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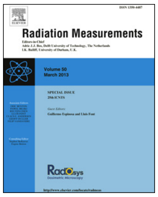

UV-induced blue thermoluminescence of annealed Na-rich aluminosilicates

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HIGHLIGHTS

- UV-induced blue TL of annealed albite is studied.
- Different annealing on several aliquots (from 480 to 960 °C) induces several changes in the TL sensitivity.
- Such changes are linked to dehydration, dehydroxylation processes and changes in the lattice structure.
- The Al/Si disorder gives rise to alkali self-diffusion and thermal stresses.

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ABSTRACT

The luminescence properties of gemstones (silicates, carbonates, phosphates, etc.), are of interest since could be potentially employed as personal dosimeters not only in case of radiation accident or radiological terrorism where conventional monitoring was not established, but also as a UV personal dosimeter. We, herein, report on preliminary results of the UV-induced 480 nm-thermoluminescence (TL) response of a well-characterised natural albite ($NaAlSi_3O_8$) from Minas Gerais (Brazil) that, due to its high transparency, is used as gemstone. Different annealing on several aliquots (from 480 to 960 °C) induces several changes in the sensitivity of the sample probably associated with (i) dehydration and dehydroxylation processes and (ii) changes in the lattice structure increasing the Al/Si disorder giving rise to ionic self-diffusion of alkali atoms, thermal stress into atomic positions and variation in the concentration of alkali ions in planar defects. The complex structure of albite has several planar defects (twinning and exsolution interphases which contain hydroxyl groups, water molecules, etc.) that can act as luminescence centres. When the thermal preannealed samples are exposed to 1 h at 254.7 nm under lab conditions one can appreciate changes in the TL behaviour that allows us to think in the albite as a potential UV dosimeter. ©2006 Elsevier Ltd. All rights reserved.

1. Introduction

Na-rich aluminosilicates are industrially important in the manufacture of ceramics as a component of ceramic clays and some of them, albite ($NaAlSi_3O_8$) for instance, can be used as an ornamental stone or even as gemstone that is of interest since can be potentially employed in situations where retrospective dosimetry for individuals is required but where monitoring was not planned (radiation accident or radiological terrorism). As observed in several aluminosilicates, albite shows a strong UV-induced blue thermoluminescence (TL) emission that could be observed for both ionising and non-ionising (UV) radiation dosimetry purposes (Correcher et al., 1999; Garcia-Guinea et al., 1999). The 380 nm UV emission band is characteristic of mineral phases containing lattices with SiO_4 tetrahedra and is detected when interstitial alkalis are placed in adjacent positions to aluminium ions (Garcia-Guinea et al., 1999). To obtain this emission band is necessary: (i) a thermal sensitisation of the lattice with different thermal treatments and (ii) an activation of emission centres under short wave radiation such as, X-ray, gamma, or UV. The combination of this double effect induces the mobility of the alkalis due to the radiation causing a high number of electron–hole pairs in the lattice where some holes can be trapped forming $[AlO_4/M^+]$ centres. When the given energy is enough, recombination of the electrons with the hole trapped adjacent to $Al{-}M^+$ reduces the presence of ionic charge compensators at the Al sites and gives rise to the 380 nm luminescence emission to $[AlO_4]^o$ centres (aluminium-hole centres) (Garcia-Guinea et al., 2001; Martini et al., 1994). It could be appreciated how alkali-rich aluminosilicates in general are highly sensitive to

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ionising and non-ionising radiation, displays a reasonably good reproducibility, possess low fading and good dose linearity in the ranges of interest (Correcher et al., 2004).

We herein study the effect of the thermal annealing (from 480 to 960 °C) on the blue TL emission of a well-characterised albite from Minas Gerais (Brazil) after UV exposures of 1 h under laboratory conditions.

2. Samples and experimental

Measurements were carried out on a natural Albite sample collected in Minas Gerais, Brazil (Fig. 1). The sample, a sodium-rich feldspar ((Na,K)[$AlSi_3O_8$]) with a mean molecular composition of Orthoclase (Or) and Albite (Ab) of Or_1Ab_{99}, has been elementarily analysed by X-ray fluorescence (XRF), using a PHILIPS PW-1404 with an Sc–Mo tube having analyser crystals of Ge, LIF220, LIF200, PE and TLAP giving rise to Fe_2O_3 0.06%, SiO_2 67.63%, Al_2O_3 19.52%, MgO >0.01%, MnO 0.02%, CaO 0.17%, Na_2O 11.38%, K_2O 0.30%, TiO_2 0.09%, P_2O_5 0.14%. A Super-Q manager (Panalytical-Spain) was used as the analytical software. Three pellets of 8 g of milled sample with 0.1 g of an acryl type resin (Elbacite 2045, produced by E.I. Dupont de Nemours Co., Ltd.) were pressed under 2×10^4 kg (about 2000 kg/cm^2) and dried at 40 °C in an atmospheric chamber. Trace elements were analysed by a Finnigan MATSOLA (FinniganMAT, Bremen, Germany) plasma source mass spectrometer (ICP-MS). Each sample was introduced into 27.12 MHz argon plasma, using a Meinhard concentric nebulizer and a Gilson Minipuls 2 peristaltic pump. The instrument was calibrated using pure NIST traceable single-element stock solutions, supplied by Alfa Products Ltd., (Karlsruhe, Germany). The chemical analysis of trace elements is as follows (in ppm): Rb 258, Ba 22, La 5, Ce 2, Y 11, Th 8, Cu 6, Mn 19. TL measurements were performed using an automated Risø TL DA-12 system (Bøtter-Jensen and Duller, 1992). This reader is provided with an EMI 9635 QA photomultiplier and the emission was observed through a blue filter (a FIB002 of the Melles-Griot Company) where the wavelength is peaked at 320–480 nm; FWHM is 80 ± 16 nm and peak transmittance (minimum) is 60%. The TL reader is also provided with a $^{90}Sr/^{90}Y$ source with a dose rate of 0.012 Gy s^{-1} calibrated against a ^{60}Co photon source, in a secondary standards laboratory (Correcher and Delgado, 1998). All the TL measurements were carried out using a linear heating rate of 5 °C s^{-1} from RT up to 500 °C, in a N_2 atmosphere. Four aliquots of 5.0 ± 0.1 mg of the samples were used for each measurement. The incandescent background was directly subtracted from the TL data. The sample was carefully ground to powder with an agate pestle and mortar to avoid triboluminescence (Garcia-Guinea and Correcher, 2000). 3D-TL spectra of albite chips were performed in the high sensitivity TL spectrometer at Sussex with a spectral range of 200–800 nm and a resolution of 3 mm (Luff and Townsend, 1993). Cleaved chips of the sample were mounted onto aluminium discs using silicone oil. Measurements of the natural TL were made from 30° to 400 °C at a heating rate of 2.5 °C s^{-1}. All the luminescence spectra were corrected for response of the system. The UV exposures were carried out with an automated irradiator developed in CIEMAT (Delgado et al., 1996) that allows UV illumination with a TUV-6W Hg lamp (254.7 nm, UV irradiance value at 10 cm was 0.03 Wm^{-2}) and controlled thermal treatments. Powdered albite aliquots were irradiated with UV-C from 1 to 8 h at RT. Thermal preheating of the samples at 480, 780 and 960 °C for 4 h were performed in a programmable temperature controlled tubular oven with quartz chamber in open air. The heating rate to reach the temperature was about 100 °C min^{-1}, and the annealed samples were cooled to room temperature at 4 °C min^{-1}.

3. Results and discussion

3D-TL plots displaying intensity (in a.u.) against temperature (in °C) and wavelength (in nm) are shown in Fig. 2. Chips of natural TL (NTL) of albite shows (i) a UV-B large flat emission of multi-order kinetics (at 290 nm), related to defect-sites associated with the presence of the Na ions. The actual effect consists on the ionic motion of metastable accumulated monovalent ions along interfaces that gives rise to a continuum in the trap distribution observed and can be linked to different processes that simultaneously can occur during the readout: partial phase transformations; consecutive breaking linking of bonds; alkali self-diffusion through the void channels in the aluminosilicate lattice; redox reactions, etc. (ii) A broad UV-A band around the 380 nm linked to interlamellar strain in twinned perthite and can be assigned to the presence of defects on non-bridging oxygens (NBOHCs); this also agrees with [AlO_4]^o centres, ionic self-diffusion of alkali atoms, thermal stress into atomic positions and concentration of alkali in planar defects. The UV-blue emission is due to the recombination of thermally released electrons with hole centres. And (iii) a weak 550 nm green emission from Mn^{2+} point defects (Correcher and Garcia-Guinea, 2001).

Thermal annealing induces several changes in the sensitivity of the sample probably associated with dehydration and dehydroxylation processes and/or redox reactions, but also with changes in the lattice structure. Alkali aluminosilicates are characterized by a low Al/Si disorder at room temperature where the stable structure is the triclinic ordered lattice containing

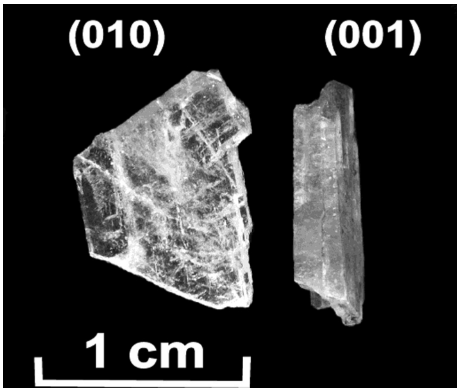

Fig. 1. Transparent monocrystals of albite samples.

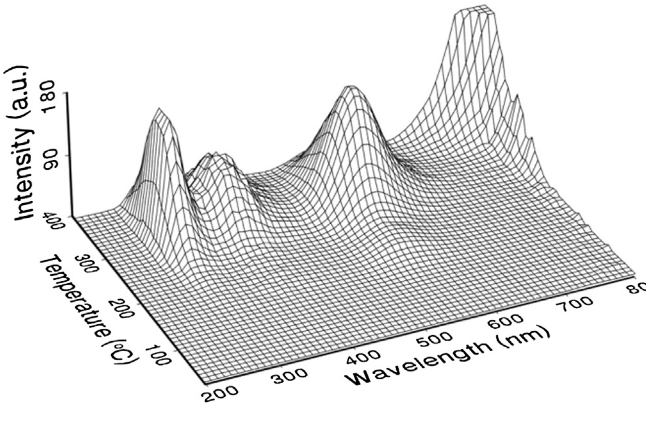

Fig. 2. Natural 3D-TL emission of albite from Minas Gerais, (Brazil).