Radiation Measurements 84 (2016) 15–25

Contents lists available at ScienceDirect

Radiation Measurements

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

Prompt isothermal decay of thermoluminescence in MgB_4O_7:Dy, Na and LiB_4O_7:Cu, In dosimeters

G. Kitis [a, *], G.S. Polymeris [b], I.K. Sfampa [a], M. Prokic [c, 1], N. Meriç [b], V. Pagonis [d]

[a] Aristotle University of Thessalonians, Physics Department, Nuclear Physics and Elementary Particles Physics Section, 54124, Thessaloniki, Greece
[b] Institute of Nuclear Sciences, Ankara University, 06100, Besevler, Ankara, Turkey
[c] Institute of Nuclear Sciences, Vinca, P.O. Box 522, 11001, Belgrade, Serbia
[d] McDaniel College, Physics Department, Westminster, MD 21157, USA

HIGHLIGHTS

- Isothermal decay of TL in MgB_4O_7:Dy, Na and LiB_4O_7:Cu, In is studied.
- The TL of LiB_4O_7:Cu, In is due to delocalized transitions.
- The TL of MgB_4O_7:Dy, Na is due to tunneling transitions.
- Recent TL expressions for delocalized and localized transitions are used to explain the results.

ARTICLE INFO

Article history:
Received 22 August 2015
Received in revised form
4 November 2015
Accepted 13 November 2015
Available online 1 December 2015

Keywords:
Thermoluminescence
Isothermal thermoluminescence
Lambert W function
TL-OSL dosimetry
Tunneling recombination

ABSTRACT

According to standard delocalized kinetic models of thermoluminescence (TL), when an irradiated sample is held at a high temperature T, the isothermal TL signal will decay with a characteristic thermal decay constant λ which depends strongly on the temperature T. This prediction of standard delocalized kinetic theory is investigated in this paper by studying two TL dosimeters, MgB_4O_7:Dy, Na and LiB_4O_7:Cu, In (hereafter MBO and LBO correspondingly). In the case of LBO it was found that the thermal decay constant λ of the main dosimetric TL peak follows exactly the predictions of standard delocalized kinetic theory. Furthermore, the thermal activation energy of the main peak evaluated by the isothermal decay method is in full agreement with values obtained from initial rise and glow curve fitting methods. However, in the case of MBO it was found that the thermal decay constant λ varies little with the isothermal decay temperature T. In order to explain these unusual results for MBO, the TL glow curves and isothermal decay curves were analyzed using analytical expressions derived recently from a radiative tunneling recombination model. Based on the different behavior of the two TL dosimeters, it is suggested that the isothermal decay of TL at high temperatures can be used to discriminate between radiative delocalized recombination and radiative localized recombination processes.

1. Introduction

Magnesium borate (MgB_4O_7) is a host matrix of interest for thermoluminescence (TL) dosimetry of ionizing radiation (McKeever et al., 1995). This material has been reported since the 1980s and 1990s as being attractive for dosimetry because of its low effective atomic number (Z_{eff}~8.4), which implies a small photon energy dependence (Prokic, 1980; Prokic, 1993; Prokic, 2007, 1993; Driscoll, 1981; Furetta et al., 2000; Lochab et al., 2007). Moreover, the possibility of developing neutron dosimeters based on these materials has also been suggested, because of the high neutron capture cross-section for ^{10}B (McKeever et al., 1995; Prokic, 1993; Price et al., 1998). Most of the literature on the TL properties of MgB_4O_7 is focused on its dosimetric properties when a variety of dopants is used.

Despite this high interest in the past, there has been no systematic study on the luminescence properties of MgB_4O_7. Recently Yukihara et al. (2014a) performed a systematic study of the TL properties of magnesium borate doped with lanthanides, in order to elucidate the thermally stimulated and recombination processes

* Corresponding author.
E-mail address: gkitis@auth.gr (G. Kitis).
[1] Dr M. Prokic has passed away last year.

http://dx.doi.org/10.1016/j.radmeas.2015.11.002
1350-4487/

in this material. Their results showed that different dopant were associated with TL peaks at different temperatures, involving different trapping mechanisms depending on the dopant element. These results stand in good agreement with Karali et al. (2002), who reported spectrally resolved TL curves which differ for MgB_4O_7 dosimeters variously doped with one or two rare earth (RE) dopant. These results provided further evidence that RE ions could form part of large complex defects, and/or long range interactions involving the charge trapping and recombination processes in TL.

Lithium tetraborate $Li_2B_4O_7$ is also a material of interest for TL dosimetry (McKeever et al., 1995), which has attracted a great deal of attention as a radiation resistant optical material and as a tissue-equivalent material for radiation dosimetry (Z_{eff}~7.25) (Park et al., 2003; Prokic, 2001a, b). In addition, lithium borate based crystals are convenient materials for neutron dosimetry due to the presence of Li and B (Tiwari et al., 2010). The original undoped material had some disadvantages like poor sensitivity, hygroscopic nature and relatively large fading rate. To improve its sensitivity, different dopant and preparation methods have been introduced by several researchers (Furetta et al. (2001); Prokic (2001a, b)). Single crystals, polycrystals and glass forms of Lithium tetraborate doped with rare-earth, copper and manganese ions are used effectively as TL dosimeters (Takenaga et al., 1980; Chandra and Bhatt, 1981; Kutomi and Takeuchi, 1996; Martini et al., 1996; Prokic, 2001a). The TL properties of $Li_2B_4O_7$ tend to vary with the synthesis method, preparation, dopant, and dopant concentrations (Wall et al., 1982). Generally, the TL curves tend to have a low temperature peak around 100 °C and a main dosimetric peak around 200 °C (Takenaga et al., 1980; Wall et al., 1982; Furetta et al., 2001; Prokic, 2001a, b). In some cases, a high temperature peak around 300 °C has been also reported (Kutomi and Takeuchi, 1986).

Both materials have regained scientific interest during the last few years, mostly for their possible use for passive temperature sensing using thermoluminescence (Yukihara et al., 2014a; Yukihara et al., 2014b; Doull et al., 2013, 2014), due to their low light sensitivity and, therefore, of potential application as a temperature sensor.

Among the various methods used to evaluate the kinetic parameters of a trap responsible for a TL glow-peak is the isothermal decay method (Chen and McKeever, 1997). There are two commonly used experimental versions of the isothermal decay technique. The first version is 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. The second version is the prompt isothermal decay (PID) in which TL is measured directly while the sample is held at a stable temperature in the TL reader. The basic measured parameter in an isothermal experiment is the thermal decay constant $\lambda=1/\tau$, where τ represents the thermal lifetime. According to standard TL models involving delocalized transitions, in the case of first order kinetics the parameter λ is expected to vary exponentially with the sample temperature T, according to $\lambda=s\,exp(-E/KT)$. In this expression s is the frequency factor, E is the thermal activation energy and T is the absolute temperature of the sample.

In a recent study Sfampa et al. (2014) reported a PID study of Durango apatite, which is a material exhibiting strong anomalous fading (AF). AF consists of the anomalous loss of TL and optically stimulated luminescence (OSL) signals after irradiation, which contradicts the predictions of standard luminescence kinetic theory, and is commonly attributed to a quantum tunneling effect (Wintle, 1977; Visocekas et al., 1976; Kitis et al., 1991). Sfampa et al. (2014) reported that the decay constant λ of the PID curves in Durango apatite was practically independent of isothermal temperature T, instead of the strong dependence expected from standard delocalized kinetic models. These authors analyzed successfully the PID curves of their experiment using analytical expressions for PID derived by Kitis and Pagonis (2013). These analytical expressions were based on the recently developed tunneling kinetic model of Jain et al. (2012).

The PID results of Sfampa et al. (2014) confirmed the prevalence of radiative tunneling recombination processes in Durango apatite. It is interesting to investigate whether this behavior is restricted in Durango apatite, or whether it exists in other natural material like feldspars, which also suffer from AF effects. Equally interesting is to investigate if a similar PID behavior exists in synthetic TL dosimeters.

The aims of this work are to investigate the behavior of PID signals in MgB_4O_7:Dy, Na and LiB_4O_7:Cu, In dosimeters, and to examine whether the PID behavior can be used to discriminate between delocalized radiative recombination and localized radiative tunneling recombination processes.

2. Experimental procedure

2.1. Sample details

The samples used in these experiments were *MgB_4O_7:Dy,Na* (hereafter termed as MBO) and *LiB_4O_7:Cu,In* (hereafter termed as LBO) obtained from the same production batch used by Furetta et al. (2000) and Kitis et al. (2000). Furetta et al. (2000) presented a complete dosimetric characterization of these materials, whereas Kitis et al. (2000) determined their characteristic kinetic parameters.

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 0.105 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 350 °C.

The PID method of isothermal luminescence is used in all experiments described 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 0: Test dose and TL measurement up to $T = 350$ °C for LBO and $T = 450$ °C for MBO at 2 °C/s.
- Step 1: The previously annealed aliquot is irradiated with a test dose TD = 1 Gy, in order to populate the traps and centers.
- Step 2: TL measurement up to a temperature T_{dec} at 2 °C/s. The sample is left to decay thermally for 500 s at this temperature, and the isothermal decay signal is measured.
- Step 3: After the end of the decay period, the sample is cooled down to room temperature.
- Step 4: TL measurement at 2 °C/s in order to obtain the residual TL glow curve (R-TL).
- Step 5: Repeat steps 1–4 for a new decay temperature T_{dec}.
- Step 6: Repeat step 0.

The peak maximum temperature (T_m) of LBO is 200 °C, whereas the corresponding value of T_m for MBO is 190 °C. In the case of LBO the sample was pre-heated before step 2 at 2 °C/s, up to a