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### RESEARCH ARTICLE

## STRUCTURAL AND PHOTOLUMINESCENCE CHARACTERIZATION OF MgAl<sub>2</sub>O<sub>4</sub>:Eu PHOSPHOR SYNTHESIZED BY COMBUSTION METHOD.

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### Abstract

The MgAl<sub>2</sub>O<sub>4</sub> spinel doped with Eu<sup>2+</sup> ions powder phosphor was prepared at temperature as low as 500 °C using the combustion route. A structural property of the powder was characterized by X-ray diffraction (XRD). This XRD pattern shows the well crystallized cubic phase, Fd-3m space group of MgAl<sub>2</sub>O<sub>4</sub>. The estimated average crystalline size is about 36 and 30 nm for MgAl<sub>2</sub>O<sub>4</sub> and Eu doped MgAl<sub>2</sub>O<sub>4</sub> particles respectively. The photoluminescent property of prepared powder was investigated using excitation and emission spectroscopy at room temperatures. Energy levels scheme is proposed for emission from MgAl<sub>2</sub>O<sub>4</sub>:Eu<sup>2+</sup>.

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### Introduction:-

In recent years, rare earth ion doped inorganic nanomaterials have received significant attention in the field of science and engineering [1, 2]. Inorganic phosphors doped with rare earth ions have been used in lighting systems and for multicolor displays to overcome the drawback such as poor color rendition, high toxicity and complicated synthesizing process [3,4]. Among them, magnesium-aluminum oxide (MgAl<sub>2</sub>O<sub>4</sub>) spinel crystals have received a great deal of attention due to its mechanical strength, high resistance to chemical attacks, and its outstanding dielectric and optical properties [5]. Europium (Eu) doped in host can improve the photoluminescence properties of MgAl<sub>2</sub>O<sub>4</sub>:Eu<sup>3+</sup> phosphors [6]. MgAl<sub>2</sub>O<sub>4</sub>:Eu<sup>3+</sup> phosphors have been prepared by various methods such as solid-state reaction, sol-gel, spray-drying, co-precipitation, microwave assisted hydrothermal method and combustion synthesis. Among the methods combustion is a simple method which requires low temperature and less time as compare to other one. In the combustion process, the phosphors can be synthesized within 5 minutes at low temperatures. In the present investigation we have reported the synthesis of Eu<sup>2+</sup> activated MgAl<sub>2</sub>O<sub>4</sub> (MgAl<sub>2</sub>O<sub>4</sub>:Eu<sup>2+</sup>) and it was characterized by using techniques such as powder X-ray diffraction (XRD), and photoluminescence (PL).

### Experimental:-

In the present study analytical grade reagents were used without any further purification. MgAl<sub>2</sub>O<sub>4</sub>:Eu<sup>2+</sup> was prepared by combustion technique [7] using precursors Magnesium Nitrate (Mg(NO<sub>3</sub>)<sub>2</sub>), Aluminium Nitrate, (Al(NO<sub>3</sub>)<sub>3</sub>.9H<sub>2</sub>O) and Urea (NH<sub>2</sub>.CO.NH<sub>2</sub>). Metal nitrates are used as oxidizers, and urea is employed as fuel. Starting materials (Al(NO<sub>3</sub>)<sub>3</sub>.9H<sub>2</sub>O) and NH<sub>2</sub>.CO.NH<sub>2</sub> were taken in the stoichiometric ratios to crushed and ground in an agate mortar to form gel-like paste with the help of distilled water. The china dish containing the mixture was inserted in a vertical furnace preheated at 500 °C. Initially, the paste melts and undergoes dehydration followed by decomposition with the evolution of large amounts of gases. The mixture then foamed and finally

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ignited, yielding a white fluffy powder, which was collected after removing the dish from the furnace. The structural characterization of the product was performed by an X-ray diffractometer (Rigaku rotating anode H-3R). Photoluminescence spectra at room temperature were recorded in the range 200-750 nm on Spectrofluorophotometer (Shimadzu RF-5301 pc).

## Results and Discussions:-

### Structural Characterizations:-

The phase composition of the synthesized powder was analyzed by XRD using Cu-K $\alpha$  radiation (1.54184 Å) in the 2 $\theta$  range of 10 $^{\circ}$  to 80 $^{\circ}$ . Fig. 1 shows the XRD pattern of the as-prepared MgAl $_2$ O $_4$  sample and Eu doped MgAl $_2$ O $_4$  sample. The observed diffraction peaks correspond to those of the standard patterns for cubic MgAl $_2$ O $_4$  (JCPDS, No.77-0435). The XRD data of sample is very similar to standard pattern and there are no additional peaks observed for the doped MgAl $_2$ O $_4$  materials. This pattern indicates that the well crystallized cubic phase, Fd-3m space group of MgAl $_2$ O $_4$  was obtained by the urea-assisted combustion method even at furnace temperature 500  $^{\circ}$ C. The XRD pattern of this phosphor contains three phases: the main spinel type MgAl $_2$ O $_4$  phase (Fd3m space group) and some additives of MgO (Fm3m space group) and  $\alpha$ -Al $_2$ O $_3$  (space group R3c). Eight diffraction peaks can be indexed as the (111), (2 2 0), (3 1 1), (4 0 0), (4 2 2), (5 1 1), (4 4 0) and (5 3 3) diffraction lines of high intensities. A stronger peak (3 1 1) at 2 $\theta$  of around 36 $^{\circ}$  is used to calculate the average crystalline size ( $D$ ) of the MgAl $_2$ O $_4$  and Eu doped MgAl $_2$ O $_4$  particles.

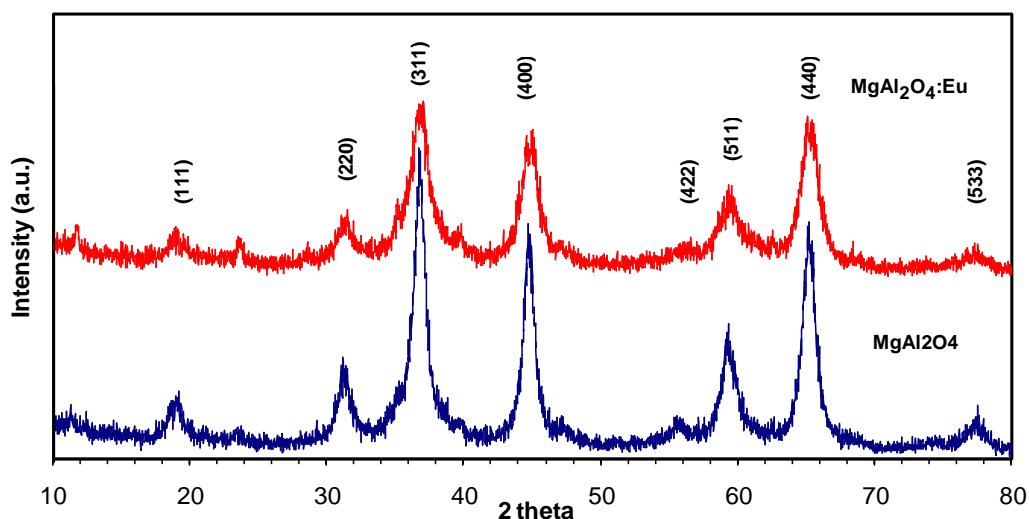


Fig. 1:- X-ray diffraction of MgAl $_2$ O $_4$  and MgAl $_2$ O $_4$ :Eu $^{2+}$  phosphor powder.

The average crystallite size was estimated from the full width at half maximum (FWHM) of the diffraction peaks by using the Scherrer formula [8]

$$D = \frac{0.9\lambda}{\beta \cos\theta}$$

The estimated average crystalline size is about 36 and 30 nm. It has been reported that the particle size of MgAl $_2$ O $_4$  spinel synthesized by various methods have been in the range of 43–55 nm [9].

## Photoluminescence studies:-

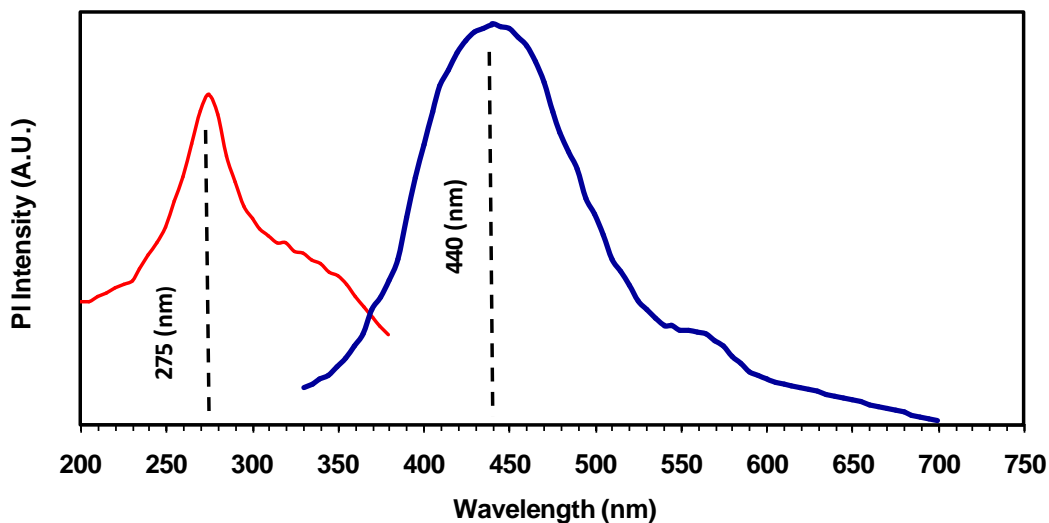


Fig. 2:- shows the photoluminescence spectrum.

Fig. 2 shows the photoluminescence spectrum (excitation and emission spectra) of Eu ions incorporated in  $\text{MgAl}_2\text{O}_4$  ranging from 200 to 750 nm. Excitation spectra of  $\text{MgAl}_2\text{O}_4:\text{Eu}$  materials prepared at  $500^\circ\text{C}$  were recorded under emission monitored at 440 nm. The excitation spectra display intense peaks at 275 nm would be due to the defects within the host material [5].

From the spectra it can also be noted that the broad emission peak is located at 440 nm, which could be attributed to the emission of  $\text{Eu}^{2+}$  ion. The short-wavelength part of the spectrum, bluish green broad band emission at 430–570 nm, is attributed to the  $4f^65d^1 \rightarrow 4f^7(^8S_{7/2})$  transitions of  $\text{Eu}^{2+}$  [10,11]. Europium is the most versatile among all lanthanides. It occurs more often in the divalent than trivalent state, which is in the contrast to all the other lanthanides. The high stability of the divalent state originates from the stability of the half filled shell configuration ( $4f^7$ ) for  $\text{Eu}^{2+}$ . The valence state of europium has a great influence on its luminescence properties. The 5d-4f transitions of  $\text{Eu}^{2+}$  emissions are parity allowed. The luminescence of divalent europium consists mostly of broad intense band emission in the visible spectral range. The ground state of the  $4f^7$  configuration of  $\text{Eu}^{2+}$  is  $8S_{7/2}$ , while the lowest  $4f^7$  excited state is the  $^6P_J$  state in the UV region. In most  $\text{Eu}^{2+}$  activated phosphors, the lowest level of the  $4f^65d$  configuration lies below the  $^6P_J$  state, leading to broadband luminescence due to the inter-configurational electronic  $4f^65d - 4f^7(^8S_{7/2})$  transition [12].

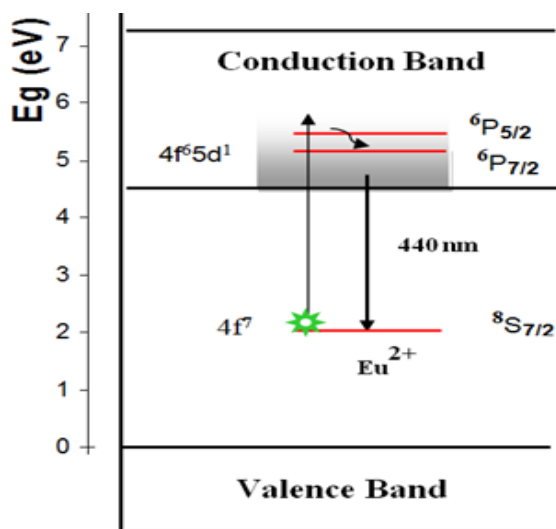


Fig.3:- Proposed energy levels scheme for emission from  $\text{MgAl}_2\text{O}_4:\text{Eu}^{2+}$

On excitation with UV, two types of lowest excited states are possible, namely  ${}^6P_j$  (f-f) or  $4f^65d$  (f-f), depending on the host matrix [13]. In case of  $\text{Eu}^{2+}$  ion in  $\text{MgAl}_2\text{O}_4$  the possible lowest excited states is  $4f^65d$  (f-f) [5]. The optical band gap energy value of magnesium aluminate is estimated as 4.5 eV [14]. Based on above discussion the possible emission mechanisms from the  $\text{Eu}^{2+}$  activated  $\text{MgAl}_2\text{O}_4$  is illustrated in Fig 3.

### Conclusions:-

The nanocrystals of the  $\text{MgAl}_2\text{O}_4:\text{Eu}^{2+}$  spinel was prepared via combustion method. Present investigation revealed that urea-nitrate combustion synthesis has outstanding potential for producing Eu-doped  $\text{MgAl}_2\text{O}_4$  phosphors in a single step. Nanopowder demonstrated a strong and quite short lived luminescence, characteristic for the doped  $\text{Eu}^{2+}$  ions. The broad emission peak at 440 nm is attributed to the usual near UV emission of  $\text{Eu}^{2+}$  ion from the  $4f^65d^1 \rightarrow 4f_7(8S_{7/2})$  transition.

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