Nuclear Instruments and Methods in Physics Research B 320 (2014) 57–63

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Prompt isothermal decay of thermoluminescence in an apatite exhibiting strong anomalous fading

I.K. Sfampa [a], G.S. Polymeris [b,d], N.C. Tsirliganis [d], V. Pagonis [c], G. Kitis [a,*]

[a] Aristotle University of Thessaloniki, Nuclear Physics Laboratory, 54124 Thessaloniki, Greece
[b] Aristotle University of Thessaloniki, Solid State Physics Section, 54124 Thessaloniki, Greece
[c] McDaniel College, Physics Department, Westminster, MD 21157, USA
[d] Laboratory of Radiation Applications and Archaeological Dating, Department of Archaeometry and Physicochemical Measurements, 'Athena' R. & I. Center, Kimmeria University Campus, GR67100 Xanthi, Greece

ARTICLE INFO

Article history:
Received 1 July 2013
Received in revised form 6 November 2013

Keywords:
Thermoluminescence
Anomalous fading
Apatite
Tunneling luminescence
Luminescence dating
Isothermal TL

ABSTRACT

Anomalous fading (AF) is one of the most serious drawbacks in thermoluminescence (TL) and optically stimulated luminescence (OSL) dating. In the present work the isothermal decay of TL signals from Durango apatite is studied for temperatures located on the rising part of the main TL peak. This material is known to exhibit strong AF phenomena, and its isothermal TL decay properties have not been studied previously. The experimental results show that the characteristic decay time of the isothermal signal does not depend of the temperature, and that this signal does not exhibit the strong temperature dependence expected from conventional TL kinetic theories. This is further direct experimental evidence for the possible presence of tunneling phenomena in this material. The isothermal decay curves are analyzed and discussed within the framework of conventional theories of TL, as well as within the context of a recently developed tunneling kinetic model for random distributions of electron-hole pairs in luminescent materials.

1. Introduction

In some inorganic materials a rapid fading of the luminescence signals is observed experimentally within short times after irradiation, instead of within the long lifetimes predicted by standard kinetic models. This rapid fading process has been termed anomalous fading (AF), and is one of the most serious problems in TL and OSL dating [1,2].

Explanations of the AF effect have been based on various proposed models, such as the tunneling model [3–5], the localized transition model [6,7] and a model based on competition with radiationless transitions [8]. Currently, the most accepted explanations of AF are based on quantum mechanical tunneling from the ground state or from the excited state of the trap [9–13].

For the investigation and understanding of the anomalous fading effect it is desirable to find experimental conditions and sample preconditioning which would strongly influence the AF phenomena. Durango apatite is a natural material which is known to exhibit strong anomalous fading effects [14], with AF in this material exhibiting a remarkable resistance to a variety of experimental conditions and sample preconditioning. Kitis et al. [15] studied the effect of varying the heating rate during a TL measurement on the AF properties of this crystal [16,17], while Tsirliganis et al. [18] investigated the dependence of the AF of TL/OSL signals in this material on the occupancy of the recombination sites. While the influence of AF on the TL and OSL signals has been studied extensively for a variety of materials, the effect of AF on the isothermal TL signals has not been studied previously in a quantitative manner.

In a review article, Visocekas [19] discussed the nature and experimental importance of tunneling afterglow signals observed in a variety of materials. Baril [20] and Baril and Huntley [21] carried out detailed isothermal experiments in feldspars and showed that at prolonge times the isothermal signals follow a power law, with a power law coefficient very close to 1.

The general aim of the present work is to investigate the isothermal decay signals from Durango apatite, and the possible influence of AF on these isothermal signals. The specific goals of this work are:

- To analyze the isothermal luminescence signals using several standard methods of analysis.
- To study the effect of AF on isothermal signals. This was achieved by comparing isothermal signals measured immediately after irradiation, with signals measured 24 h after irradiation.
- To discuss the experimental results within the framework of a recently proposed model which is based on localized transitions and tunneling.

---

* Corresponding author. Tel.: +30 2310998175.
E-mail address: gkitis@auth.gr (G. Kitis).

http://dx.doi.org/10.1016/j.nimb.2013.12.003

2. Experimental procedure

2.1. Sample details and preparation

The sample for these experiments was a natural crystal of Durango apatite with dimensions of 8 × 4 × 3 mm. The single piece crystal was crushed gently with an agate mortar and grains of dimension 80–140 μm were obtained after sieving. The grains were annealed at 900 °C for 1 h, followed by rapid cooling to room temperature. This annealing treatment is necessary in order to empty all traps, which have been filled by the natural irradiation of the material. Previous work has shown that this annealing process does not influence the anomalous fading effect in Durango apatite [14].

Aliquots (sub-samples) with the same mass of 5 mg were attached to stainless steel disks. Each data point reported in this paper was the average of two measurements carried out on two different aliquots/disks.

2.2. Apparatus and measurement conditions

TL measurements were carried out using a Risø TL/OSL reader (model TL/OSL–DA–15), equipped with a ^{90}Sr/^{90}Y beta particle source, delivering a nominal dose rate of 0.075 Gy/s. A 9635QA photomultiplier tube with a combination of Pilkington HA-3 heat absorbing and Corning 7–59 (320–440 nm) blue filter were used for light detection. All measurements were performed in a nitrogen atmosphere with a low constant heating rate of 2 °C/s, in order to avoid significant temperature lag, and the samples were heated up to the maximum temperature of 500 °C. There are two experimental methods to study the decay of TL under stable temperature. (a) The Residual isothermal decay method (RID) in which the irradiated sample is post irradiation annealed in a furnace and then the residual TL glow-curve is measured. (b) The prompt isothermal decay (PID) method in which TL is measured directly while the sample is held to a stable temperature in the TL reader. The PID method is used in the present work.

2.3. Experimental protocols

The experimental procedure for the PID study of TL was performed according to the following protocol.

- Step 1: The previously annealed aliquot is irradiated with a test dose TD = 15 Gy, in order to populate the traps and centers.
- Step 2: TL measurement up to a temperature T at 2 °C/s. At this temperature, called T_{dec} the sample is left to decay thermally for 1000 s.
- Step 3: After the end of the decay period the sample is cooled down to room temperature.
- Step 4: Repeat steps 1–3 for a new aliquot and for a new decay temperature T_{dec}.

The prompt isothermal TL decay temperatures T_{dec} used in step 2 are shown in Fig. 1. The glow curve yields a small peak around 140 °C and the main TL peak is located at 310 °C. The T_{dec} values range from 160 °C up to 250 °C in steps of 10 °C. The starting T_{dec} of 160 °C was chosen to erase sufficiently the low temperature TL, and the highest T_{dec} of 250 °C is just below the temperature corresponding to half of the maximum TL intensity. Higher T_{dec} values were avoided for the following reasons. (a) The percentage of trapped electrons thermally released during the TL readout up to T_{dec} must be kept in a relatively low level, in order for the isothermal decay to represent accurately the decay of the whole population of trapped electrons. (b) The thermal decay constant is expected to depend very strongly on temperature T_{dec}, so that at temperatures close to, or higher than, the peak maximum temperature the isothermal decay will become extremely short lived and difficult to measure.

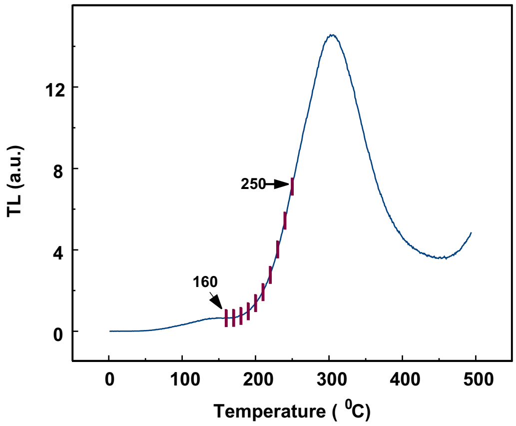

Fig. 1. Glow-curve of Durago apatite. The lines indicates the position on the glow-curve of the PID temperature used, in steps of 10 °C.

The above protocol was run twice. In the first run the TL measurement in step 2 was performed immediately after the end of irradiation in step 1. This means that the time left for anomalous fading is practically zero, so this case will be referred as “zero storage” case. In the second run the sample after the irradiation in step 1 was stored at room temperature for 24 h. During the 24 h storage period a substantial amount of anomalous fading has taken place, reducing the TL intensity by about 50% [16,17]. Longer storage times cause a substantial TL signal loss, making a PID experiment statistically poor.

The application of the experimental protocol produces two sets of prompt isothermal decay curves. In the “zero storage” set the isothermal decay curve will represent the thermal decay effect without a serious loss of TL signal due to anomalous fading. The second 24 h storage set of measurements will represent a substantial loss of TL signal due to the anomalous fading, and will be referred to as the “24 h storage” measurements.

3. Results

3.1. Analysis based on exponential functions

The decay curves obtained from the PID experiment for “zero storage” time are shown in Fig. 2(a). The decay curves for the “24 h storage” samples were very similar and are not shown here. Due to the very different intensity at each T_{dec}, the curves are normalized over the initial intensity at time $t = 0$. The results of Fig. 2(a) are unexpected, in the sense that they contradict well–established TL theories based on delocalized transitions through the conduction band. As is well known from TL kinetic theory [22,23], the decay constant λ (T) is expected to depend very strongly on T_{dec}. However, the PID curves in Fig. 2(a) do not follow this expected behavior.

An example of the expected typical behavior of experimental PID curves in dosimetric materials is given in Fig. 2(b) for the case of Al_2O_3:C. As an additional example based on simulation, thirteen (13) pairs of (E, s) values were found which would give the same peak maximum temperature T_m at 300 °C. Using these pairs of parameters, the PID curves were simulated at the temperatures of Fig. 1. A typical result of the simulation is shown in Fig. 2(c), showing clearly a different behavior than the experimental data in Fig. 2(a).