Radiation Measurements 47 (2012) 236–240

Contents lists available at SciVerse ScienceDirect

Radiation Measurements

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

Technical report

Preparation and TSL studies in Tb activated $LiMgPO_4$ phosphor

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ARTICLE INFO

Article history:
Received 15 October 2010
Received in revised form
2 November 2011
Accepted 31 December 2011

Keywords:
Thermally stimulated luminescence
Kinetic parameters
Peak shape method

ABSTRACT

$LiMgPO_4$:Tb^{3+} phosphor was synthesized by solid state reaction. The thermally stimulated luminescence (TSL) glow curve of Tb doped $LiMgPO_4$ exhibits a main TSL peak at 170 °C with shoulders at 100 and 260 °C on either side of this peak. The TSL sensitivity of the phosphor was found to be about 2.5 times that of $CaSO_4$:Dy phosphor. TSL emission and photoluminescence (PL) studies show that Tb^{3+} ion acts as luminescence centre in this phosphor. The kinetic parameters, namely activation energy (E) and frequency factor (s) associated with the main glow peak have been determined using peak shape method. The activation energy and frequency factor obtained are 1.35 ± 0.03 eV and (6.53 ± 0.43) × 10^{14} s^{-1} respectively. The paper discusses the dosimetric characteristics like dose response, fading, energy response and minimum detectable dose and results thereof.

1. Introduction

Thermally stimulated luminescence (TSL) technique is extensively used in radiation dosimetry. The phosphors which are used commonly are LiF:Mg,Ti (TLD-100), $CaSO_4$:Dy, LiF:Mg,Cu,P etc. $CaSO_4$:Dy has good TSL sensitivity but has poor tissue equivalence. LiF-TLD-100 has good tissue equivalence but its TSL sensitivity is about 30 times less than that of $CaSO_4$:Dy (McKeever, 1985). LiF:Mg,Cu,P is a sensitive TSL material which is tissue equivalent also. However, its response depends critically on the heat treatment (Sahare and Moharil, 1990). It has been reported that in the commercially available LiF:Mg,Cu,P TLD's an increase of 1 °C in the annealing temperature above 240 °C reduce the TL sensitivity significantly (Luepke et al., 2006). It has been found that thermal treatment needed for the encapsulation of LiF:Mg,Cu,P in Teflon for making a commercial TLD badge reduces the TSL sensitivity by more than a factor of 2 (Moscovitch et al., 2006). Kim et al. (2008) have reported the development of LiF:Mg,Cu,Si phosphor which could overcome the drawback of LiF:Mg,Cu,P phosphor. During the course of our investigation of TSL properties in Li based compounds we have found a new phosphor $LiMgPO_4$:Tb having interesting TSL properties that could be used for dosimetric applications. The antiferromagnetic properties and nuclear magnetic resonance (NMR) studies on $LiMgPO_4$ doped with Fe have been reported (Goni et al., 2001, 1998). No report is available in the literature on the TSL properties of $LiMgPO_4$:Tb. Dhabekar et al. (2011) have synthesized $LiMgPO_4$ based phosphor with Tb and B as dopants and observed it to be a sensitive OSL phosphor. However its TSL sensitivity was found to be much less than that of $LiMgPO_4$:Tb phosphor. Dosimetric characterization of new TSL materials also include the determination of kinetic parameters, order of kinetics b, activation energy E and frequency factor s (Kitis et al., 2000). In this paper we report the preparation, TSL properties, dosimetric characteristics and the kinetic parameters of $LiMgPO_4$:Tb phosphor.

2. Materials and methods

$LiMgPO_4$:Tb phosphor was prepared by solid state reaction between LiOH, $Mg(NO_3)_2 \cdot 6H_2O$, Tb_4O_7 and $NH_4H_2PO_4$ in air (Goni et al., 1996). The structure of the as-prepared sample was analyzed by Rigaku D/max-II X-ray diffractometer using monochromatic $CuK_{\alpha 1}$ ($\lambda = 1.5405$ Å) radiation in the 2θ range of 10–80°. The samples were ground and sieved to get the powder of grain size in 75–210 μm range. All the experiments were carried out using this sample. Calibrated ^{60}Co gamma chamber having a dose rate of 3 Gy/min was used for irradiating the phosphor samples to gamma rays unless otherwise specified. For the determination of the energy response, the photon irradiations were performed at the photon energies of 10, 30, 64, 83, 101, 120, 167, 213 and 662 keV using an X-ray generator and ^{137}Cs source (662 keV). A locally made PC based TSL reader (Seethapathy et al., 1999) with 9125A photomultiplier tube (PMT) having S11 response was used for TSL measurements. TSL glow curves were recorded one day after the exposure. 5 mg of sample was used to record the TSL glow curve each time with the heating rate of 5 °C/s unless otherwise specified.

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doi:10.1016/j.radmeas.2011.12.013

To record the fading, exposed samples were stored in dark and glow curves were recorded at regular intervals.

TSL emission spectrum was recorded at 100 °C so that fading of the TSL signal during the recording is not appreciable. The TSL emission spectrometer having Hamamatsu R 928 PM tube was used for this purpose.

The room temperature photoluminescence (PL) spectra were recorded on F-4500 Fluorescence spectrophotometer (Hitachi, Japan) with 150 W Xenon lamp, in the wavelength range of 200–800 nm. Slit width for excitation and emission was 2.5 nm each.

The kinetic parameters of the main TSL peak were determined using peak shape method (Chen, 1969). This method is valid for any order of kinetics. The expression for activation energy is given by

$$E_\alpha = c_\alpha\left(kT_m^2/\alpha\right) - b_\alpha(2kT_m)$$

Where α stands for τ, δ and ω; in which $\tau = T_m - T_1$, $\delta = T_2 - T_1$ and $\omega = T_2 - T_1$. T_1 and T_2 are the temperatures at the half of the maximum intensity on the ascending and descending parts of the TSL peak respectively. T_m is the peak temperature of the TSL peak. k is the Boltzmann constant. The empirical expressions for b_α and c_α are:

$$c_\tau = 1.51 + 3.0\left(\mu_g - 0.42\right),$$

$$b_\tau = 1.58 + 4.2\left(\mu_g - 0.42\right),$$

$$c_\delta = 0.976 + 7.3\left(\mu_g - 0.42\right), \quad b_\delta = 0,$$

$$c_\omega = 2.52 + 10.2\left(\mu_g - 0.42\right), \quad b_\omega = 1,$$

where $\mu_g = \delta/\omega$. b can be evaluated from the measured value μ_g from the graph of μ_g against order of kinetics b given by Chen (1969). Once the activation energy and order of kinetics have been determined, the frequency factor s can be estimated by

$$s = \left(\beta E/kT_m^2\right)\exp(E/kT_m)[1 + (b-1)2kT_m/E]^{-1}$$

β is the heating rate (Chen, 1969; Halperin and Branner, 1960).

To obtain the kinetic parameters the TSL reading were carried out on powder samples at a heating rate of 1 °C/s. The low temperature peak was removed by thermal cleaning.

3. Results and discussion

3.1. X-ray diffraction

$LiMgPO_4$ is a diamagnetic member of the olivine type family of lithium phosphates (Goni et al., 1996). Fig. 1 shows the XRD pattern of $LiMgPO_4$ phosphor along with the standard XRD pattern (JCPDS card no. 32-0574). The XRD pattern shows the formation of pure $LiMgPO_4$ phase. The addition of the dopant has no effect on the XRD pattern. The structure of $LiMgPO_4$ is orthorhombic, with the space group P_{nma}, and with the lattice parameter 'a' about twice that of 'b' and 'c'.

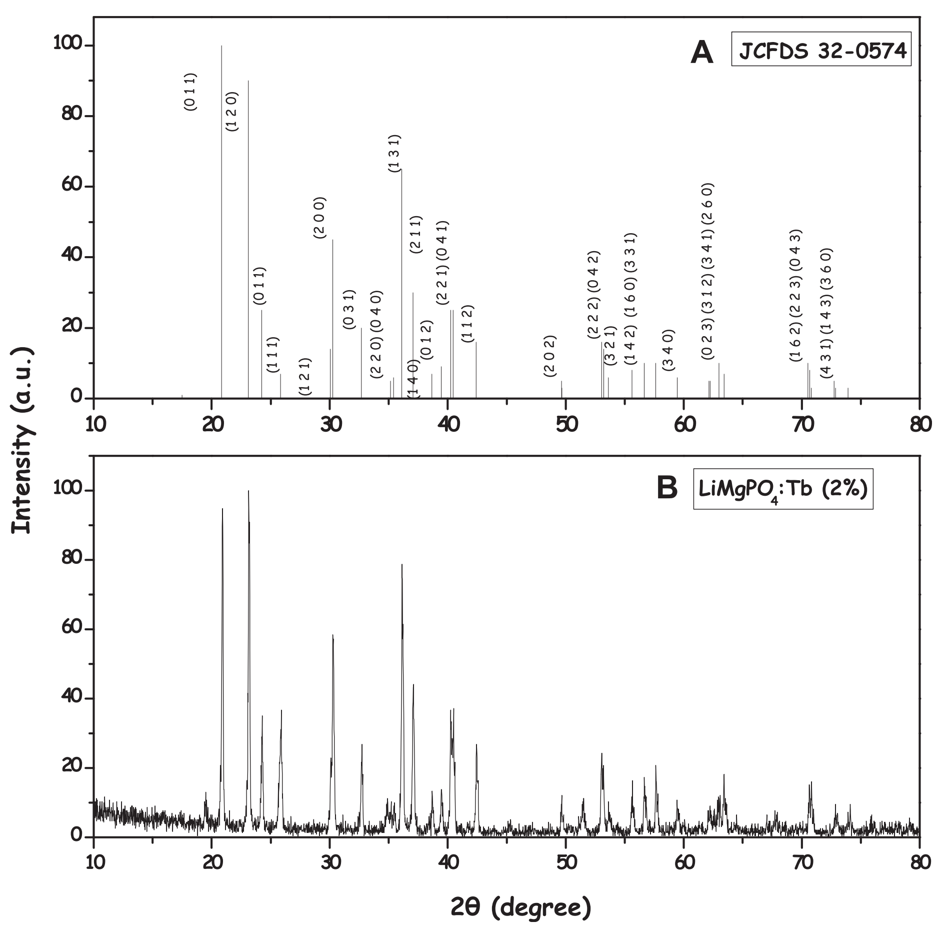

Fig. 1. XRD patterns of (A) JCPDS card no. 32-0574 (B) $LiMgPO_4$:Tb.