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Photoluminescence Studies Of Cerium Doped Magnesium Aluminate Nanophosphors ($MgAl_2O_4$: Ce)

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ABSTRACT : Magnesium aluminate doped with ;Ce^{3+} ($MgAl_2O_4$:Ce^{3+}) was prepared by combustion synthesis. The prepared samples were characterized by X-ray diffraction technique(XRD) sample particle size was found to be form in nano range. With variable concentration of Ce (0.05-0.8 mol%), no phase change was found with increase in concentration of Cerium(Ce). The FTIR Studies were also done which confirmed the composition of prepared phosphor. PL emission spectrum showed that Ce^{3+} acts as the luminescent centre. Effect of variable concentrations of Cerium on Photoluminescence (PL) was studied. In photoluminescence (PL) studies, the excitation wavelength was found around 330 nm and an intense broad emission peak appeared around 380 nm which may be due to the 5d-4f transition. Photoluminescence peak intensity versus concentration graph shows that intensity is maximum for 0.4 mol% of doping concentration, and then it decreases with further increase in doping percentage. Sample was prepared by Combustion technique which is less time taking and suitable for large scale production of nanophosphors.

Keywords:- photoluminescence, combustion ,magnesium aluminate.

1. *INTRODUCTION*

Magnesium aluminium oxide ($MgAl_2O_4$) is used in various technological applications such as light emitting devices including laser, optical, electrical applications and also in radiation environment [1]. Rare-earth-doped $MgAl_2O_4$ have been reported to be a long persistent afterglow emission phosphors used in light emitting device and color display [1,2]. Many of its properties viz. density, melting point are intermediate between those of its constituent oxides (MgO and Al_2O_3) [3]. The crystal structure of $MgAl_2O_4$ is a face-centered cubic lattice of oxygen ions, with a lattice parameter of 8.08 A˚. It has the cubic space group. Unit cell is formed with eight molecules, in which there are 64 tetrahedral symmetry sites and 32 octahedral sites. $MgAl_2O_4$ of natural origin have 8 magnesium ions occupying tetrahedral sites and 16 aluminium ions occupying octahedral sites. However, synthetic $MgAl_2O_4$ crystal can have up to 30% of cation antisite disorder [4–8]. Antisite formation due to interchange of the ions on tetrahedral and octahedral lattice positions by divalent and trivalent ions causes numerous trapping sites for the electron and hole on irradiation. Further, irradiation causes damage to the defect centres and impurities by changing their charge states [9]. These defect centres play a vital role in many of the luminescent and optical properties of the crystal. Optically stimulated luminescence (OSL) emission in pure $MgAl_2O_4$ crystal is reported by Yoshimura and Yukihara [10].

$MgAl_2O_4$:Dy^{3+} is a promising material for solid state displays, ceramic color pigments and solid state display applications. The photometric studies of magnesium aluminate revealed that the prepared samples were quite useful for the fabrication of white light emitting diodes [11].

Combustion synthesis or self-propagating high temperature synthesis (SHS) provides an attractive practical alternative to the conventional synthesis of producing advanced materials, such as ceramics, composites etc. The underlying basis of SHS is highly efficient energetic exothermic reaction with the evolution of various gases along with the high intense flame. It volatilizes low boiling point impurities and results in purer products than those produced by the other conventional synthesis [12]. This paper reports photoluminescence (PL) properties

of $MgAl_2O_4$ doped with Cerium, prepared by combustion synthesis. XRD and FTIR studies were carried out to identify the chemical composition , groups present , and Crystallinity of the phosphor.

2. EXPERIMENTAL

Magnesium nitrate, aluminium nitrate, urea and Cerium nitrate were used as starting materials. $MgAl_2O_4$ was prepared in stoichiometric ratio (oxidation to reducing valency). Metal nitrates, urea and desired amount of dopant are dissolved in water in a glass beaker. The beaker was kept on magnetic stirrer for ½ hr at 70^0C. The clear solution of beaker was transferred in silica crucible . the crucible was kept inside a pre heated furnace, which was set at 650 ^{0}C. Once the water boiled off, the metal nitrate and urea react and ignite the energy realeased from the reaction can produce temperatures in excess of 1500 ^{0}C [13,14]. The reaction is self-propagating and is able to sustain this high temperature long enough, from 1 to 5 s typically, to form the desired product. The entire combustion process was over in about 5-7 This result confirms the formation of the crystal phases of $MgAl_2O_4$, which includes the Al^{3+} ions in octahedral symmetry and Mg^{2+} occupying the occupy the site of bigger ions, i.e. Mg $^{2+}$ ions ($r_{Mg^{2+}}$ = 173 pm and $r_{Al^{3+}}$ = 67pm) in the $MgAl_2O_4$ lattice..

$$d_{hkl} = a/\sqrt{(h^2 + k^2 + l^2)}$$

where d_{hkl} is the distance between adjacent lattice planes.

h,k,l are the miller indices.

The photoluminescence and lifetime data were collected using a nanosecond optical parametric oscillator/amplifier (Spectra-Physics MOPO-730) operating at a 10 Hz repetition rate and tunable between 220 and 900 nm. The output of the MOPO system was frequency doubled in a KDP crystal and directed onto the particles. Emission was collected at right angles to the excitation and focused into a 1/8 metre monochromator equipped with either a gated intensified CCD detector (for emission spectra) or a standard photomultiplier tube (for lifetime measurement).

3.RESULTS

min. Voluminous polycrystalline $MgAl_2O_4$:Ce was prepared by this method. The identity, crystallinity, crystalline structure, size, and shape of the nanoparticles in the nanocomposites were determined by x-ray powder diffraction (XRD) . The XRD pattern was recorded with a diffractometer using monochromatized Cu $K_{\alpha 1}$ (λ = 1.540 56 A) radiation with Si (a = 0.5430 88 nm) as an internal standard. The crystallite size (D_{xrd}) of the calcined powder were determined by using Scherrer's equation as follows:

$$D_{xrd} = \frac{0.9\,\lambda}{\beta\cos\theta}$$

Where λ is the wavelength of the X-ray (1.5406 Å), θ is the scattering angle of the main peaks used for calculation β is the corrected peak at full width at half-maximum (FWHM) intensity. For the simple cubic crystal, the lattice constant (a) of synthesized magnesium aluminate nanoparticle was calculated from the equation below: (111),(210),(311),(400),(422),(511) and (440). tetrahedral symmetry site. The trivalent Ce^{3+} with ionic radius of 180 pm is expected to preferentially

.. Particle size calculated by Scherrer's equation is in nano meter range .

3.1 *XRD*

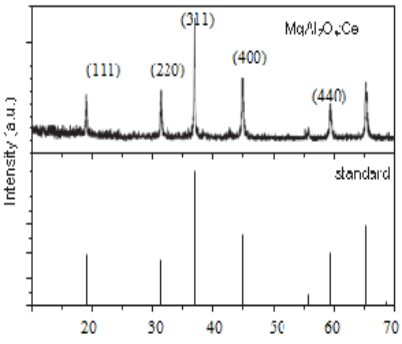

Figure 1. XRD of Ce Doped MgAl2O4

XRD pattern of prepared $MgAl_2O_4$:Ce and standard XRD pattern (JCPDS file 75-0905) shown in fig1. XRD of MgAl2O4 shows various planes of diffraction with miller indices

3.2 *FTIR*

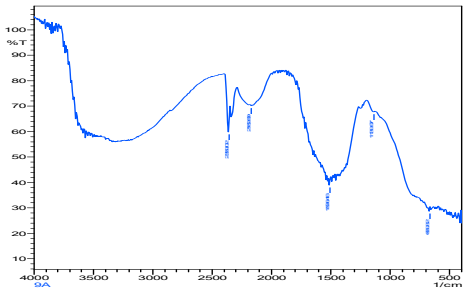

Figure 2

Fig. 2 illustrates the FTIR spectrum of the Cerium doped magnesium aluminate complexes. Several characteristic bands of TEA ligand were observed. The broad peak centered at 3500 cm^{-1} was assigned to the O-H stretching which could be the moisture absorption or triethanolamine residue from the reaction. The peaks between 2169 and 2359 cm^{-1} were corresponding to asymmetric and symmetric stretching modes of methylene (-CH2-) group, respectively. The N-O stretching of nitrate group showed moderate absorption bands in region of 1550-1250 cm^{-1} [26]. The Mg-O-C bending resonated at 1139 cm^{-1} . Moreover, the small absorption peaks at 669 and 499 cm^{-1} could be assigned to M- O or M- N stretching of metal (M = Mg, Al and Ce) coordinated with TEA ligand.

3.3 PHOTOLUMINESCENCE

PL emission spectrum shows that Ce^{3+} acts as the luminescent centre. Effect of variable concentrations of Cerium on Photoluminescence (PL) was studied. In photoluminescence (PL) studies, the excitation wavelength shown in fig 3(a) it was found around 330 nm and an intense emission peak appears around 380 nm and 411nm which may be due to the 5d-4f transition . Photoluminescence peak intensity versus concentration graph shows in fig 3(b) that intensity is maximum for 0.4 mol% of doping concentration, and then it decreases with further increase in doping percentage .

4. *CONCLUSION*

As Cerium (Ce^{3+}) **i**on (180pm) is expected to occupy the site of bigger ion magnesium (Mg) preferably, we may conclude that more number of Ce ion may occupy the site. Consequently we say that the more luminescence center may be produced, which will produce the stability in PL. it is present requirement . More stable nanophosphors for solid state lighting purpose are expected to be produced in near future.

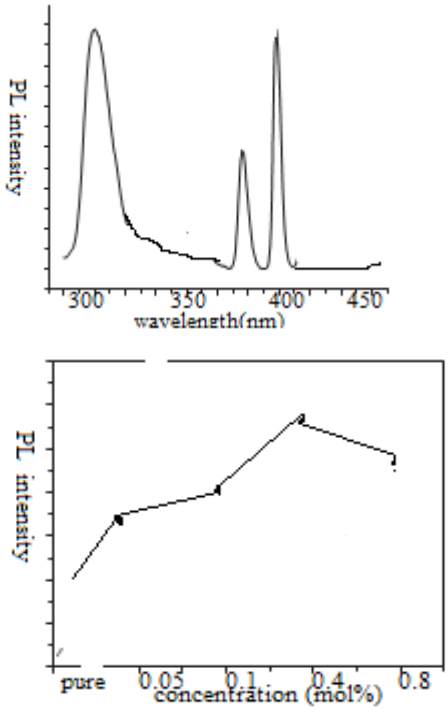

Figure 3(a) ,3(b)

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