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EFFECTS OF DIFFERENT UV EXPOSURES ON THE THERMOLUMINESCENCE OF HIGH ALBITE

V. Correcher (1), J. Garcia-Guinea (2), A. Delgado (1)
(1) CIEMAT, Avda Complutense 22, 28040 Madrid, Spain. Email: JLUIS@CIEMAT.ES
(2) MNCN-CSIC, C/ Jose Gutierrez Abascal 2, 28006 Madrid, Spain.

ABSTRACT

High albite can be used as a new UV dosimeter. Heated natural albite posseses a special property: it has strong thermoluminescence (TL) blue emissions under UV radiation. The TL glow curves of high albite after different times of UV exposure show PhotoTransferred ThermoLuminescence (PTTL) from deeps traps (345^{o}C - 450^{o}C) to shallow traps (150^{o}C) which can be used to store UV damage. The point of inflexion of the process occurs after one hour under UV radiation, the low temperature peaks start gaining in intensity while the high temperature peaks stop dropping and stabilize into a plateau. Samples which have been subjected to UV radiation for 1 hour at different temperatures (RT, 70^{o}C, 105^{o}C and 140^{o}C) give glow curves which display an increase of the maxima peaks according to temperature (up to 300^{o}C). High albite, UV irradiated (for one hour) at 70^{o}C displays a TL signal decrease of 30% after 1000 hours of storage at room temperature with high intensity emissions, for this reason this material could be a good UV dosimeter.

I.- INTRODUCTION

Gartia & Robertson (1989) stated a new type of regenerated thermoluminescence in albite. The TL zeroing in archaeological dating is the time of heating of the sample and the TL zeroing in geological dating is the moment of last exposure to sunlight. In preheated albites, the opposite is true - sunlight (UV radiation) produces strong TL blue emissions. A recuperation of the luminescent signal (IRSL) occurs on preheating treatments and it has consequences for dating young sediments (Rees-Jones & Tite, 1994).

This effect allows high-albite to be used as a UV dosimeter with improved properties such us low-cost and non-solubility. The depletion of stratospheric ozone and the health risks (cataractogenesis, Sliney 1994; skin cancer, Herlihy et al., 1994; etc.) associated with the consequent increase in UVB radiation have prompted efforts to measure solar UV radiation using new dosimeters. In turn, heated feldspars (i.e., albite) are environmental materials which were exposed during thermal/radiation accidents and can be useful for retrospective dosimetry purposes (Correcher & Delgado, 1996).

Albite, $NaAlSi_3O_8$ is monoclinic C2/m above 1000°C (monalbite) but shrinks around the Na atom to the triclinic $C\bar{1}$ structure below this temperature. When working with thermal treatments of feldspars it is important to take into account the following: a) the noncollapsible feldspar framework is extremely flexible and tilts with temperature changes, b) the fast alkali (Na+) self-diffusion at low temperatures and c) the frequently sluggish kinetic activity of the solid state processes which involve atomic displacements (i.e. Al/Si order-disorder).

2.- ULTRAVIOLET RADIATION DOSIMETERS

Quantifying individual exposures to ultraviolet B radiation (UV dosimetry) is crucial for the understanding of the etiology of skin cancers, for the control of phototherapy for psoriasis and, in short, in most living processes. UV dosimeters are found in many different materials and mechanisms: (Fig. 1) a) Photoinduced damage in molecular materials, such as DNA blocks - from mononuclear leucocytes, saccharomices cerevisiae, etc. (Yoshida & Regan, 1995), or RNA (uracil, dioxypyrimidine, etc.), b) Inactivation of dried spores of *Bacillus subtillis* on glass and plastic biofilms (Elnaggar et al., 1995), c) The human cornea as a biological dosimeter for ocular exposure to UV radiation because human photokeratitis is well defined (Sliney, 1994), d) Engineering polymers as polysulphone (Parisi & Wong, 1994), Rise B3 films and CR-39 which is allyl diglycol carbonate (Sydenham et al., 1994), e) Standard silicon CMOS integrated circuit processes allow the fabrication of device structures sensitive to ultraviolet radiation. These UV-sensitive structures can be integrated with detection and signal-conditioning circuitry to make monolithic smart UV sensors (Kerns, 1993), f) Oxides and phosphors doped with rare earth oxides such as Gd_2O_3:Eu, Gd_2O_3:Tb, Gd_2O_3:Dy and Y_2O_3:Eu^{2+} (Yeh & Su, 1996), g) Salts doped with rare earth such as KBr:Eu^{2+} (Melendrez, 1996), $CaSO_4$:Dy Goyet, 1993, etc.), g) Glasses e.g. Vycor (Justus, 1995), h) Electronic devices (Roelandts at al., 1995) and, i) Environmental minerals, such as, albite (this work) which moreover are insoluble mono-block, low-cost materials which are simply measured with TL and additionally, could permit UV retrospective dosimetry studies.

3.- SAMPLES AND METHODS

To avoid undesirable physical variables we have selected transparent monocrystal samples of albite (CLBR) from Minas Geraes (Brazil); albite is a representative choice because it is both an alkali feldspar and a plagioclase term. The sample CLBR is a cleavelandite low albite (K_2O=0.35%, Na_2O = 10.78%, CaO = 0.13%) previously studied by optical microscopy, Infrared spectroscopy (IR), X-ray diffraction (XRD) and X-ray fluorescence (XRF), differential thermal analyses (DTA) and other previous 3DTL luminescence analyses in Sussex (Garcia-Guinea et al., 1996). These natural Brazilian albites were preheated at 1140°C for 30 minutes to transform low albite into monalbite.

Spectral measurements of preheated CLBR sample with UV at RT were made upon the Sussex high sensitivity thermoluminescence spectrometer (Luff & Townsend, 1993). The starting point of the experiment was Townsend's suggestion that the 3DTL signal from a preheated CLBR albite (1140°C/15 days) came from the UV light produced by the flourescent tubes of the room. To check this, the preheated albite was irradiated under a mercury UV lamp for 5 minutes and the 3DTL signal (at 420nm) rose to 1000 arbitrary units (Figs. 2A&B).

More rigorous testing of high-albite and UV relationships was carried out to clarify the effects that UV exposure at different temperatures and for different amounts of time had on the thermoluminescence of this preheated albite (CLBR). A monochromatic UV radiation (254.7 nm) from a Hg lamp was used to stimulate the emissions. The temperature control during the UV irradiation was carried out using an original UV thermo-irradiator developed by Delgado et al., (1996). The thermo-UV-irradiated samples were measured using an automated RISØ TL system model TL DA-12 (Bøtter-Jensen and Duller 1992). Signals were detected using a blue filter (FIB002) situated in front of an EMI9635QA photomultiplier tube. The RISØ TL measurements were performed by linear heating at 5°C/s up to 500°C in N_2 atmosphere.

4.- EXPERIMENTAL RESULTS

4.1.- TL of High albite after different times of exposure to UV at RT.

Natural thermoluminescence (NTL) glow curves (Fig. 3) were generated after exposure of the sample to enviromental light (electrical and sunlight). NTL albite curves consisted of three maxima, peaked at 150°C, 345°C and 450°C, and all of them (24 aliquot samples) presented the same curve. The intensity of the curve areas range from 50 to 500°C, showed a standard deviation of 9.6%. Preheated albite, after differing lengths of UV exposure at RT, displays significant bleaching of the 345°C and 450°C peaks (Fig. 3). This bleaching is progressive according to the time under UV. However, after exposures of more than an hour the high temperature region seems to be resistant to the bleaching effect, forming a plateau. In comparison with NTL, the UV induced thermoluminescence curves displayed the presence of a low temperature peak (150°C) which becomes visible after 5 minutes of UV exposure. From 5 minutes to 1 hour, the low temperature regions are little affected by UV light, but from one hour on, high albite samples show a growth in intensity with increasing UV irradiation.

4.2.- TL of High albite after one hour under UV at different temperatures

Preheated samples, irradiated under UV for one hour at different temperatures (RT, 70°C, 105°C and 140°C) gave glow curves which clearly display an increase of the maxima peaks according to temperature (up to 300°C) (Fig. 4). The maxima of TL peaks display an ascending curve which fits the equation $y = 298.97 \cdot \exp(x/71.01)$ that approximately corresponds with the Arrhenius formula that governs the thermal detrapping of electrons in isolated glow peaks in accordance with conduction band theory. 300°C is a common maxima in TL of Na feldspars (albite and perthite). The 3DTL of high albite after 5 minutes of UV radiation (Figure 2) displays a multiorder kinetic (MOK) glow curve which is commonly attributed to continuous trapping-detrapping. All glow curves of CLBR high albite after UV radiation doses and thermal treatments display MOK shapes (Fig. 4).

4.3.- Decay of TL in High albite previously irradiated under UV at different temperatures.

Aliquots of high albite, irradiated under UV for one hour (at temperatures of 70ºC and 140ºC) were stored at RT in darkness to check the TL fading after a maximum of 2500 hours (Fig. 5). High albite, UV irradiated (for 1 hour) at 70ºC displays a TL signal decrease of 30% after 1000 hours of storage. These 70ºC samples display a stable TL signal and its glow curve reaches high intensity values (approximately 18 000 a.u./mg) certainly, this thermo-UV-irradiated High albite could be considered a good UV dosimeter. Conversely, the High albite which was UV irradiated for 1 hour at 140ºC, shows a TL signal decrease of 50% after being stored 1000 hours. The distinct drop of intensity after approximately 500 hours storage makes this 140ºC sample less suitable for UV dosimetry. All thermo-UV-irradiated CLBR aliquots display good TL signals, for this reason, CLBR High albite could be used for UV dosimetry even in unfavorable conditions**.**

5.- DISCUSSION AND CONCLUSIONS

When the TL glow curves of high albite after different times of UV exposure (5, 10, 20, 30, 60, 120, 240 and 480 minutes) are placed together, a clear example of phototransferred thermoluminescence (PTTL) can be observed. This is caused by the optically stimulated transfer of electronic charge from deep traps (345°C and 450°C peaks) to shallow traps, and results in the generation of thermoluminescence peaks at low temperatures, i.e. 150°C (Fig. 3). The spectra position of these TL curves was achieved using a blue filter (FIB002) and a PM tube (EMI9635QA) in a RISØ TL system and corresponds with the 420nm effect obtained in a 3DTL obtained in Sussex University (Fig. 2). Taking into consideration the role of $[AlO_4/M^+]^0$ centres in the 380nm TL of quartz (Martini et al., 1995) and the studies on the time and wavelength response of phototransferred thermoluminescence in quartz (Alexander et al., 1997), it is possible to speculate that there is a similar case of PTTL in high albite, including $[AlO_4/M^+]^0$ centres ($M^+=Na^+$) with a similar spectra range of 420nm instead of 380nm in quartz.

The TL glow curves of high albite after one hour under UV at different temperatures such as RT, 70°C, 105°C and 140°C, display an increase of the maxima peaks according to temperature. For the same dose of UV irradiation (one hour), the increase of sample temperature (up to 300°C) creates deeper traps. 300°C is a common maximum temperature in TL of Na feldspars (albite and perthite) and 300°C is also the point where ionic conductivity of albite begins (thermal Na self diffusion). The multiorder kinetics of these glow curves of high albite are commonly attributed to a continuous trapping-detrapping phenomenon. However, it is very difficult to associate the trapping and recombination sites with specific defects of albite structure, e.g. Kronenberg et al. (1996) studied hydrogen defects in potassium feldspar and found that the kinetics of diffusional exchange, colouration through radiation damage and deformation of feldspars are all influenced by the presence of hydrogen defects.

High albite, UV irradiated for 1 hour at 70ºC, displays a TL signal decrease of 30% after 1000 hours of storage. However, this decreased TL signal is still intense. The properties of high albite appear to make a good UV dosimeter. Further experiments under sunlight are being carried out to test the high albite as a new environmental UV dosimeter. More thermo-UV-irradiated CLBR High albite stored aliquots will allow testing times to be lengthened. In addition, insoluble monocrystals of thermo-UV-irradiated CLBR High albite have been placed in different geographical areas to accumulate natural ultraviolet radiation under distinct atmospheric conditions.

ACKNOWLEDGEMENTS

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FIGURE CAPTIONS:

Fig.1.-Schematic representation displaying the main types of known UV dosimeters.

Fig.2.- A) 3DTL displaying no emissions corresponding to high albite which was stored, heated and analyzed in darkness. **B)** 3DTL displaying high intensity emissions corresponding to high albite which was stored and heated in darkness, but before analysis, irradiated for 5 minutes under a mercury UV lamp.

Fig.3.- Association of TL curves of high albite after different periods of time under a UV lamp. The 3D model displays a phototransferred thermoluminescence effect from high temperature to low temperature peaks.

Fig.4.- TL glow curves displaying a clear increase of the maxima peaks according to temperature (up to 300^{o}C).

Fig.5.- A) Graph displaying the signal intensity loss of high albite (heated at 70^{o}C under UV radiation) during storage time. **B)** Graph displaying the signal intensity loss of high albite (heated at 140^{o}C under UV radiation) during storage time.

SECOND ANNOUNCEMENT AND CALL FOR PAPERS
3rd INTERNATIONAL SYMPOSIUM
LUMINESCENT DETECTORS AND TRANSFORMERS OF IONIZING RADIATION

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About 110 persons responded to the first circular. The Organizer is glad to inform that Radiocarbon Foundation in Gliwice will co-organize the symposium and provide free administration and banking services.

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The proceedings of the symposium will be published by Elsevier in the international journal Radiation Measurements and only the papers presented by the author at the conference (in either form) will be considered for publication. The papers will be refereed according to the rules of the Journal.

The instructions for authors will be included in the third circular.

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The potential use of annealed high-albite ($AlSi_3O_8Na$) as an ultraviolet radiation dosimeter

J. GARCIA-GUINEA
MNCN-CSIC, C/Jose Gutierrez Abascal 2, Madrid 28006, Spain
E-mail: guinea@mncn.csic.es

V. CORRECHER, A. DELGADO
CIEMAT, Avda Complutense 22, 28040 Madrid, Spain

Annealed natural albite possesses the special property of having strong thermoluminescence (TL) blue emissions after ultraviolet (UV) irradiation that could be considered for dosimetric purposes. After 5 min of UV irradiation, the spectra TL UV-blue emissions of high albite grow by a ratio of over 1 : 100. Using the RISØ TL reader, the glow curves of high albite after different periods of UV exposure show Photo transferred thermoluminescence (PTTL) from deep traps (345–450 °C) to shallow traps (150 °C). The TL measurements of high albite aliquots, at distinct temperatures for different amounts of time, show, above 100 °C, a consistent plateau after 1 h of UV irradiation. This large increase of the UV-induced blue TL (∼400 nm) in preheated albite could be linked with the fast alkali (Na^+) self-diffusion in the lattice and the activation of the $[AlO_4]^\circ$ centers; this is comparable to the case of quartz.

Gartia and Robertson [1], while recording the high-temperature peaks of an Amelia albite, found that if the sample was heated to 550 °C and then reheated to measure the black-body radiation, a clear peak around 300 °C appeared in the later run. They explain this by a thermally assisted mechanism. In annealed albites, sunlight and/or artificial UV produces strong TL blue emissions. A recuperation of the luminescent signal (IRSL) occurs after preheating treatments, and this can cause problems in geological and archaeological dating [2] and retrospective dosimetry [3] when natural feldspars that have been subjected to heating are used.

The same effect (strong preheating and X-ray or UV irradiation to enlarge the luminescence) has been described in quartz where prolonged high-temperature annealing of the samples in vacuum (10 h at 1400 °C, 1 Pa) reduces the presence of ionic charge compensators at the Al sites and induces an intense 380 nm emission from $[AlO_4]^\circ$ centers [4].

Transparent monocrystal samples of natural low albite (CLBR) from Minas Geraes (Brazil) were analyzed. These crystals ($K_2O = 0.35\%$, $Na_2O = 10.78\%$, $CaO = 0.13\%$) were previously studied by optical microscopy, infrared (IR) spectroscopy, X-ray diffraction (XRD), X-ray fluorescence (XRF), differential thermal analysis (DTA) and other previous 3D TL luminescence analyses [5]. The samples were annealed at 1050 °C for 15 days to transform them from low albite to high albite.

Spectral TL measurements of this high albite after UV irradiation at room temperature (RT) were taken using the University of Sussex high-sensitivity thermoluminescence spectrometer [6] within a spectral range of 200–800 nm and a resolution of 3 nm. Cleaved chips of non-irradiated annealed albite, X-irradiated with 50 Gy, were mounted onto aluminum discs using silicon oil. TL measurements were made from 30 to 400 °C at a heating rate of 2.5 °C s^{-1} Photon emission from the sample was detected via a pair of spectrometers, with gratings blazed for the UV-blue (200–450 nm) and blue-green-red (400–800 nm) parts of the spectrum, and a pair of sensitive photomultiplier tubes. Signals were recorded over the wavelength range of 200–800 nm, with a resolution of 5 nm for 100-point spectra and 3 nm for 200-point spectra, and all signals were corrected for the spectral response of the system.

More rigorous testing of the relationships between high albite with UV was carried out to clarify the effects of UV exposure at different temperatures and for different amounts of time on the TL of this annealed albite. A monochromatic UV radiation (253.7 nm) from a Hg lamp was used to stimulate the emissions. The temperature control during the UV irradiation was carried out using a UV irradiator [7]. An automated RISØ TL system model TL DA-12 was used to carry out the TL measurements [8] with a blue filter (FIB002) situated in front of an EMI9635QA photomultiplier tube. These measurements were performed by linear heating at 5 °C s^{-1} up to 500 °C in N_2 atmosphere.

To check the spectra TL of a high albite (in the high sensitivity TL spectrometer) after different times of exposure to UV at RT, an annealed sample (1050 °C/ 15 days) was irradiated under a mercury UV lamp at RT for 5 min. The 3D TL signal (at 420 nm) rose, from the initial 10 arbitrary units (a.u.) to 1000 a.u. (Fig. 1a and b). In the RISØ TL reader, a set of TL measurements was recorded to compare this 3D TL observation and test the effects, on its TL, of different UV exposures at different temperatures (70, 105 and 140 °C). The TL glow curves were generated after a short exposure (approximately 15 s) of the high albite to environmental light (electrical and sunlight). These glow curves consisted of three maxima, peaked at 150, 345 and 450 °C, and all of them (24 aliquot samples) presented the same curve (Fig. 2).

The intensity of the curve areas, ranging from 50 to 500 °C, displayed a standard deviation of 9.6%. Annealed albite, after differing lengths of UV exposure

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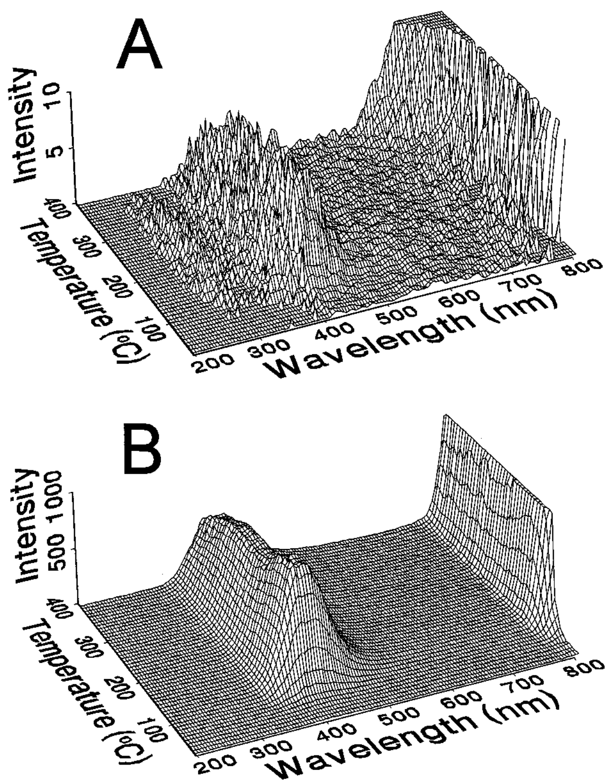

Figure 1 (a) CLBR albite 1050 °C for 15 days. 3D TL displaying small emissions corresponding to high albite that previously was irradiated with an insignificant amount of environmental light; (b) 3D TL displaying high-intensity emissions corresponding to high albite irradiated for 5 min under a mercury UV lamp.

at RT, displays significant bleaching of the 345 and 450 °C peaks (Fig. 2). This bleaching is proportional to the time it was exposed to UV radiation. After exposures of more than an hour, however, the high-temperature region seems to resist the bleaching effect, forming a plateau. In comparison with the first TL glow curve (high-albite sample with a little environmental light), the UV-induced TL curves displayed the presence of a low-temperature peak (150 °C), which becomes visible after 5 min of UV exposure. From 5 min to 1 h, the low-temperature regions are little affected by UV light, but from 1 h on, high-albite samples show a growth in intensity with increasing UV irradiation (Fig. 2).

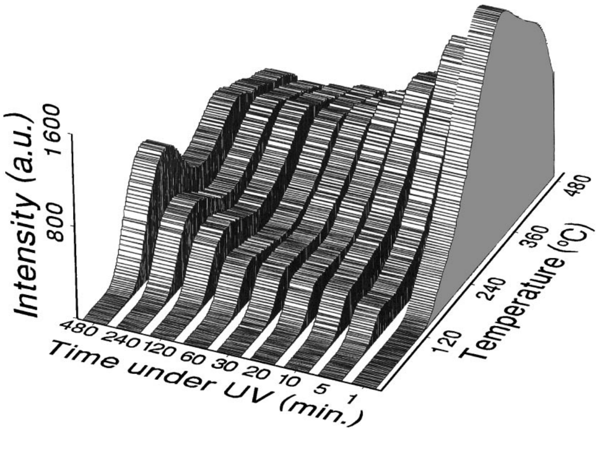

Figure 2 Association of TL curves of high albite after different periods of time under a UV lamp. The three-dimensional model displays a PTTL effect from high-temperature to low-temperature peaks.

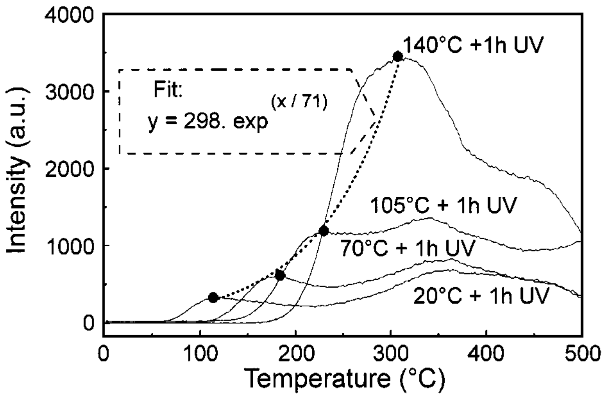

Figure 3 TL glow curves of annealed samples following UV irradiation at different temperatures for 5 min showing a clear increase of the maxima peaks according to temperature (up to 300 °C).

The annealed albites, irradiated under UV for 1 h at different temperatures (RT, 70, 105 and 140 °C) gave glow curves that clearly displayed an increase of the maxima peaks according to temperature (up to 300 °C) (Fig. 3). The maxima of TL peaks display an ascending curve that approximately fits the equation $y = 300 \cdot \exp(x/71)$ (result obtained using the Origin fitting program). The 3D TL of high albite after 60 min of UV radiation (Fig. 1) displays a glow curve resulting from a wide trap depth distribution. All glow curves of CLBR high albite after UV radiation doses and thermal treatments display these wide trap depth distributions (Fig. 3).

Ninety-six aliquots of high albite, irradiated under UV for 1 h (at temperatures of 70 and 140 °C), were stored at RT in darkness to test the TL fading after a maximum of 2500 h (Fig. 4). While high albite, UV irradiated (for 1 h) at 70 °C displays a TL signal decrease of 30% after 1000 h of storage, this decreased TL signal is still intense. These 70 °C samples display a stable TL signal and a glow curve of high-intensity values (approximately 18 000 a.u. mg^{-1}). Conversely, the high albite, which was UV irradiated for 1 h at 140 °C, shows a TL signal decrease of 50% after being stored for 1000 h. The distinct drop of intensity after storage for approximately 500 h makes this 140 °C sample less suitable for possible dosimetric purposes. All CLBR aliquots of high albite that had been previously irradiated under UV at different temperatures display good TL signals.

The TL glow curves of high albite after different times of UV exposure show PTTL from deep traps (345–450 °C) to shallow traps (150 °C), which can be used to measure UV irradiation. The point of inflexion of the process occurs after 1 h under UV radiation; the low-temperature peaks start gaining in intensity while the high-temperature peaks stop dropping and stabilize into a plateau. Samples that have been subjected to UV radiation for 1 h at different temperatures (RT 70, 105 and 140 °C) give glow curves that display an increase in the intensity of the maxima peaks according to temperature (up to 300 °C). The TL curves of the annealed albite after 1 h under UV at different temperatures such as RT, 70, 105 and 140 °C display an increase of the amplitude of the TL peaks with the

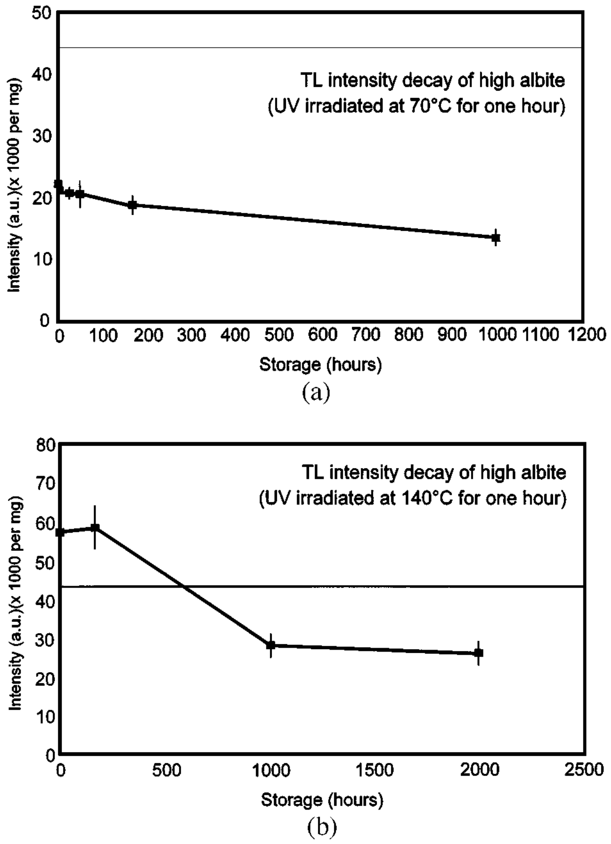

Figure 4 (a) Graph displaying the signal intensity loss of high albite (heated at 70 °C under UV radiation) during storage time; (b) graph displaying the signal intensity loss of high albite (heated at 140 °C under UV radiation) during storage time.

temperature (Fig. 4). For the same dose of UV irradiation (1 h), the increase of sample temperature (up to 300 °C) creates deeper traps. 300 °C is a common maximum temperature in TL of Na feldspars (albite and perthite). The glow curve results from a wide trap depth distribution; it is very difficult, however, to associate the exact trapping and recombination sites with specific defects of albite structure. Taking into consideration the role of $[AlO_4/M^+]^\circ$ centers in the 380 nm TL of quartz [4] and the studies on the time and wavelength response of PTTL in quartz [9], it is possible to speculate that there is a similar case of PTTL in high albite, involving $[AlO_4/M^+]^\circ$ centers ($M^+ = Na^+$) at the 420 nm wavelength instead of the 380 nm wavelength in quartz. Further experiments under sunlight are being carried out to test the high albite as a new environmental UV dosimeter.

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