

SYNTHESIS AND PRELIMINARY STUDIES OF LIMGPO₄:EU³⁺ PHOSPHOR FOR SOLID STATE LIGHTING APPLICATION

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Abstract : Lithium Magnesium Phosphate doped with Europium (LiMgPO₄:Eu³⁺) phosphor was successfully synthesized by Stearic acid Sol-Gel Method. The structural properties of prepared phosphor were studied using X-Ray Diffraction technique. XRD peaks were matched with the standard International Center for Diffraction Data (ICDD) file and the synthesis method did not affect the XRD patterns. 0.005 mole of Eu were doped in the LiMgPO₄ host, which actually replaced Mg-ions in the host lattice. The PL excitation spectra of the phosphor showed the excitation peak at 394 nm while the PL emission spectra were monitored at 614 nm. The emission of Eu³⁺ is observed due to the 5D₀ → 7F₂ transition giving the red-orange luminescence.

Index Terms - Sol-Gel method, ICDD, luminescence, effective atomic number.

I. INTRODUCTION

After Edison's futuristic statement over 100 years ago –“We will make electricity so cheap that only the rich will burn candles”[1] – the wishful dream of cheap, abundant electricity has not come true yet not only because of the continuous increase in population but also due to the increasing demand of electricity. Only about 24% of the people living in sub-Saharan Africa had access to electricity in 2000 [2]. Also in most of the developing countries the electrical networks are limited to urban areas only. Rest of the people, who do not have access to the electric supply, use biomass and petroleum fuels for lighting. Fuel based lighting is not only inefficient and expensive compared to electric lighting, but is also a severe cause of respiratory and cardiac health problems [2], [3], [4]. Light Emitting Diodes (LEDs) are rapidly evolving light sources. Cost analysis of LED based lighting systems driven with renewable energy sources in different parts of developing countries have shown them to be cost effective in comparison with the existing options [4], [5], [6]. Currently W-LEDs are preferred owing to low energy consumption and reduction of greenhouse gas emissions due to their high efficiency, long lifetime and environmentally friendly characteristics [7], [8], [9]. LiMgPO₄ belongs to a large family of phosphates with formula ABPO₄ where A and B are mono and divalent cations, respectively, with structures dependent on the relative size of the ions [10]. If the size of A and B are small, such as Li¹⁺ and Mg²⁺, the resulting compound adopts the olivine structure [11]. Also recently it has been reported that LiMgPO₄ can be a future material for OSL dosimetry because its chemical and physical properties show some advantages over commercial available materials [12]. Hence, the research shows that LiMgPO₄ is a phosphor having multi applications [13]. In recent years, rare-earth ions activated inorganic material become important due to their high chemical stability and high luminescence efficiency [14]. Rare earth luminescent materials have considerable practical applications in almost any device involving the artificial production of light, such as cathode ray tubes, lamps, and X-ray detectors, etc. [15-21]. Among the rare earth elements, europium is an exceptional dopant because it can exist in divalent and trivalent states. Both these ions show many different characteristics as emitting centers. In the case of Eu³⁺, photoluminescence typically originates from the intra-configurational 4f → 4f transitions which are almost independent of ligand field strength and even the effect of the local symmetry. Mostly, the room temperature emission of Eu³⁺ originates from the 5D⁰ state and locates about 595 nm (5D⁰ → 7F¹) and 610–620 nm (5D⁰ → 7F²). As a result, Eu³⁺ ion provides an orange-red luminescence [13]. Also Eu³⁺ ions is an excellent activator have been widely used in red-emitting phosphors [22]. Eu³⁺ doped inorganic materials such as borates, aluminates, phosphates, tungstets, vanadates and silicates, give strong red emission [23].

II. EXPERIMENTAL

As per the reports LiMgPO₄ was mostly made by Solid State Diffusion method. In the present work we prepared the LiMgPO₄:Eu³⁺ by the Stearic acid Sol-Gel Method. Firstly the 0.5M stearic acid was taken in

the china dish and it was placed on the hot plate at 100°C for 10 min till the acid get melted. Then all the dried precursors along with the dopant, taken in the stoichiometric proportion, were added in the molten stearic acid. The precursors used were Lithium Nitrate [LiNO₃], Magnesium Nitrate Hexahydrate [Mg(NO₃)₂·6H₂O], Ammonia Dihydrogen Orthophosphate [NH₄H₂PO₄] and Eu₂O₃ was added as dopant. The molten stearic acid along with the dried precursors was then added and 0.005 mole of Eu₂O₃ with 2-3 drops of acetic acid. The mixture was then kept on the hot plate at 200°C for around 2 hours. The solution was then allowed to cool. After cooling, the transparent gel was formed. The dry gel was then slowly heated and burnt slowly into red flame. The residue was then sintered at 950°C for 2 hours. Finally we got the white powder which was nothing but the required LiMg(1-0.005)PO₄:0.005Eu³⁺.

Table1. Molar ratio of ingredients used for material preparation and corresponding chemical reaction.

Product	Products Corresponding reaction with balanced molar ratios of precursors
LiMg _(1-0.005) PO ₄ :0.005Eu ³⁺	LiMg _(1-0.005) PO ₄ : 0.005Eu ³⁺ = LiNO ₃ + Mg _(1-0.005) (NO ₃) ₂ ·6H ₂ O + NH ₄ H ₂ PO ₄ + 0.005Eu ₂ O ₃ {in stock solution form 1 g = 100 ml}= LiMg _(1-0.005) PO ₄ :0.005Eu ³⁺ + gaseous products (H ₂ O, NH ₃ and NO ₂)

III. RESULTS AND DISCUSSIONS

XRD (X-RAY DIFFRACTION) PATTERN

The crystalline structure of the sample was studied using powder X-ray diffraction (XRD) technique. The XRD pattern of LiMgPO₄:Eu³⁺ is shown in figure 1. The experimental pattern of was compared with the standard ICDD (International Center for Diffraction Data) pattern with card no. 01-084-0342. From the comparison it is clear that the position of the main peaks is same. Hence, there were no impurity lines which confirm the crystalline product. LiMgPO₄ was cubic crystal structure with lattice constants as a=b=c=4.452 Å. The addition of dopant Eu³⁺ did not affect the XRD pattern of the phosphor which suggests that the dopant was well incorporated in the host lattice.

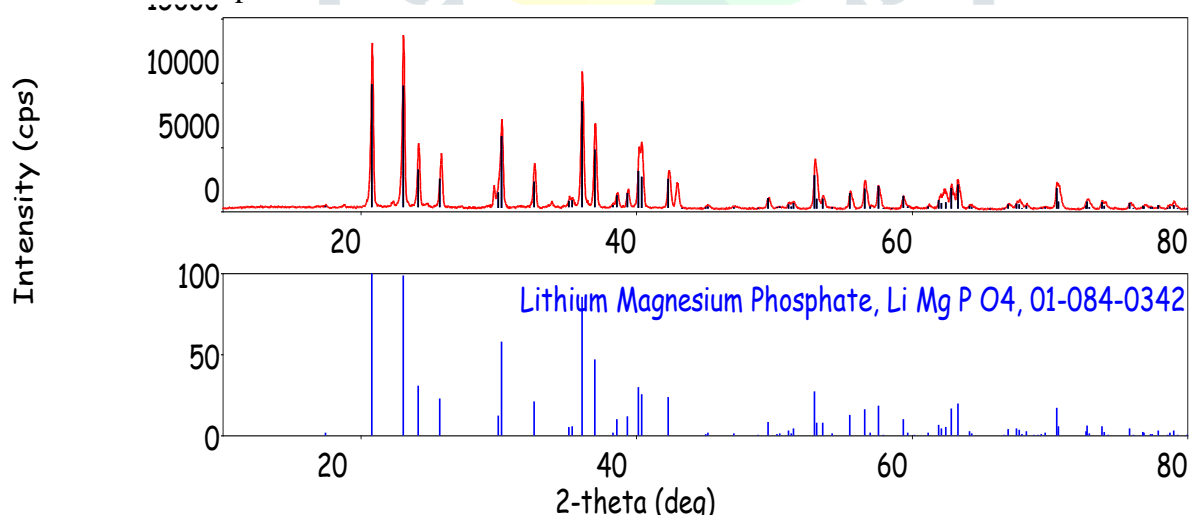


Figure1. X-ray diffraction patterns of LiMgPO₄:Eu³⁺ phosphor matched with the standard data ICDD file.

PHOTOLUMINESCENCE (PL) PROPERTIES

The photoluminescence properties of LiMgPO₄:Eu³⁺ were studied and the excitation and emission spectra of LiMgPO₄:Eu³⁺ phosphors are shown in Figure 2. The excitation and emission peaks were monitored at 394 nm and 614 nm respectively. The excitation spectra consist of a number of peaks in the wavelength range from 350 nm to 420 nm with the main peak at 394 nm. The emission spectra possess two characteristic peaks at 593 nm and 614 nm. Main emission at 614 nm is attributed to 5D⁰ to 7F² transition of Eu³⁺ ions which results into red-orange luminescence.

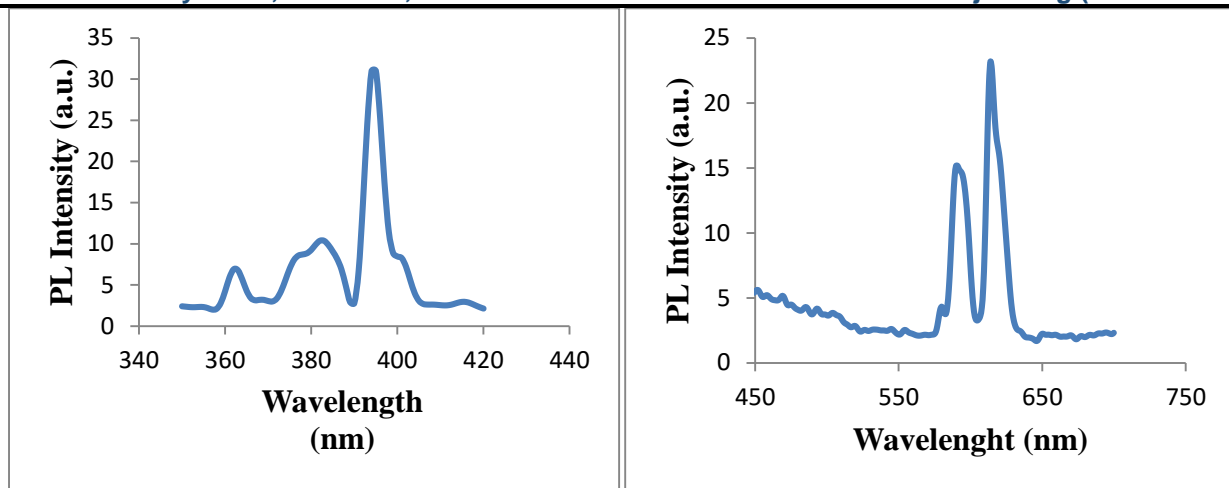


Figure 2. Excitation and Emission Spectra of LiMgPO₄:Eu³⁺ phosphor synthesized by Sol-Gel Method.

IV. CONCLUSIONS

The XRD pattern of LiMgPO₄: Eu³⁺ matched well with the ICDD data which implies that the LiMgPO₄: Eu³⁺ was successfully prepared by using Stearic acid Sol-Gel Method, by keeping concentration of Eu as 0.005 mole. Thus this method can be utilized for the preparation of LiMgPO₄: Eu³⁺ as this method is simple, low cost, uses readily available raw materials. The photoluminescence study of the prepared phosphor shows the excitation spectra in the range 350 to 420 nm with the highest peak at 394 nm. The prepared sample exhibits the PL emission peak at 614 nm which indicates 5D⁰ to 7F² transition of Eu³⁺ ions. As a result, Eu³⁺ ion provides an orange-red luminescence. Hence, the synthesized LiMgPO₄: Eu³⁺ can be the promising phosphor for the Solid State Lighting application.

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