Synthesis and Electroluminescent property of bis (2-(benzimidazol-2-yl) quinolinato) Magnesium

Pin-Wen Cheng, Chien-Chih Lin, Anchi Yeh

Department of Chemical and Materials Engineering, Chengshiu University, Kaohsiung, 83347, Taiwan, R.O.C
E-mail Anchi Yeh: acyeh@csu.edu.tw

Abstract: An emission material, (2-(benzimidazol-2-yl)quinolinato)magnesium (MgBIQ) used for organic light emitting devices, has been synthesized. The melting transition (T_m) of MgBIQ is 436℃ and no glass transition temperature (T_g) was observed up to 430℃. The emission spectrum of organic emitting device using MgBIQ as emitted layer exhibits a broad maximum at 596 nm. The color of the emitted light is in the red region in the CIE coordinate of x = 0.46 y = 0.46.

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1. Introduction

Since an organic light emitting diode was reported by Tang and Vanslyke [1], LEDs based on organic or polymeric materials have generated considerable interest and enabled the development of low-cost, full-color, flat-panel displays along with other emissive products [2-5]. Organic and polymer devices provide advantages over their inorganic counterparts, such as high luminous efficiency and fine-pixel formation. The best-known EL metal chelate compound is Alq_3, not only a good emitter but also a highly efficient electron-transporting material, where q is the 8-hydroxyquinolinato ligand [6, 7]. Via the modification of the ligand of metal chelate compound, the emission color of a metal chelate compound may be tuned. Other properties, such as thermal stability and carrier mobility, may also be improved upon. In the present work, we report the synthesis and electroluminescent (EL) property of (2-(benzimidazol-2-yl)quinolinato)magnesium (MgBIQ). The attachment of the benzimidazol group at 2-position would allow the ligand to form stable complexes with metal ions similarly to 8-hydroxyquinoline. Therefore, the thermal stability, an important character for the practical application in the electronic fields, of this metal complex is investigated by thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC). The organic emitting device using MgBIQ as emitting layer has been fabricated to study the electroluminescent property of this metal complex.

2. Experimental Setup

The synthesis of the title compound was accomplished by following processes, as shown in Scheme 1. The dibutylmagnesium solution (0.5M in heptane, 0.6927mL, 10mmol) was slowly added to 100 ml of THF solution containing benzimidazol-2-yl-quinoline (2.45g, 10mmol) at –10℃ under N_2. After the resulting mixture was stirred at room temperature for 4 hours, 5 ml isopropyl alcohol was added to quench the reaction. The solvents were removed under vacuum condition, and the residual solid was sublimed to purify the final product. reddish orange powder of MgBIQ was obtained in 75% yield. The formula of this compound has been determined by 1H NMR and elemental analysis. The organic light emitting device, Fig. 1, using MgBIQ as the emitting layer were fabricated on the transparent conductive indium-tin oxide (ITO) glass substrate. The organic layers and the cathode were sequentially deposited by conventional vacuum vapor deposition in the same chamber without breaking the vacuum under 3×10^{-7} Torr. In the present work, the N,N'-bis-(1-naphthyl)- N,N'-diphenyl-1,1'-biphenyl-4,4'-diamine (NPB, 1c) was used as the hole-transport material (HTM), and tris (8-quinnolinolato) aluminum (Alq_3, 1d) was employed as the electron-transporting material (ETM). The EL spectrum and the Commission Internationale de l'Eclairage (CIE) co-ordinates were measured by Pro-650 Spectroscanner, the current-voltage (I-V) characteristic was measured by Keithley 2400 Source meter.

Thermogravimetric analysis (TGA) was performed on a Perkin-Elmer thermogravimeter (Pyris 1) under a dry nitrogen gas flow at the heating rate of 20℃/min. Glass transition temperature (T_g) and melting point (T_m) of materials were determined by differential scanning calorimetry of the Perkin-Elmer differential scanning calorimeter (DSC-7).

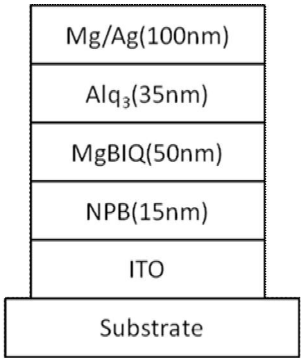
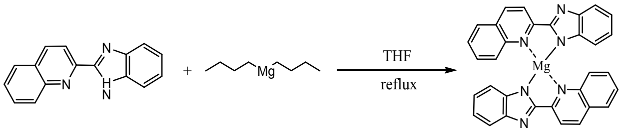


Fig. 1. Device structure of organic light emitting device (OLED) fabricated in this work

Scheme 1.



Results and discussion

Fig. 2 shows the TGA of MgBIQ that possesses a maximum rate of weight loss occurring at 501℃ and no weight loss was observed at the temperature lower than 362℃. Above 600℃, there is about 16 wt % of residue composed of Magnesium ash. This Magnesium complex is reasonably stable upon exposure to air and exhibited a very high thermal stability in nitrogen, which is attributed to the fact that the Mg-N (imidazole) bond is highly polarized [8, 9]. The DSC curve of MgBIQ, Fig. 3, shows that the melting transition (Tm) of MgBIQ is 436℃and no glass transition temperature (T_g) was observed up to 430℃. The DSC results indicate that the MgBIQ possesses a very high transition temperature, which may serve as an advantage for the fabrication of organic light emitting device because the use of the materials with high transition temperature as the active emissive layer or carrier transporting layer may provide the device with greater longevity [10, 11].

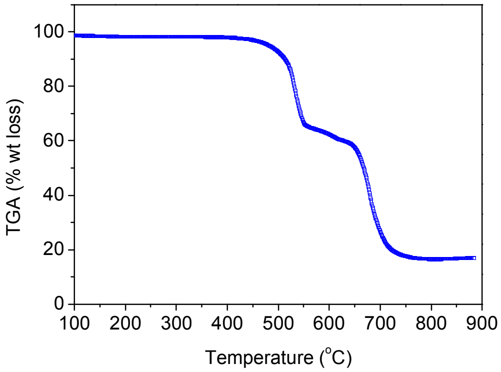


Fig. 2. TGA curve of MgBIQ.

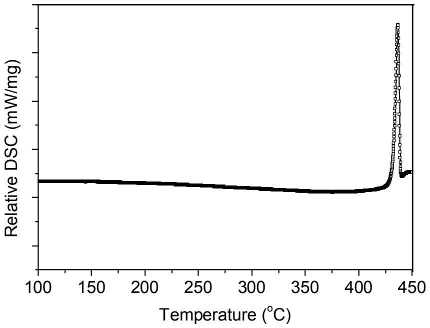


Fig. 3. DSC curve of MgBIQ, under nitrogen gas flow at the heating rate of 20℃/min.

The Photoluminescent (PL) spectra of the MgBIQ solutions and neat film, excited with 323 nm laser line, were illustrated in Figure 4. At low concentration, 5×10^{-5} M in DMSO, only one emission band is observed with maximum at 436 nm, corresponding to the relaxation of MgBIQ from the excited state of a single molecule into ground state. Besides the 436 nm band, a new emission band appeared while the concentration of MgBIQ increased from 5×10^{-5} to 1×10^{-3} M. This new emission band having a maximum at 500 nm is observed in the spectrum of the MgBIQ neat film. We have assigned this new emission band to the excimer and higher aggregates emission [12,13] resulting from the relaxation of collision complex into the lower energy state.

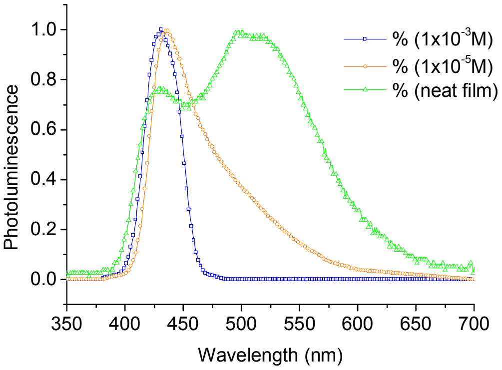


Fig. 4. Photoluminescent spectra of the MgBIQ in solutions and neat film

The EL spectrum of organic light emitting device at the bias voltage of 11.5 V, Fig. 5, shows the broad emission band in the 550-700 nm region with the maximum at 596 nm. The emission is almost fixed in the red region in the CIE coordinate of x = 0.46 y = 0.46, Fig. 6. For the small molecular organic materials, to develop the new type of material with red emission is very important because this kind of material is very seldom prepared so far, and it is very important for the fabrication of full color display panels.

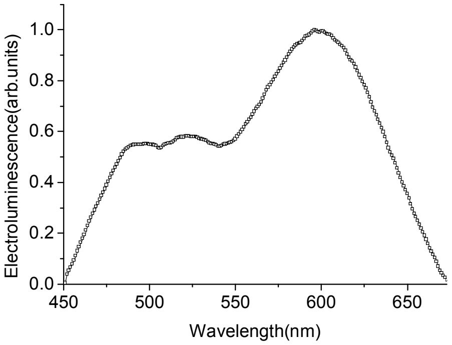


Fig. 5. EL spectrum of OLED fabricated in this work.

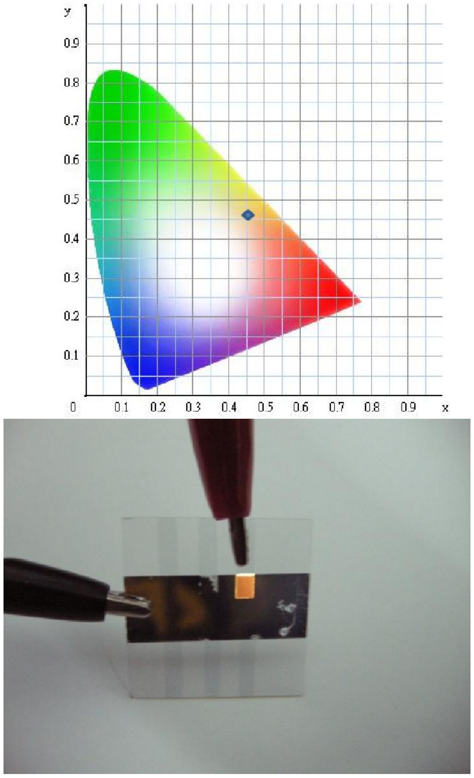


Fig. 6. CIE color coordinates (CIEx,y = 0.46，0.46) for the light emission produced by the OLED devices.

Figure 7 shows the energy level diagram of the HOMO and LUMO of the different organic materials and the work function of cathode and anode. The LUMO energy of MgBIQ is 2.6eV determined form the HOMO energy (5.9eV) obtained from the cyclic voltammetry (CV) method and the optical band gap estimated from the absorption onset. Comparing the energy level of MgBIQ with NPB, it is clear that the MgBIQ has the much higher hole injection barrier than that of NPB; in fact, it is impossible for the hole injection from ITO into MgBIQ without the assistance of NPB or some other kind of HTLs. This diagram also pointed out that the Alq_3 has the lower electron injection barrier than that of MgBIQ, so the electron injection from the MgAg into MgBIQ will be enhanced and confines the recombination zone at the interface between NPB and MgBIQ. Fig. 8 shows the current-voltage and luminance- voltage characteristics of this device having a low turn on voltage of about 5.5V for current and luminance. This device shows a brightness of 1315 cdm^{-2} at the driving voltage of 11.5V with current density of $167 mA/cm^2$.

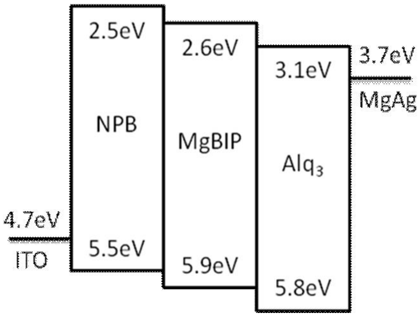


Fig. 7. Energy level diagram of OLED materials, ITO, and Mg-Ag alloy

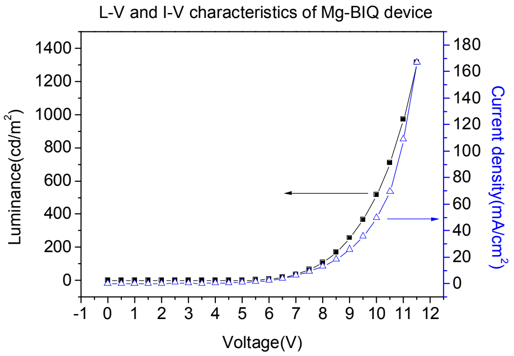


Fig. 8. Current-voltage and luminance- voltage characteristics of OLED fabricated in this work.

4. Conclusion

A novel metal complex, bis (2-(benzimidazol-2-yl) quinolinato) magnesium, was successfully prepared by the reaction of benzimidazol-2-yl-quinoline and dibutylmagnesium. Because of its high thermal stability and excellent electrical characteristics, MgBIQ and its related compound suggest a possible application for the use of the organic light emitting devices.

Acknowledgements

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