glow curves in the region from room temperature up to 400°C. The linear behavior ($Y = 5.160 + 0.692X$) of the PTTL, where Y represents the PTTL signal, X represents the γ dose. This implies that PTTL in obsidian can be used for γ dose reassessment over the dose range from 0.1Gy to 1000Gy.

3.5 Storage Effects

The use of the PTTL phenomenon allows us to transfer the TL signal from the deeper traps to the classical dosimetric traps and, thus, to read this information with the normal reading apparatus.

Figure 9 shows the TL fading of laboratory induced PTTL signal (50Gy of ^{60}Co gamma dose, annealing at 400°C in air for $t = 30$ min, exposed to an UV for 30 min at 0°C).

The large amount of treated powder obsidian was 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 treated as above and read out immediately on the same day to avoid variations from instrumental drift.

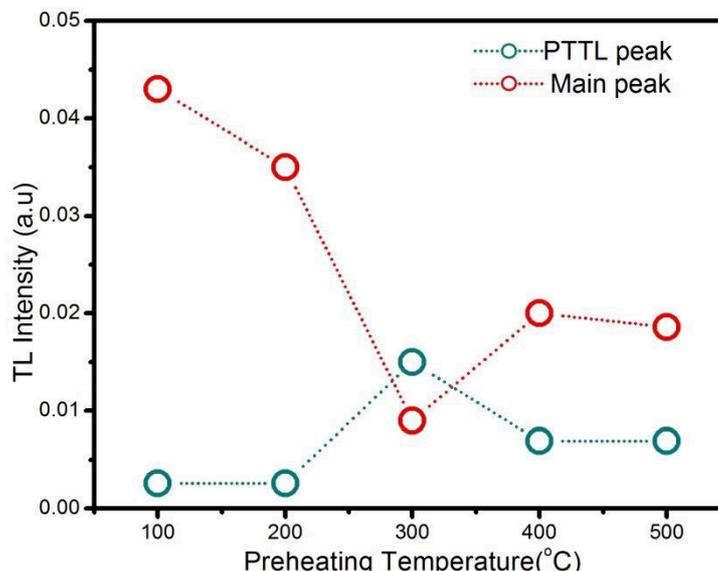


Figure 7. TL peak intensity of the PTTL curves as a function of the preheating temperature.

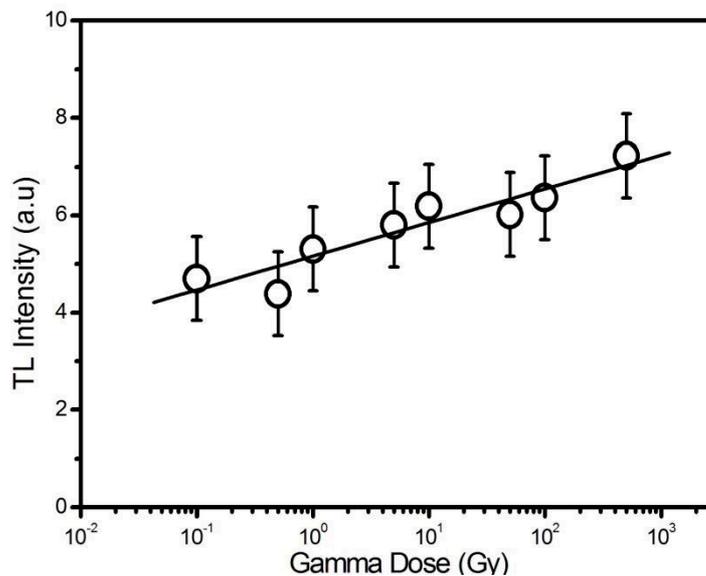


Figure 8. The dose dependence of the PTTL signal.

A small growth in the PTTL is observed within the first 2 days storage time. This is thought to result from the migration and aggregation of low-temperature trapping centers to form more stable complex centers (Driscoll et al. (1986)). The stable PTTL signal was found to be large (95%) after a delay period of 14 days of storage.

The physical process can be fitted to of the form $Y = \{0.947 + 0.18 \exp(-X/4.02)\}$ where Y corresponds to the relative intensity of the PTTL signal, X is the time after irradiation. The time dependence arises from the concept of a potential barrier with an exponentially decreasing tunneling probability with increasing separation between trap and recombination site, and a random spatial distribution of traps and centers (McKeever (1985)).

4. Conclusion

Obsidian powder glow curves exhibit a simple, with limited variety of shapes depending upon the treatment condition. Measurement of dose range for the PTTL response of obsidian is helpful in establishing the TL application for which it might be useful in accident and industrial dosimetry. The experimental results have shown that the γ induced main peak appear at nearly the same temperature as the PTTL curves.

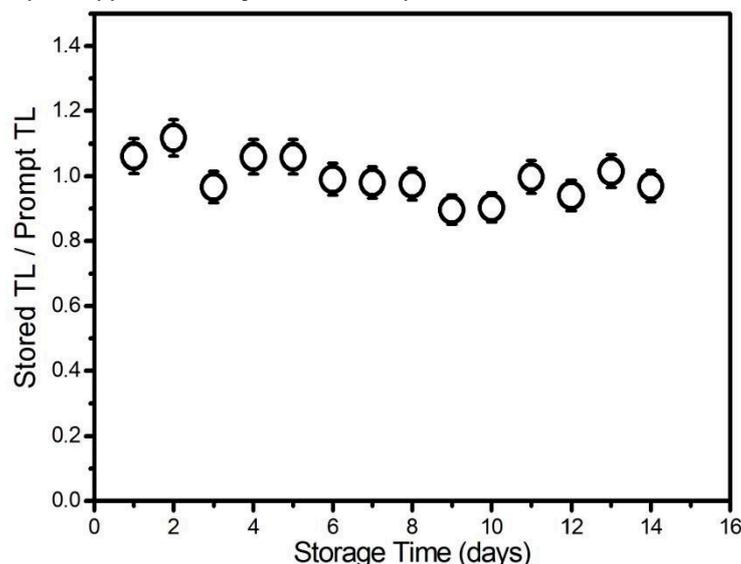


Figure 9. PTTL fading of obsidian at room temperature.

In the PTTL the 75°C appeared at low doses ≤ 1 Gy and has max. height after annealing at 300°C due to transfer from the main trap at 310°C. After the main peak was destroyed the PTTL restored it when preheating the sample at 500°C. Which means that the deep traps, may be considered as the source of the PTTL signal, located near 500°C.

These results show a primary study on obsidian PTTL for use in UV dosimetry. The PTTL responses as

a function of gamma dose are linear in a large range. The behavior of the PTTL curves strongly depends on the preheating temperature and is correlated with the intensity of the main TL peak.

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