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Natural blue thermoluminescence emission of the recently fallen meteorite in Villalbeto de la Peña (Spain)

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Abstract

This paper reports on the thermal stability of the natural blue thermoluminescence (TL) of a well-characterised meteorite fallen in Villalbeto de la Peña (Palencia, Spain) in January 4, 2004. The meteorite specimen exhibits a complex structure of the spectra emission a wide broad maximum peaked at about 320 °C that can be associated with consecutive breaking linking of bonds (e.g. of Al–O, Cr–O, Fe–O) from lattices of several phases, i.e., olivine, low-Ca pyroxene, plagioclase and redox reactions. The natural TL emission seems to be essentially caused by the Na–Ca plagioclase as the brighter material inside this L6 chondrite. The thermal stability tests, at different temperatures, confirm a continuous trap distribution with progressive changes in the glow curve shape, intensity and temperature position of the main peak. In consequence, the glow curve analysis methods commonly used for synthetic materials based on single-discrete traps cannot be applied for this material.

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Keywords: Thermoluminescence; Meteorite; Continuous trap distribution

1. Introduction

The luminescence studies of meteorites are usually performed on fragments fallen down on the Earth's surface long time ago. Consequently, the natural thermoluminescence (NTL) emission of the samples is mainly attributed not only to the extraterrestrial radiation (high-energy cosmic rays and cosmogenic isotopes) but also to the environmental radioactivity from the Earth. This information can be used not only to determine the time that the meteorite fell to the Earth [1] but also it should be an indicator of meteorite orbit and useful to characterise the metamorphism and type of the chondrite [2]. NTL levels of recently fallen meteorites are a guide (i) to estimate the closest approach to the Sun, i.e., to know the thermal history of the sample and (ii) the irradiation history strictly linked to extraterrestrial exposure [2]. The study of the thermoluminescence (TL) properties of mineral phases is of great interest in solid-state dosimetry applied to dating and retrospective dosimetry [3]. Accordingly, the radiation environment of the outer space can be estimated using extraterrestrial natural materials as a first and good approach.

NTL is the energy stored in crystals of the mineral phases (mostly plagioclases feldspars in meteorites) by ionising radiation released as visible light by heating. Several studies carried out on terrestrial mineral phases demonstrate that quartz [4] and feldspars [5], ubiquitous in nature, (i) are highly sensitive to radiation, (ii) their TL signals possess low fading and good dose linearity in the ranges of interest and (iii) the results are reproducible. These facts make them undoubtedly useful for radiation dose assessment in uncontrolled dosimetric areas.

The Villalbeto de la Peña meteorite is an L6 ordinary chondrite mass of 760 ± 150 kg (before going through the Earth's atmosphere) that has been employed for this study because: (i) is one of the nine meteorites with a known orbit, it comes from the main asteroid belt [6]; (ii) there are enough sample to be studied, 32 meteorite fragments in a

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range of 11 g to 1.4 kg; and (iii) the elapsed time between the meteorite fireball dropping and the recovery of the fragments was very short (no more than 1 month). It is of great interest to determine the absorbed dose of the sample with a negligible contribution of terrestrial environmental radiation and, in addition, to estimate the presence of cosmogenic radionuclides of short lifetime.

The main aim of this paper is to study the thermal stability of the natural blue TL of a well-characterised meteorite recently fallen in Villalbeto de la Peña (Palencia, Spain) to determine the trap structure. The value of the activation energy (E_a) has been estimated employing the initial rise approach.

2. Materials and methods

Samples of the Villalbeto de la Peña meteorite (3.4 g cm^{-3} of bulk density), fallen down in January 4, 2004, has been collected in situ during January and February in the northern part of Spain.

The meteorite fragments (Fig. 1) have been characterized by means of X-ray diffraction (XRD), polarizing microscopy and scanning electron microscopy (SEM) together with the isotope analysis. The structural data were assessed by a Siemens D-5000 X-ray automated diffractometer using the CuK$_\alpha$ radiation with a Ni filter at a setting of 40 kV and 200 mA. SEM observations were done using a Zeiss DSM-90 (40 kV electron microscope with a resolution of 70 Å). The microanalysis detector was a Si–Li model, using a Tractor Northern Z2 computer. Sample metallization was made using gold vapour in vacuum (50 Å of gold cover). The activity of the cosmogenic radionuclides was estimated by gamma spectrometry using a high-purity Ge detector in the Lab. Nazionali del Gran Sasso (Rome, Italy) [7].

Fig. 1. Fragment of Villalbeto de la Peña meteorite.

The TL measurements were performed using an automated Risø TL system model TL DA-12 [8], reader with an EMI 9635 QA photo-multiplier and the emission was observed through a blue filter (FIB002 Melles-Griot) with transmission in the 320–480 nm range; FWHM is 80 ± 16 nm and peak transmittance is 60%. It is also provided with a ^{90}Sr/^{90}Y source with a dose rate of 0.018 Gy s^{-1} calibrated against a ^{60}Co photon source in a secondary standards laboratory [2]. The TL measurements were performed at 5 °C s^{-1} from room temperature to 500 °C in a N_2 atmosphere. A set of four aliquots of the sample with 5.0 ± 0.1 mg each was used for the measurement. The sample was carefully powdered with an agate pestle and mortar to avoid triboluminescence [9]. The incandescent background was subtracted from the TL data.

3. Results and discussion

The blue TL emission has been analysed on the two parts that make up the fragments of the whole meteorite: the inner part constituted by olivine, low-Ca pyroxene, plagioclase, merrillite and minor amounts of some other aluminosilicates, troilite and native Fe and Ni (Fig. 2a) and the fusion crust, 1 mm black outermost layer mainly composed by neo-formed Fe–Ni spinel phases (Fig. 2b). As expected, the levels of NTL of the fusion crust of this freshly fallen meteorite are close to zero due to the atmospheric heating that could reach up to 1500 °C.

The NTL emission of the inner part of the meteorite specimens (Fig. 3) is very intense compared to other aluminosilicates measured under similar conditions. The Villalbeto meteorite samples reach intensity light levels of ~2 × 10^5 counts (in arbitrary units, a.u.), whereas emissions from natural adularia (K-rich feldspar), kaolinite or polymineral materials, i.e., lava flows, reach levels of ~500, ~14,000 and ~400 a.u., respectively [10–12]. This TL emission can be attributed to the presence of plagioclase in the polymineral composition of the meteorite since: (i) it is the most abundant mineral phase in the chondrite, (ii) it has no iron in this lattice that can act as a TL inhibitor in

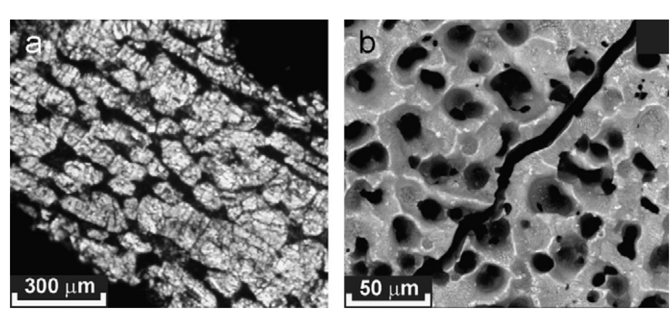

Fig. 2. (a) Polarized photomicrograph of the Villalbeto L6 chondrite exhibiting olivine crystal strips of a characteristic chondrule. (b) SEM image of a representative part of the fusion crust showing needles of native Fe and Ni and craters.

the UV–blue region and (iii) the feldspar group displays the most intense glow TL signals. Cosmic rays and cosmogenic isotopes, namely ^{22}Na, ^{48}V, ^{51}Cr, ^{54}Mn, etc., induce the intensity of the NTL curve that is proportional to the absorbed dose [6].

Curves of NTL were analysed in terms of both first- and second-order kinetics equations, respectively, corresponding to the cases where the intensity of the TL is proportional to the concentration of thermally released charges, and where the thermally released charges are retrapped at least once before the recombination process. However, the best-fit parameters obtained, based on the value of the factor of merit, were unsatisfactory; TL emissions of the meteorite cannot be explained employing the commonly accepted model based on the discrete trap distribution model, since it is not possible to determine some physical parameters such as trap-energies or pre-exponential factors. Therefore, due to the complexity of the NTL glow curves a structure of a continuous trap distribution, as in other polymineral phases involving multiorder kinetics [12], might be considered. Thus, the experimental work was focused on the determination of the trap structure of several aliquots of the meteorite, which can be confirmed by the evolution of the TL glow curve after progressive thermal treatments. The samples were preheated at temperatures ranging from 270 to 430 °C in nine steps of 20 °C to check the response of the stability of the group of components after different sample pre-annealing, T_{stop} (Fig. 3).

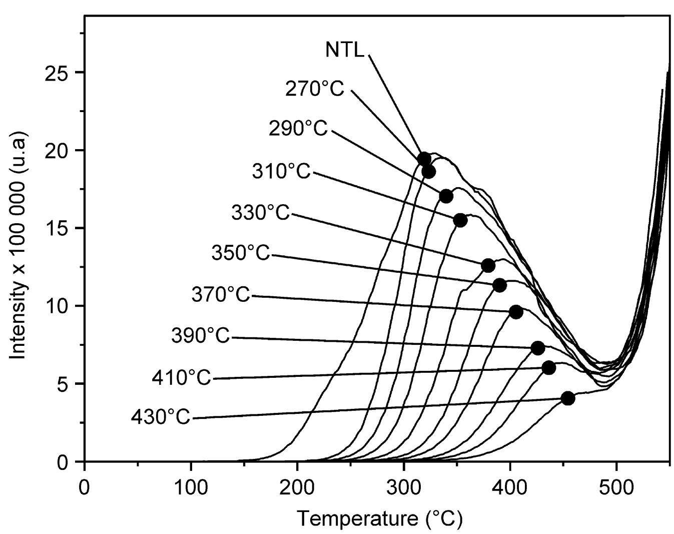

Fig. 3. Thermal stability of the natural TL emission of the meteorite after different thermal preannealings in the range of 270–430 °C.

The NTL feature typically involves the maximum peak shifting to higher temperatures and a change in the shape and intensity of the TL distribution in good agreement with the specific thermal pre-treatment. This means that the luminescence process stems from a multiplicity of traps with different depths. The thermolabile broad band of blue emissions shows TL glow curves of multi-order kinetics involving continuous processes of trapping–detrapping, namely a continuous trap distribution structure.

This complex structure of the emission spectra based on a wide broad maximum peaked at about 320 °C (Fig. 4) could be associated with consecutive breaking linking of bonds (e.g. of Al–O, Cr–O, Fe–O) and redox reactions that take place in the luminescent mineral phases.

The value of the activation energy (E_a in eV) has been calculated by the initial rise (IR) method that assumes the number of electron trapped in the low-temperature tail of the analysed peak to be constant [13]. Thus, the initial part of the TL glow peak might follow an exponential dependence regardless of the kinetic order and the assumption of the quasi-equilibrium approximation that is necessary to solve the equation $I_{TL} \sim \exp(-E_a/kT)$, where I_{TL} is the intensity of the TL emission (in arbitrary units, a.u.), k is the Boltzmann's constant and T is the absolute temperature (in degrees Kelvin, K). The Arrhenius plot (ln I_{TL} vs. $1/T$) allows us to calculate the activation energy from the slope $-E/k$ that is not dependent on the order kinetic parameter.

The increase of thermal preheating treatment performed on different aliquots gives rise to a growth in the value of the activation energy in the studied range (270–430 °C)

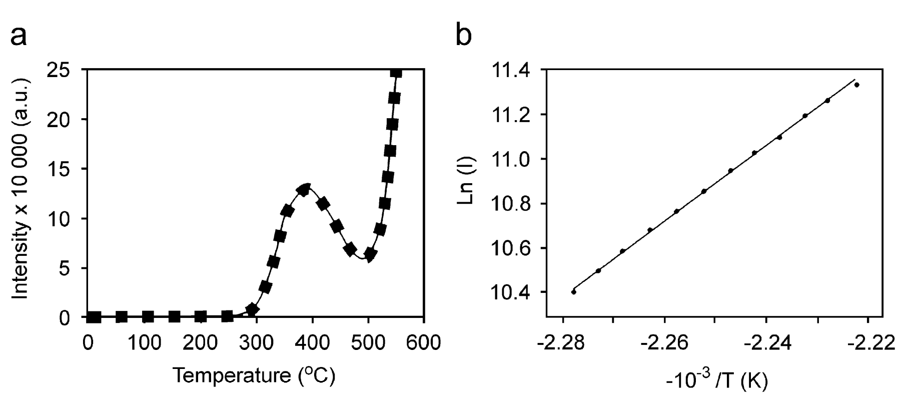

Fig. 4. (a) TL emission of the meteorite obtained after a thermal preheating of 330 °C. The region of the curve selected for the estimation of the activation energy (E_a) employing the IR method was 290–345 °C. (b) Arrhenius plot of the low-T side of the TL curve of the meteorite where the $E_a = 1.33 \pm 0.011$ eV.

Table 1
E_a of the main peak obtained by the IR method after the thermal preheating (T_{stop}) performed on NTL emission of meteorite aliquots

T_{stop} (°C)	E_a (eV)
270	1.21 ± 0.021
290	1.35 ± 0.022
310	1.38 ± 0.024
330	1.33 ± 0.011
350	1.58 ± 0.014
370	1.60 ± 0.015
390	1.72 ± 0.005
410	1.76 ± 0.005
430	1.73 ± 0.005

which corresponds to the energy distribution (from 1.21 to 1.73 eV) of the active traps and indicates the thermal stability of the sample (Table 1). Fig. 3b displays the Arrhenius plot of the 330 °C preheated aliquot activation energy employing the IR method.

4. Conclusions

The NTL glow curve of Villalbeto de la Peña meteorite exhibits emission spectra with a very complex structure. It consists of a broad maximum peaked at about 320 °C that can be associated with consecutive breaking linking of bonds (e.g. of Al–O, Cr–O, Fe–O) and redox reactions. According to the mineralogical analysis, the NTL emission is due to the Na–Ca plagioclase since this is the brighter material inside the chondrite. The tests of thermal stability at different temperatures indicate a continuous trap distribution with progressive changes in the glow curve shape, intensity and temperature position of the main peak. The value of the activation energy, which has been estimated by the IR method, is progressively higher increasing the thermal preheating of the chondrite aliquot. These smooth variations in the energy values confirm the continuum or quasi-continuum distribution of trap levels in the range of 1.21–1.73 eV for the natural TL of this L6 ordinary chondrite.

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