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Thermoluminescence properties of Ce-doped MgB_4O_7 phosphor

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In this study, the polycrystalline samples of undoped and different amount of Ce-doped MgB_4O_7 were firstly prepared by a melting point method in air conditions with high temperature and then their thermoluminescence properties were reported. The most effective amount of dopants on MgB_4O_7 is found to be 1% Ce. The main dosimetric properties which are namely TL sensivity, fading, linearity and re-usability of 1 % Ce-doped MgB_4O_7 were examined in detail. The trap parameters namely order of kinetics *(b)*, activation energy *(E)* and frequency factor *(s)* associated with the glow peaks in beta irradiated undoped and Ce doped MgB_4O_7 samples were also obtained by computer glow curve deconvolution (CGCD) method.

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Keywords: Magnesium tetraborate, thermoluminescence dosimetry

1. Introduction

The thermoluminescence (TL) studies of some borate compounds are of interest because of their near tissue equivalent absorption coefficient, low cost and easy handling process and therefore it is particularly suited for applications in radiation dosimetry, especially radiation therapy and clinical applications. Therefore, the TL studies of lithium borate compounds were started in 1967 by the work of Schulman et al. [1] and then, detailed TL studies on various alkali and alkaline earth tetra borates, especially on the lithium and magnesium borate compounds, were continued up to present times [2–6]. It is well known that the most of pure borates efficiency are very low. The dopants have to be added in order to increase their TL output. According our knowledge the TL properties of magnesium tetra borate activated by Ce with melting point method have not been reported up to now. In this study, the TL properties of pure and Ce-doped MgB_4O_7 were reported for the first time.

2. Experimental

In this study, polycrystalline samples undoped and Ce-doped MgB_4O_7 are synthesized in an inert atmosphere by melting point method. Firstly, the magnesium tetraborate compounds were synthesized by mixing magnesium carbonate ($MgCO_3$) and boric acid (H_3BO_3) which were purchased from Merck company in stoichiometric ratio and the mixture was heated at 300 ^oC in a porcelain crucible for 4 hr to remove carbonates and water from the samples. Then, the desired amount of Ce is added to mixture, and sample heated to 900 ^oC for 10 hr which assures a complete crystallization. Then samples cooled to room temperature rapidly up to 500 ^oC. The samples are taken out of furnace and dried in a desiccators. The resultant glassy mass was ground in an agate mortar again then sintered at 400 ^oC for 4 hour and sieved to obtain 100-200 mesh powder [7-8]. The samples were irradiated at room temperature by a ^{90}Sr-^{90}Y beta-source which delivering about 0.9 Gy/min. The glow curve measurements were made using a Harshaw TLD System 3500 Manual TL Reader. Glow curve readout was carried out on a platinum planchet at a linear heating rate of 1 $^oC/s$ up to 400 ^oC. The recorded glow curves were analyzed using computer glow curve deconvolution (CGCD) program. In order to determine the identity of produced samples, a Rigaku D/Max-I IB X-ray diffractometer at 40 kV and 20 mA with Cu Kα1 (λ=1.5405 Å) radiation were used. The X-ray diffraction measurements were taken at the interval of Bragg angle 2θ ($5^o \leq 2\theta \leq 80^o$) at scanning rate of 4^0 per min at room temperature. Inter plane spacing (d),

lattice parameters of unit cell and miller indexes are found from 2θ values by using a computer program package called ''POWD'' [9-10]. It is an interactive powder diffraction interpretation indexing program version 2.

3. Results and discussions

X-ray diffraction pattern of 1% Ce-doped MgB_4O_7 is shown in Fig.1. Diffraction lines are indexed by using Powd program and it was found that the magnesium borate prepared by melting method has monoclinic crystal structure. The lattice parameters of unit cell were found to be *a*=6,45 Å, *b*=3.74Å, *c*=5.59 Å and unit cell volume *V=125,8 Å*^{3}.

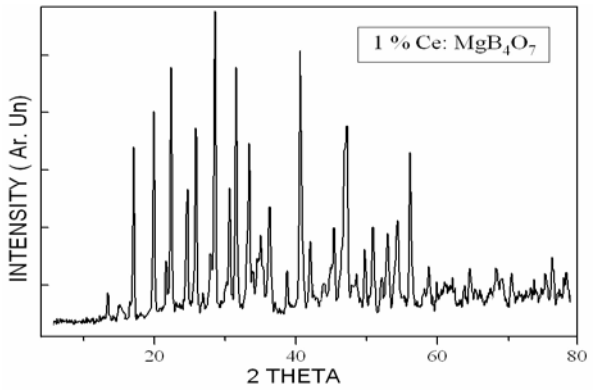

Fig. 1. XRD pattern of MgB_4O_7:1%Ce which synthesized with melting method

The glow curves of undoped MgB_4O_7 samples after different dose levels were shown in Fig.2. As seen, it has been acquired two sharp and separated peaks at about 170 ^oC and 290 ^oC. The intensity of 290 ^oC is the strongest. The appearance of two different glow peaks indicates that two different species of traps are being activated within the particular temperature. On the other hand, the TL glow curve of Ce-doped MgB_4O_7 crystal exhibits one strong TL glow peak at around 240 ^oC and two shoulder peaks to this peak, one of them at the low temperature side about 170 ^oC and the other at the high temperature side about 300 ^oC. Therefore, we can say that the concentration of Ce-doping is greatly altered the distributions of traps produced by β-ray irradiation in MgB_4O_7 host. The TL dose response curve of Ce-doped MgB_4O_7 was studied as a function of dose levels between 2.25 Gy and 36 Gy and the resulting dose response curve is shown in inset of figure 3. The resulting dose response curves are an average of three measurements. As seen, the intensity of main glow peak (T_m≈240 ^oC) of Ce-doped MgB_4O_7 is linearly increased with increasing applied dose levels in the range of investigated region with no observed superlinearity and saturation effects.

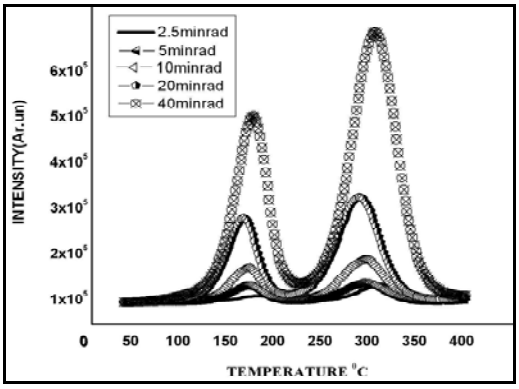

Fig. 2. The influence of the ^{90}Sr-^{90}Y β-ray as a function of irradiation time on the of MgB_4O_7 phosphor at room temperature.

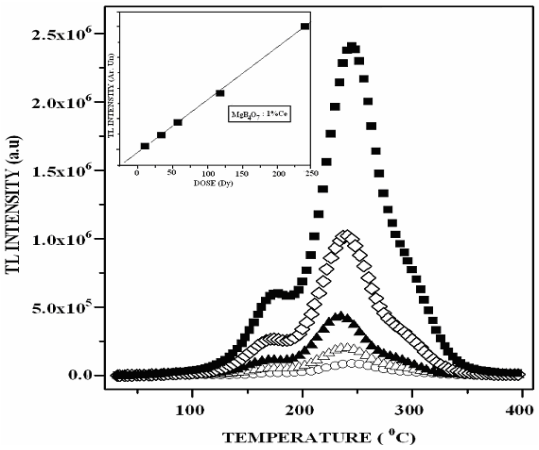

Fig. 3. The influence of the ^{90}Sr-^{90}Y β (beta)-ray as a function of irradiation time on the 1% Ce doped MgB_4O_7 phosphor at room temperature. The inset shape indicates linearity range of the TL dose response of main glow peak (T_m≈240 ^oC) of this material. [(○): 2.25 Gy, (Δ): 4.5Gy, (▼): 9 Gy, (◊): 18 Gy, (■): 36 Gy].

For sample to be useful in dosimetry, the TL intensity of the glow peaks, at least its dosimetric peak(s), should be stable and no fade upon storage after exposure. The experimental glow curves acquired during fading study showed that the peak temperature at the low temperature (170 ^oC) slightly shifted towards higher values as the elapsed time from irradiation increased shown in figure 4. A small fading acquired for this peak about 5 % but there isn't an

important fading in the main glow peak. This effect shows that MgB_4O_7 phosphor can be useful for personal dosimetry.

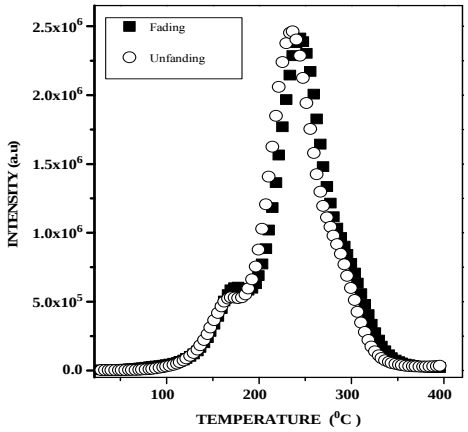

Fig. 4. The comparison of glow peaks of 36 Gy irradiated MgB_4O_7:1%Ce phosphor (○) directly and (■) that of after stored at dark room for 24 hour.

If the sensitivity of a sample does not change after several cycles of exposure and readouts, it can be considered as a good phosphor. Thus the produced MgB_4O_7 and 1%Ce doped samples were also tested for reusability. The reference samples were exposed to beta rays for 10 min and then its glow curves were recorded between room temperature and 400 °C. These procedures were repeated seven times and it was observed that the sensitivity of TL glow peaks have good stabilities during the successive readings. The obtained repeatability of the samples for seven cycles is always below 10% (see fig.5).

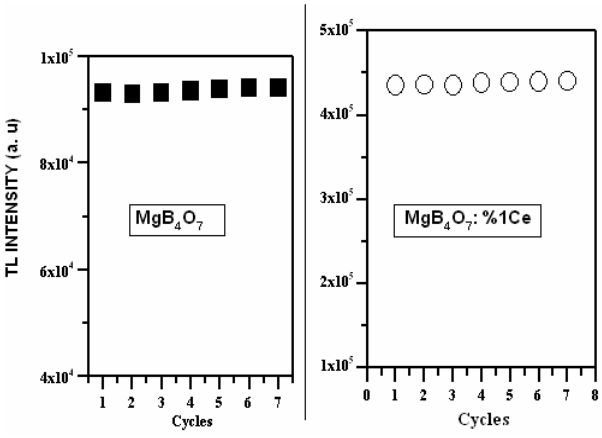

Fig. 5. Reproducibility of undoped (■) and 1% Ce-doped MgB_4O_7 (○) samples thought seven times repeated annealing-irradition-readout.

The glow curves of Ce-doped MgB_4O_7 were also analyzed by the computerized glow curve deconvolution (CGCD) method to obtain the number of glow peaks and their kinetic parameters. This method has become very popular to obtain the number of glow peaks in the complex glow curves and their kinetic parameters for the last three decades [11]. It is also commonly used in the study dosimetric properties of TL dosimeters. On the other hand, the previously published studies have shown that the determination of E_a and s mainly depends on the prior knowledge of b and the exact number of glow peaks which were used in the deconvolution program [12-13]. Therefore, to form an opinion about the b of all individual glow peaks in the glow curve structure of examined samples, the results of glow curves after different dose levels were firstly utilized in the current study. This is a simple test for the first-order kinetics. In TL theory [14], the peak temperatures of glow peaks are expected to change only with heating rate for b=1. Hence, for a constant heating rate, the peak maximum should not be affected by other experimental parameters and should be fairly constant within the limit of experimental uncertainties. However, for $b\neq1$ and below the trap saturation points {n_o(concentration of trapped electrons)<N_t (concentration of traps)}, it is generally received that the peak temperatures are shifted to the lower temperature side with increasing dose levels. It is seen from Fig.3 that the structures of the TL glow curves of Ce-doped MgB_4O_7 samples and the peak temperatures of glow peaks at around 170 and 240 °C were not changed with increasing dose levels. On the other hand, the peak temperature of glow peak 3 is slightly shifted to the low temperature side with increasing dose levels. These results indicate that the glow peaks 1 and 2 should be considered under the first-order kinetics whereas the peak 3 under the general-order kinetics. As a result, it was concluded that the glow curve of MgB_4O_7:Ce is well described by a linear combination of at least three closely overlapped glow peaks (P1, P2 and P3) corresponding to peak temperatures at around 170, 240 and 275 °C, respectively. An analyzed glow curve of Ce-doped MgB_4O_7 measured after 36 Gy irradiation at RT is shown in figure 6 along with the components obtained from CGCD. The obtained kinetic parameters were given in Table1. The statistical error of these parameters deriving from the analysis of glow peaks after different irradiation doses is of the order of 10%.

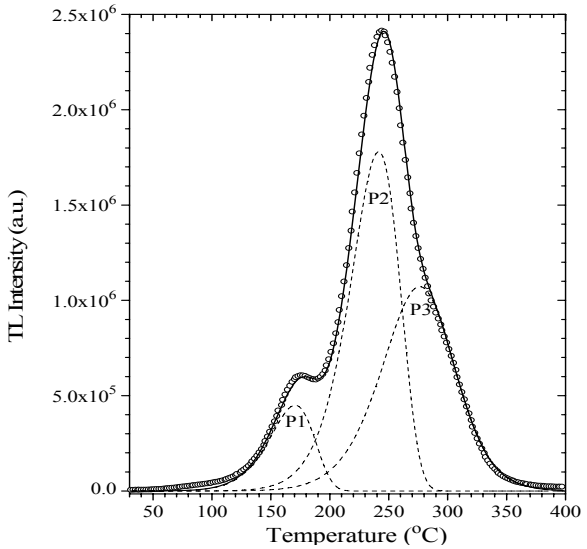

Fig. 6. An analyzed glow curve of Ce-doped MgB_4O_7 measured after 36 Gy irradiation at room temperature at a heating rate of 1°C s^{-1}.

Table 1. The trapping parameters of Ce-doped MgB_4O_7 samples calculated by the CGCD method (Heating Rate= 1 °C/s).

Trapping Parameters	Glow Peaks		
	Peak 1	Peak 2	Peak 3
T_m (°C)	170	240	275
E_a (eV)	0.86	1.06	0.98
ln(s) (s^{-1})	19.6	20.8	17.5
b	1.00	1.00	1.45

Comparison with data concerning undoped and Ce-doped MgB_4O_7 allows for identification of the contributions of impurities to the glow curves. The recent investigations showed that, besides metal impurities such as Cu and Ag-ions, rare earth dopants (which are well known as very efficient activators in other materials, for example in alkali halide crystals or in oxides) originate good TL features in borate compounds [15-16]. In general, the undoped samples, whose emission can be related to intrinsic defects or to some unwanted impurity in the starting powder, exhibits a poor TL efficiency which seems to be insufficient for reliable measurements in the low dose range measurements. On the other hand, the TL signal was steadily increased after incorporation of Ce-ions, as expected. In addition, the glow curve shape of MgB_4O_7 is highly changed after the doping of Ce-ions. This result indicates that there are large interactions between intrinsic defects and Ce-ions. However, further work is necessary to clarify details of the defect and glow curve structure in this material after beta irradiation.

4. Conclusions

The polycrystalline of Ce-doped MgB_4O_7 compounds were synthesized with melting method in air condition successfully. The growth material is quite suitable for use in high dose measurements in medical or industrial area between 2.25 Gy and 36 Gy due to its good linear dose response during this range, its low fading and its reusability after many experimental cycles. Generally supralinearity is a characteristic of a large variety of TL materials, so TL linearity in the high dose region, over 1 Gy, is exceptionally rare. However, the result suggested in the given study that the main peak of Ce-doped of MgB_4O_7 was shown that the dose response of this peak is quite linear with applied dose levels therefore it can be safely used as dosimetric material in the medical applications.

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