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Mechanoluminescence Characterization of Gamma Irradiated TB Doped $Mg(VO_3)_2$ Phosphors

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Abstract: ***Rare earth doped materials attracted much attention of the researcher due to characteristics emission in the visible region. In present investigation the ML intensity of γ-irradiated Tb doped divalent metal metavanadate phosphors have been studied. It has been found that single peak is observed in Tb doped $Mg(VO_3)_2$ phosphors relative to the ML intensity versus time curve. It is also observed that ML intensity increases with mass of the piston dropped on to the samples without any considerable change in peak positions, for all the samples. We have also observed that, the total ML intensity increases with impact velocity.The ML intensity is found optimum for the sample having 0.05 mol% concentration of Tb. ML intensity is found optimum for a particular concentration(0.05 mol%) of Tb then decrease with further increase in dopant concentration. ML intensity also increases with γ-dose given to the samples and seems to saturate for higher γ-doses. ML spectra of Tb doped divalent metal metavanadate phosphors show the characteristic emission of rare earth impurities. In Tb doped metavanadate phosphors a single peak at 482 nm was due to $^5D_4$-$^7F_6$characteristics emissionof Tb^{3+} ions.The prepared samples are characterized by x-ray diffraction XRD, scanning electron microscopy SEM and photoluminescence PL.***

Keywords: Phosphor, X-ray diffraction XRD, scanning electron microscopy SEM and photoluminescence PL and solid state reaction method

1. Introduction

Mechanoluminescence (ML) is an interesting luminescence phenomenon, which is caused by mechanical stimuli such as grinding, cutting, collision, striking and friction [1]. It can convert mechanical energy into visible light efficiently. The ML sensor to detect environmental stress by emitting light is expected to be used widely in various applications such as the forecasting of an earthquake, the damage detection of an air plane or car [2, 3]. In particular, the ML sensors have been found that they can be used as self-powered luminescence sensors [4]. The phenomenon of ML links the mechanical, electrical, spectroscopic and structural properties of solids. This technique offers a number of interesting possibilities such as detection of cracks in solids and for mechanical activation of various traps present in the solid.

The introduction of rare earths has resulted in a drastic improvement of the performance of luminescent devices based on these phosphors. Many efficient lasers are developed in vanadates host crystals in recent years [5-9]. The systematic study on ML and PL properties of magnesium metavanadate has not been investigated so far. Present paper discussed the comparison of ML properties of γ-irradiated Tb activated $Mg(VO_3)_2$ with their PL properties may help to understand the basic mechanism of luminescence in these materials.

2. Materials and Method

Pure and Tb doped $Mg(VO_3)_2$ phosphors of different impurity concentrations were prepared by solid state reaction method. Analargrade (99.9% pure) powder used as starting material. Requisite amount of host compound like $Mg(NO_3)_2.6H_2O$and NH_4VO_3 were mixed thoroughly in 1:2 mole proportion for different samples. In case of doped samples known amount of impurity Tb_4O_7 (from 0.05 to 1mole%) were added to above mixture and then transferred to J-mark porcelain crucibles. These powders were annealed in a high temperature muffle furnace by slowly raising the temperature to about 400^0C for 4 hours in air and heated at 650^0C for 12 hours then cooled to room temperature. The resulting compounds were crushed again and heated up to 1 hour at 650^0C, then quenched to room temperature. In present investigation vanadates based phosphors were prepared and XRD data of $Mg(VO_3)_2$ measured by using X-ray diffractometer and shown in below (figure A) respectively. The data matched well with the standard data (JCPDS) of corresponding compounds.

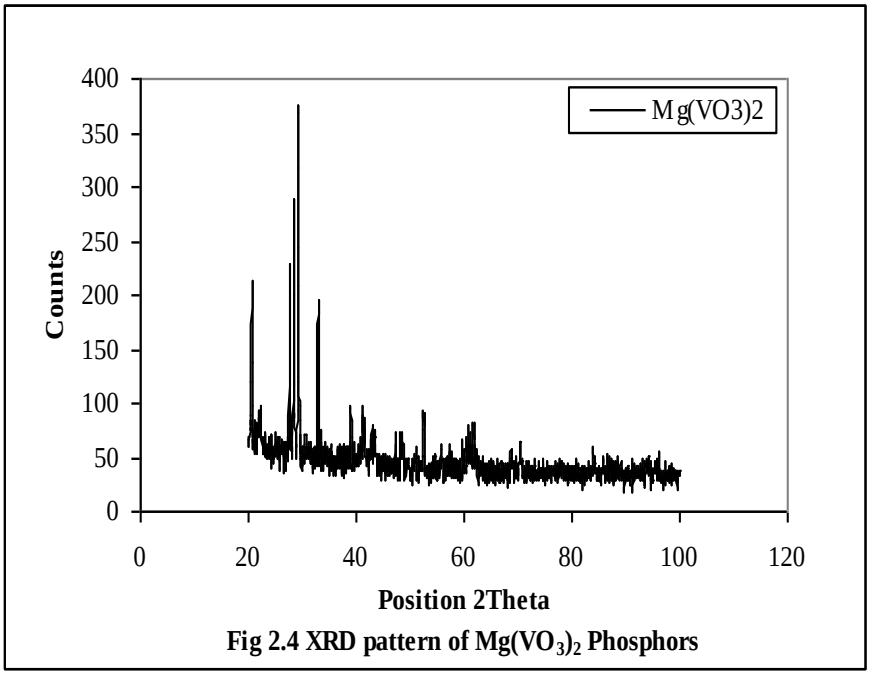

Fig 2.4 XRD pattern of $Mg(VO_3)_2$ Phosphors

Figure A

Samples were exposed to gamma rays using ^{60}Co source having the exposure rate 0.93kGy/h. The ML glow curves were recorded by the routine ML unit. Two milligrams of sample was used every time for recording the glow curves. The ML was excited impulsively by dropping a fixed mass load on to the sample. The luminescence was monitored by a RCA-931 photomultiplier tube positioned below the transparent Lucite plate and connected to storage oscilloscope (SCIENTIFIC SM-340). The ML spectra were recorded using a series of optical band pass filter. Seven different filters were used and all the filters have bandwidth in the range 20 to 40nm and with transmission 30 to 70%. Similarly, PL spectra of samples were recorded by using fluorescence spectrometer (SHIMADZU RF-1305 PC). Emission was recorded using a spectral slit width of 1.5nm.The scanning electron microscope (SEM) is a significant scientific technique for characterization. The microphotography of solid bulk specimen can be examined, as scan surface where roughness or other characteristics render their observation extremely difficult or impossible by means of a conventional transmission electron microscope, using either direct or extraction replica methods. Results show that the powder was agglomerated and particle size varies from 500nm to 5 µm. Their average particle size was found to be about 2.5µm. The narrow particle size distribution and the fine particles observed in our sample are necessary requirements of a good phosphor material(shown in figure B).

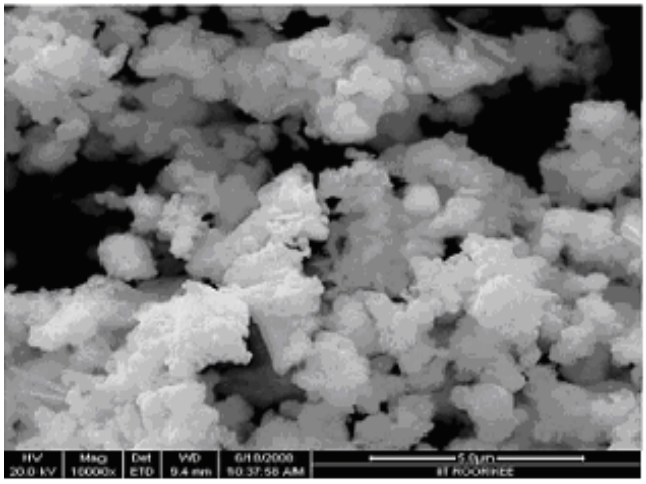

Figure B: SEM picture of $Mg(VO_3)_2$ samples

3. Results

Figure 1 shows the time dependence of ML intensity of γ-irradiated $Mg(VO_3)_2$:Tb(0.05mol%) phosphors for different mass of piston(0.1 kg to 0.4 kg) dropped on to the sample. It is observed that ML intensity increases with increasing mass of the piston dropped on to the sample without any considerable change in time corresponding to ML peaks.

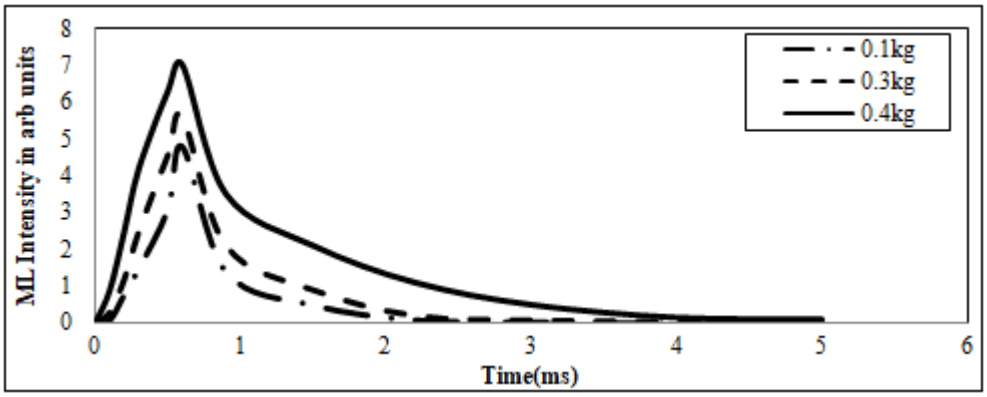

Figure 1: Time dependence of ML intensity of $Mg(VO_3)_2$:Tb(0.05mol%) phosphors for load of different masses (γ-dose 1.395 kGy, impact velocity 0.98 ms^{-1})

Figure 2 shows the time dependence of ML intensity of γ-irradiated $Mg(VO_3)_2$ phosphors for different concentration of Tb ions. It is observed that undoped $Mg(VO_3)_2$ shows very weak ML emission. ML intensity is found maximum for 0.05mol% concentration.

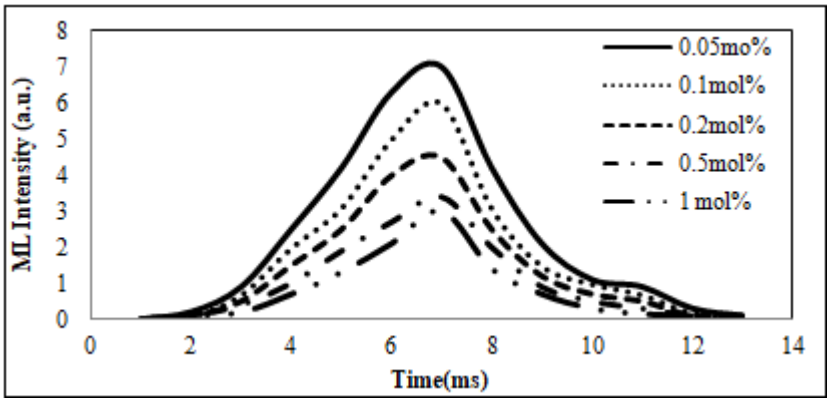

Figure 2: Time dependence of ML intensity of $Mg(VO_3)_2$:Tb phosphors for different concentration of dopant (γ-dose 1.395 kGy, impact velocity 0.98 ms^{-1}, load of piston 0.4 kg)

Figure 3 shows the time dependence of ML intensity of γ-irradiated $Mg(VO_3)_2$:Tb phosphors for different impact velocities from 0.7 ms^{-1} to 1 ms^{-1}. It is observed that ML intensity increases with increasing impact velocity and the time corresponding to ML peak shifts towards shorter time values with increasing impact velocity.

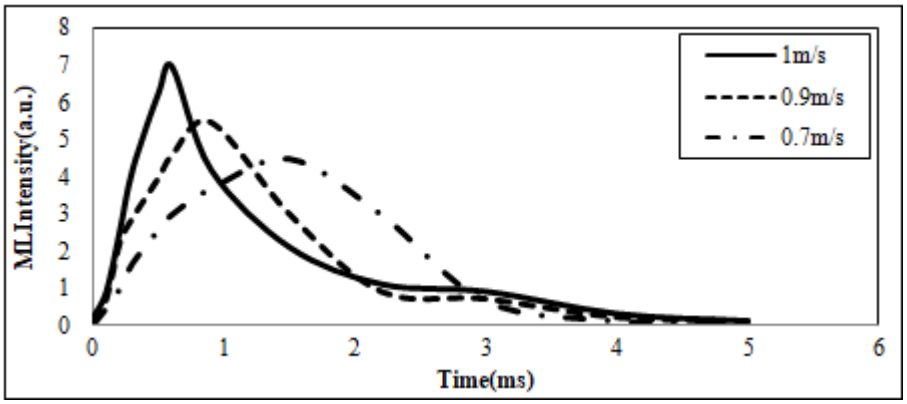

Figure 3: Time dependence of ML intensity of $Mg(VO_3)_2$:Tb(0.05mol%) phosphors for different impact velocities(γ-dose 1.395 kGy, mass of the piston 0.4 kg)

Figure 4 shows ML emission spectra of Tb doped γ-irradiated $Mg(VO_3)_2$ phosphors. Single peak is observed at 482nm, which show the characteristic emission of Tb^{3+} ions.

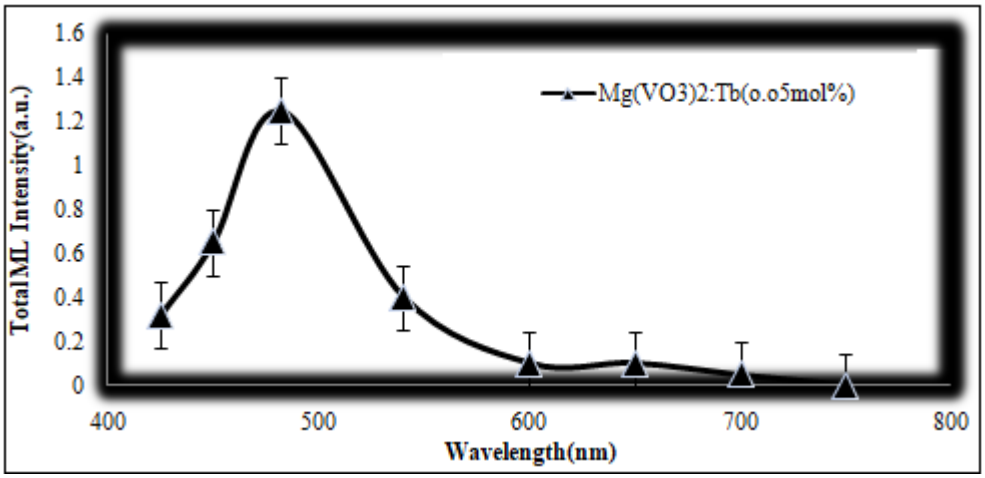

Figure 4: ML spectra of $Mg(VO_3)_2$:Tb(0.05mol%) doped vanadate based phosphors

Figure 5 shows the total ML intensity of $Mg(VO_3)_2$:Tb(0.05mol%) phosphors for different γ-doses given to the samples, in the dose range of 0.4 to 2.0 kGy. **Figure 6** shows total ML intensity of these phosphors increases with increasing γ-dose and seems to saturate for higher γ-doses.

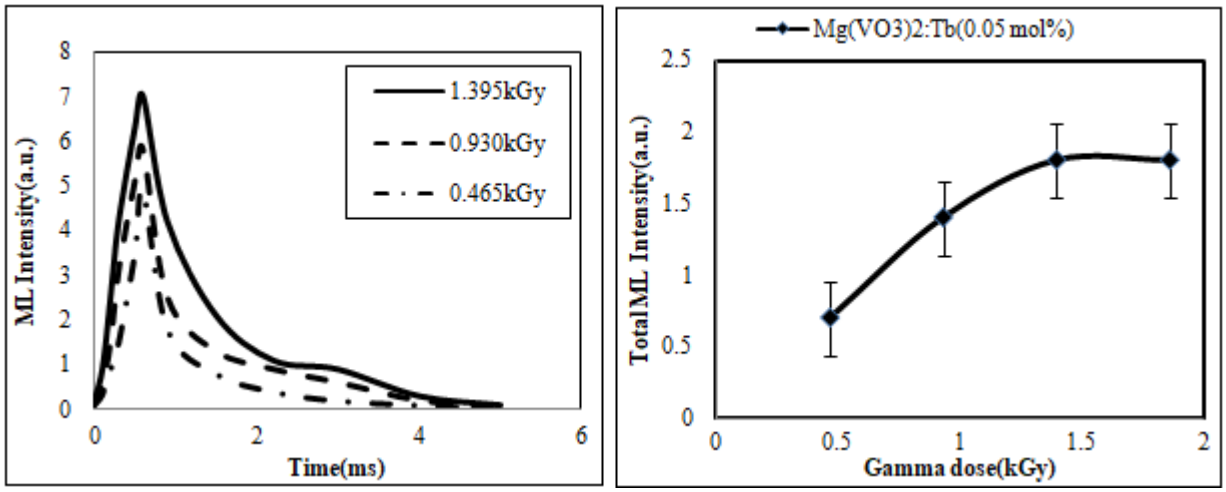

Figure 5 & 6: Time dependence of ML intensity of $Mg(VO_3)_2$:Tb(0.05mol%) phosphors at different gamma doses (Impact velocity 0.98 ms^{-1}, load of piston 0.4 kg)

Figure 7 shows PL emission spectra of $Mg(VO_3)_2$:Tb(0.05mol%) phosphors. A single peak is observed around 486 nm when phosphors are excited by 385 nm.

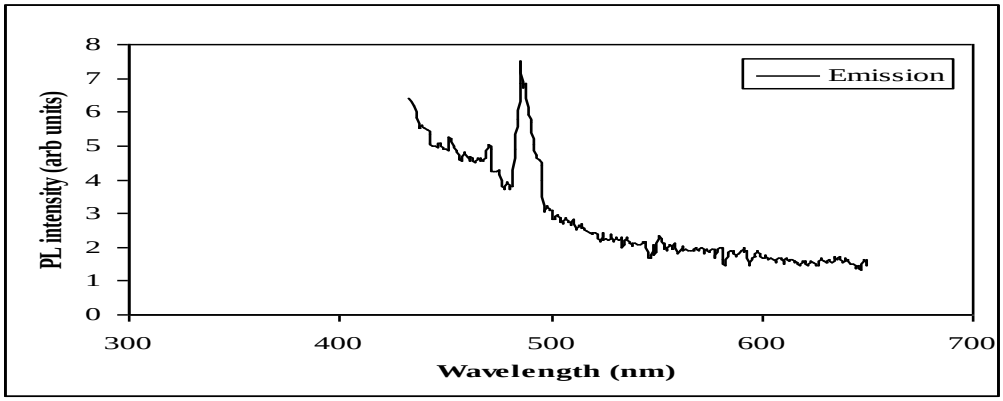

Figure 7: PL Emission spectra of $Mg(VO_3)_2$:Tb(0.05mol%) phosphors(λ_{ex}= 385 nm)

4. Discussion

In the study of mechanoluminescence, the main objective is to understand the origin of ML and to develop efficient ML materials. It is the matter of speculation that how the energy added to the system dissipate and which simultaneously passing processes of the physical nature having dominating influence on the emission of light. The recent experiments

which have been carried out to elucidate the excited state origins of the luminescence and to determine the mechanism, by which these states are populated, showed clearly that the emission of light during the mechanical deformation of solids is not attributed generally to friction. Since the mechanical energy could not populate directly the excited states of the molecules comprising the sample, some alternative process should be responsible for the ML excitation, Chandra [10]. Mechanoluminescence is one of the methods for studying radiation effects in solids. The presence of impurities enhances the generation/ recombination of luminescence centers, defects etc. The ML intensity depends upon the number of populated traps and the recombination of probability of trapping and recombination sites. When the sample is fractured by the impact of piston, it seems that very intense electric field of the order of 10^6-10^7 Vcm^{-1} is produced due to the charging of newly created surface near the crack tip (Fig.1). Due to this electric field, release of electron from electron traps may take place. The subsequent recombination of electrons moving in the conduction band with the hole trapping centers, and the hole moving the valance band with electron trapping centers may release energy. This energy may non-radiatively be transferred to $Tb^{2+} \leftrightarrow Tb^{3+}$ ions causing their excitation and subsequent de-excitation of excited Tb^{3+} ions may give rise to the characteristic luminescence of the rare earth ions.

It is observed that ML intensity depends on the concentration of rare earth impurity doped. Undoped irradiated samples show very weak ML intensity, it increases with increasing doping concentration attains maximum value for a particular concentration of dopant then decreases with further increase in centration of dopant. Initially the number of luminescence centers and defect centers increases with increasing concentration of dopant, which causes increase in ML intensity with increasing dopant concentration. For the higher values of dopant concentration, concentration quenching of luminescence centers may take place and therefore, the ML intensity decreases with further increasing in concentration of the dopant. As the mater of the fact, the ML intensity is optimum for a particular concentration of the dopant (Fig.2). At high strain rate or impact velocity, the values of β_r, β_n, and ($\lambda_r + \lambda_n$) are independent of the strain rate of rare earth doped vanadate based phosphors, however, the value of β depend on the strain rate. The dependence of β on the strain rate or impact velocity may be understood by in the following way. If $\tau = 1/\beta$ is the characteristic time related to the stopping of the piston impacted onto the rare earth doped vanadate based phosphors (Fig.3). ML emission spectrum of Tb doped γ-irradiated divalent metal metavanadate phosphors. Single peak at 482nm are observed for all the samples (Fig.4). It is observed that ML intensity increase with increasing gamma dose given to the samples and seems to saturate for higher values of gamma doses. When the vanadate based phosphors are exposed to ionizing radiation the defect centers like cation vacancies and vanadate radicals are created. On increasing the γ-dose, the defect centers N_c, increases and thereby the peak intensity I_m (for the samples having single peak in their ML glow curve) and total ML intensityincreases (Fig.5).

Menon et al [11] Tb^{3+} emission peaks is found around 486 nm which is assigned to $^5D_4 \rightarrow {}^7F_j$ (j = 6, 5, 4, 3, 2, 1) transitions. The reason is that this transition has largest probability for both the electric dipole and magnetic dipole induced transition. In present investigation peak around 486 nm has been observed in the Tb doped $Mg(VO_3)_2$phosphors which is clearly due to $^5D_4 \rightarrow {}^7F_6$ transitions of Tb^{3+} ions **(Fig.7)**.

5. Conclusion

ML intensity is found to be optimum for a particular concentration of Tb(0.05mol%) and similarity of ML spectra with PL spectra suggests that although the excitation processes are different, emission process is governed by the states of the similar nature. The relative intensities are different due to mode of excitation.

6. Acknowledgement

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