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Microwave-assisted synthesis of benazoxoazol derivatives and their applications for phosphors of white light-emitting diodes

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ABSTRACT

In this paper, several benazoxoazol derivatives have been rapidly and effectively synthesized by microwave irradiation and their physical properties (i.e. specific gravities, thermal resistance, and florescent performances) have also been investigated. Since lab-made organic phosphors (OP-1 and OP-2) exhibit high fluorescent efficiencies, low specific gravities, and excellent thermal resistance, we have applied them for the encapsulation of white light-emitting diodes (LEDs) as phosphors. Experimental results reveal that OP-1 and OP-2 possess appropriate emitting wavelengths and their electroluminescent properties highly depend upon their chemical structures, thus causing the diverse hues of white LEDs with them. The hues of white LEDs with OP-1 and OP-2 are $CIE_{x,y}$=(0.35, 0.34) and $CIE_{x,y}$=(0.25, 0.24), respectively.

1. Introduction

In 1907, H.J. Round discovered the unprecedented electroluminescent properties of semiconductors [1]. Until 1962, first red light emitting diodes (LEDs) with gallium arsenic phosphide (GaAsP) were successfully developed by General Electric (GE) Co. in United States and commercialized four years later [2]. In 1996, white LEDs were manufactured with blue chips (indium gallium nitride; InGaN) and yellow yttrium aluminum garnet (YAG; $Y_3Al_5O_{12}$:Ce) phosphors by Nichia Co. in Japan [3] and have attracted much attention owing their miscellaneous advantages such as long lifetime, small size, low driving voltage, fast response time, low energy consumption, non-mercury contamination, and so on [4]. Recently, they have been applied for displays and illumination (e.g. mobile phones, digital cameras, laptops, indoor lightings, lamps, traffic lights, etc.). However, rare-earth metals and severe processing conditions (800–2000 °C; 1–10 atm) are necessary for the preparation of inorganic phosphors (yttrium aluminum garnet (YAG; $Y_3Al_5O_{12}$:Ce) or terbium aluminum garnet (TAG; $Tb_3Al_5O_{12}$:Ce), causing huge energy consumption and high cost. Moreover, sedimentation happens while inorganic phosphors and encapsulating materials are mixed because of their high specific gravities, resulting in short pot life. Poor compatibilities also exist between inorganic phosphors and organic encapsulating materials, leading to bad dispersion and lowering the optical properties (e.g. halation, light-scattering, etc.). Therefore, simple and low cost preparation for phosphors of white LEDs has been an important issue. Although there are also a lot of studies reported in literature [5–7] in which organic materials and molecules are applied instead of inorganic ones for white light generation in combination with inorganic LEDs, their preparing procedures are complicated and long reaction durations are needed.

In this study, benazoxoazol derivatives have been fast and effectively synthesized under microwave illumination and applied for the phosphors of LEDs. Experimental results reveal that microwave synthesizing procedure facilitates the reaction and LEDs with lab-made organic phosphors, which exhibit high thermal stability, low specific gravities, and high fluorescent quantum yields, irradiate white light.

2. Experimental

2.1. Materials and instruments

All of starting materials in this paper were purchased from Aldrich Co. and utilized without further purification. The specific gravity, thermogravimetric analysis (TGA), UV/vis absorption spectra, and photoluminescence (PL) spectra of organic phosphors were measured with a MH-300E, a TA TGA Q500, a HITACHI U-3300, and a HITACHI F-2500, respectively. Moreover, we recorded the electroluminescent spectra, CIE chromaticity diagrams, color rendering indices (CRI), and color temperatures of LEDs by Keithley 2400, Spectrascan PR650, and integrating sphere. The microwave oven for synthesis was CEM Mars.

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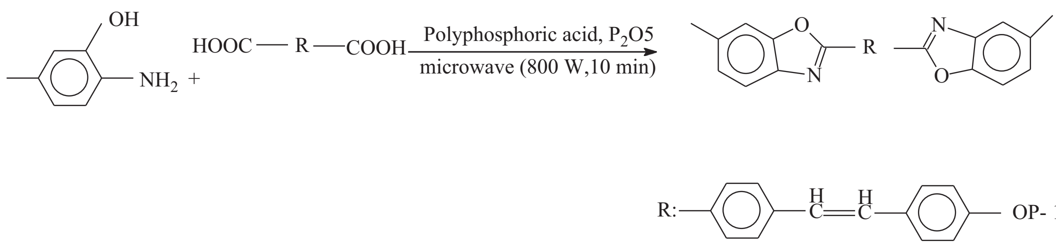
2.2. Quantum yield of fluorescence

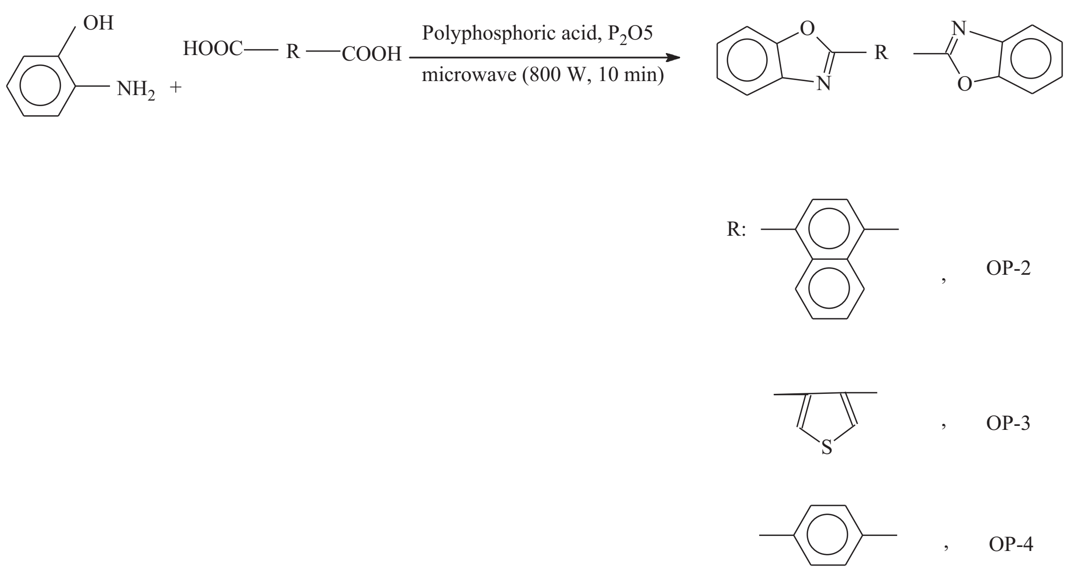
The quantum yield was calculated from the following equation [8]

$$\Phi_{sample} = \Phi_{standard} \frac{[\mathrm{Area}_{sample}/1-\exp(-A_{sample})]}{[\mathrm{Area}_{standard}/1-\exp(-A_{standard})]} \quad (1)$$

where Φ_{sample} and $\Phi_{standard}$ represent the quantum yields of the sample and standard, respectively. Area $_{sample}$ and Area $_{standard}$ represent the area of fluorescent emission band for sample and standard, respectively. A_{sample} and $A_{standard}$ represent the UV/vis absorbances of sample and standard, respectively. The standard in this paper was Rose Bengal ($\Phi_{standard}=\Phi_{Rose\ Bengal}=0.14$). The concentrations of samples were 10^{-6} M.



Scheme 1. Synthesis of OP-1.



Scheme 2. Synthesis of OP-2, OP-3 and OP-4.

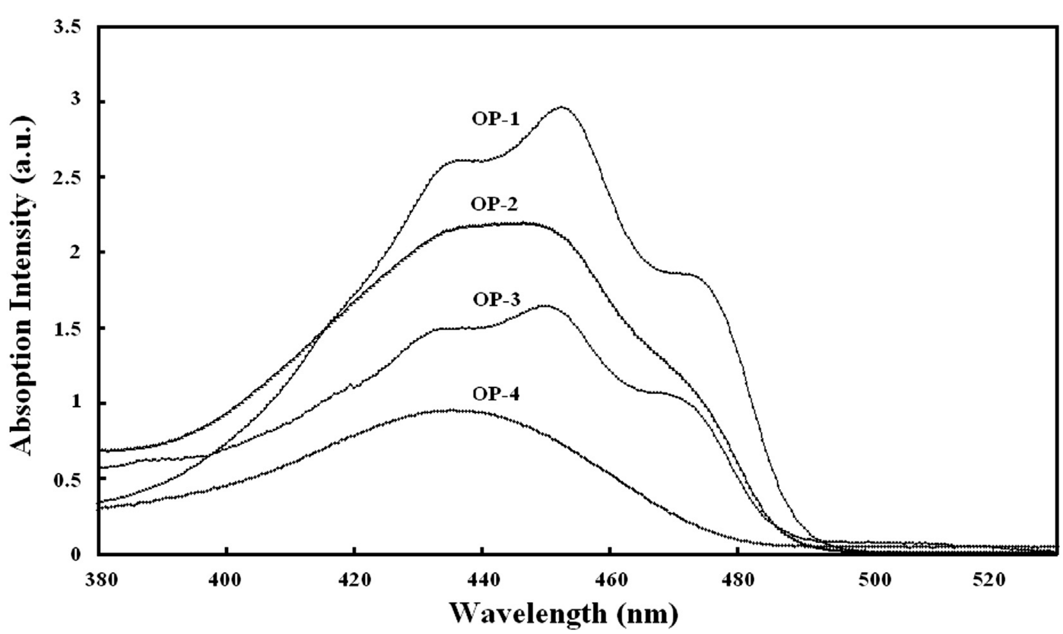


Fig. 1. UV/vis spectra of lab-made phosphors in toluene.