

Study on current–voltage characteristics of OLEDs using Alq₃ as the electron transport layer

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Received 10 September 2011, received and revised from 29 September 2011

Abstract. This paper describes changes in current-voltage (I-V) characteristic of an organic light-emitting device (OLED) stacked as a multilayer of ITO/MEH-PPV/Alq₃/Al. The ordered, stacked ITO/MEH-PPV/Alq₃/Al multi-layers were fabricated by spin-coating and thermal vacuum evaporation methods. First, dissolved MEH-PPV solution was spin-coated on ITO-electrodes that had been covered on a glass-slide. Subsequently, an Alq₃ layer was evaporated thermally on the MEH-PPV-coated layer. Finally, an Al-electrode was evaporated also on the electron transport layer. UV-vis absorption and photoluminescent characterization of the MEH-PPV and Alq₃ layer as well as their surface images were performed. All spectra obtained revealed the MEH-PPV and Alq₃ were deposited as given structure. The I-V characteristics show the present of the Alq₃-electron transport layer deposited between the MEH-PPV emissive layer and Al-cathode could enhance current-voltage characteristic reducing the threshold voltage and turn-on voltage.

Keywords: UV-vis absorption, photoluminescence, electron transport layer, current-voltage characteristics.

1. Introduction

Organic light-emitting devices (OLEDs) are composed of one or more thin layers of organic materials sandwiched between two conductors. An applied potential across the contacts injects electrons and holes from the cathode and anode, respectively. Then, a combination of holes and electrons in an emissive layer would generate visible light.

In a well-designed device, there is a high probability of photon emission from the excited state, as well as a balanced number of electrons and holes injected from both the cathode and anode, respectively. However, in a conducting polymer used for a device, some defects exist in its chains. It is these defects that are considered as traps to keep electrons and they prevent the electron mobility. Hence, the hole mobility is usually much easier than the electron mobility inside the multilayer-OLED devices. This effect usually causes unbalanced numbers between injected holes and injected electrons.

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Therefore, to resolve this problem, one of approaches is to enhance electron injection at the interface of cathode/ electron transport materials (ETM) by using low work function metals [1-2] or introducing a thin layer of electron injection material with high electron affinity between the cathode and ETM in order to increase electron injection through stepwise injection from the cathode [3, 4]. Charge injection balance is an extremely important issue in achieving high efficiency OLED devices. During device operation, unbalanced injection of electrons or holes will result in non-irradiative recombination of the charge carrier species at the organic/ cathode or the organic/anode interfaces [5]. For studying a device structure with charge-balanced operation, one needs to consider both the effect of the energy barriers on charge injection and the effect of e/h mobility on charge transport.

Aluminum tris (8-hydroxyquinoline) (Alq_3) is a thermally stable, highly fluorescent material with excellent electron-transport mobility. A limitation associated with Alq_3 is its poor process-ability because the Alq_3 layer must be vacuum-deposited. However, the addition of the Alq_3 layer between an emissive layer and a cathode electrode could enhance a number of injected electrons arriving at the interface of the emissive/ Alq_3 layer.

In this paper, we have reported changes in current-voltage (I-V) characteristics of the organic light-emitting devices (OLED) based on the structures of MEH-PPV monolayer (ITO/MEH-PPV/Al) and bilayers MEH-PPV/ Alq_3 (ITO/MEH-PPV/ Alq_3 /Al). The difference of current intensity, and turn-on voltage corresponding the MEH-PPV monolayer and MEH-PPV/ Alq_3 bilayer as well as effects of these characteristics on performance and luminescent efficiency were studied.

2. Experiments

Organic small molecules, Alq_3 , and conducting polymer, poly(2-methoxy-5-(2'-ethyl-hexyloxy)-p-phenylene vinylene) (MEH-PPV), ITO-covered glass-slides as well as toluene solvent were purchased from Aldrich-Sigma Co. Ltd. Molecular structures of MEH-PPV and Alq_3 substances are shown in left side and right one of the figure 1, respectively. A single polymer-monolayer (ITO/MEH-PPV/Al) and polymer-bilayer (ITO/MEH-PPV/ Alq_3 /Al) of the OLED device were fabricated using the corrosion method creating ITO-electrodes, subsequently the spin-coating and thermal vacuum-evaporation techniques to deposit the MEH-PPV and Alq_3 layers, respectively. Finally, Al-electrodes were thermally vacuum-evaporated on the polymers to make a single-layer and bilayer devices.

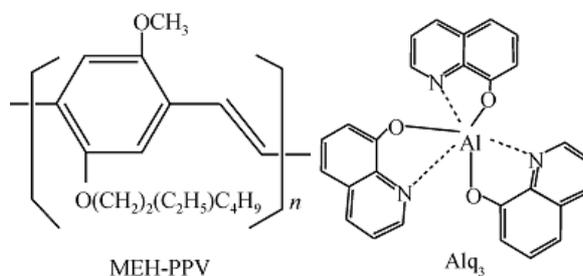


Fig. 1. The structure of MEH-PPV (left) and Alq_3 (right).

First, used as anode-electrodes of OLED devices, an ITO-deposited glass-slide was cut into pieces of 10 x 10 mm. A part of ITO-layers was then marked, avoiding to be corroded, with the solution composed of nitrocellulose, and the rest of them (without the mark) was corroded by the mixed solution of HCl: H₂O: HNO₃ response to 4:2:1 ratio, respectively. Finally, the marked layer on the ITO-layer was removed in sonicated acetone-solution for 30 min. The ITO-electrodes on the glass pieces (so-called samples) were cleaned in de-ionized water, and dried in an oven.

Prior to spin-coating to fabricate a film, the solution of MEH-PPV dissolved in toluene at a rate of 2mg/ml. Stirred in 10 min., subsequently dispersed in an ultrasonic bath for about 30 min., a MEH-PPV-layer was spin-coated on the ITO-anode at a speed of 1300 rpm for 1min., then baked in vacuum oven at 80°C for 30min.

Playing a role of the electron transport layer, Alq₃ was deposited on the MEH-PPV layer by thermal vacuum evaporation of Alq₃ powder for 90s in a chamber of the thermal vacuum-evaporation coater. The sample was then taken out of the chamber after its temperature cooled down to room one, avoiding sample's surface oxidation.

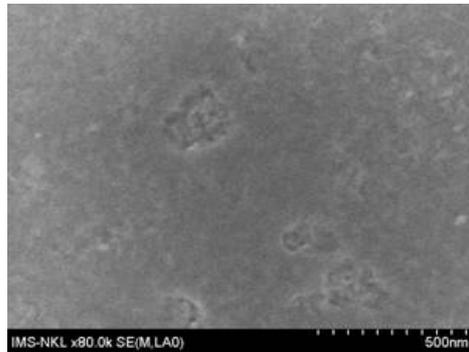


Fig. 2a. SEM micrographs showing some aggre-gated molecules on the surface of the MEH-PPV film.

Similarly, the Al-electrode served as the cathode was deposited on the Alq₃-layer by thermal vacuum-evaporation of an Al pure wire at a chamber pressure of about 10⁻⁴ Torr for 90s. The thickness of the Al-cathode layer was estimated about 80 nm. This value is also equal to the thickness, measured by a surface profiler KLA-Tencor α -step at the scan length of 10nm and scan speed of 2 mm/s.

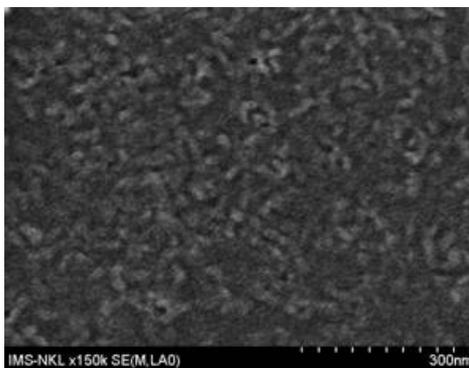


Fig. 2b. SEM micrograph showing rough, well-uniformed surface of the thermally evaporated Alq₃ layer.

Besides, the MEH-PPV and Alq₃ layer was also fabricated for optical characterization of each layer. Images of the deposited layers were identified by observing them on an ultra high resolution scanning electron microscope (FE-SEM), a model Hitachi S-4800. UV-vis adsorption and luminescent emission properties of each layer were determined by a UV-vis/NIR spectrophotometer (model Jasco V-570) at the bandwidth of 1.0 nm, a scanning speed of 200nm/min. and within the range of 200-800 nm. Photoluminescent emission of MEH-PPV and Alq₃ layers was performed with the excitation at a wavelength of 325 nm at the room temperature. And also, electronic transport in the ITO/MEH-PPV/Alq₃/Al devices was studied via current-voltage (I-V) curves taken using a 2400 Keithley source meter by sourcing voltage across the ITO (positive) and Al (negative) electrodes and measuring the resulting current.

3. Results and Discussion

Spin-coated MEH-PPV and thermally evaporated Alq₃ layers were identified by observing their images on the SEM. As can be seen, the surface of the spin-coated layer is rather well-uniformed with some MEH-PPV clusters (high molecular density). It might be attributed to the dissolved polymer, which was accumulated due to the solvent's evaporation itself, causing its viscosity during the spin-coating process. Hence, the MEH-PPV molecules were hardly uniformly dispersed on the ITO surface.

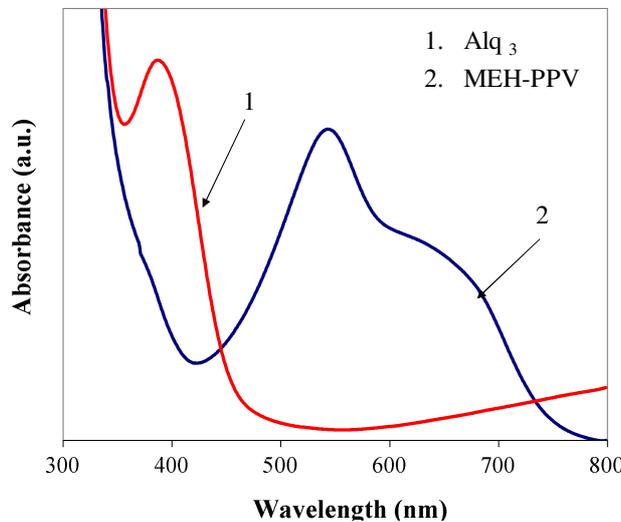


Fig. 3. Typical UV-Vis absorption spectra of Alq₃ (1) and MEH-PPV (2) thin films deposited on glass-slides.

Not similar to the surface of the MEH-PPV layer, the SEM micrograph revealed the rough and well-uniformed surface of the thermally-evaporated Alq₃-layer which can enhance the contacted surface area between the Alq₃ layer and the Al-electrode. This might be a factor that could improve the electron injection from the Al-cathode to the electron transport layer.

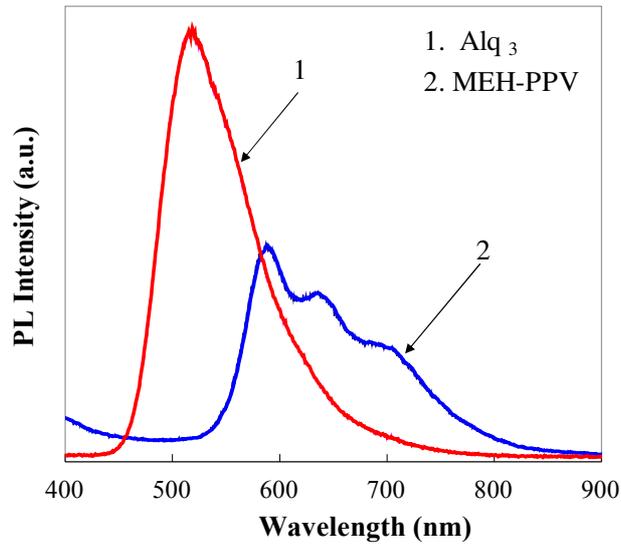


Fig. 4 Photoluminescent spectra of Alq₃ (1) and MEH-PPV (2) excited by a laser at the wavelength of 325 nm.

Absorbance peaks of MEH-PPV and Alq₃ (in Fig. 3) were located at the wavelength of 543 nm and 387 nm, respectively. Their peak-absorbance was in a near- and visible region. These peaks are rather similar to those reported by Djuricic *et al.* [6]. Moreover, It is obvious that the absorption peaks at about 387 nm originate from the π to π^* transition of organic backbone of Alq₃. The absorption peak is in a good agreement with the absorption valuation reported by X. Chunxiang *et al.* [7]. Based on the UV-Vis peak absorbance of both polymers, the excitation of the wavelength of 325nm, generated by a He-Cd laser source, was selected for the photoluminescent characterization.

Photoluminescent spectra of both polymers with the excitation of the wave length of 325 nm for MEH-PPV- and Alq₃-polymer film were shown in Fig. 4. The peak emitted of Alq₃ was found at 517 nm [7] while the emission peak of MEH-PPV was located at 587 nm with a shoulder at 635 nm at RT. These spectra, being a good evidence of the polymers deposited on each other, are suitable to those being reported by Reddy *et al.*[8, 9]

The I-V characteristics of the single layer device (ITO/MEH-PPV/Al) and the bilayer-device (ITO/MEH-PPV/Alq₃/Al) were shown in Fig. 5. . The simplest I -V characteristic involves a resistor, which according to Ohm's Law exhibits a linear relationship between the applied voltage and the resulting electric current. However, depending on characteristics of the conducting polymers sandwiched between two electrodes, the device's resistor can produce non-linear curve exhibiting diode-like behavior.

For the MEH-PPV monolayer device (ITO/ MEH-PPV/Al), the MEH-PPV served as a hole-transport layer. As the hole-mobility is much greater than the electron-one, and due to the mobility imbalance, hence running through the MEH-PPV, a number of hole accumulated at the interface of the polymer and Al-electrode. It resulted in the light-emission region located near the Al-cathode, and in the low resistor. Therefore, the I-V curve of the MEH-PPV monolayer diode shows a large value of the turn-on voltage, and low current intensity when the voltage was over the value of 1.7 -1.8 V.

Not similar to single layer device, for the (ITO/MEH-PPV/Alq₃/Al) bilayer device with Alq₃ layer served as an electron transport one, the light-emission region would be located near the MEH-PPV/Alq₃ interface. Because the HOMO (highest occupied molecular orbit) level of Alq₃ (5.9 eV) lay down lower than that of MEH-PPV (see Fig. 6) and in Alq₃ the electron mobility is superior to the hole one. Thus, Alq₃ layer served as an effective hole broker, it is difficult for holes to cross the Alq₃ layer.

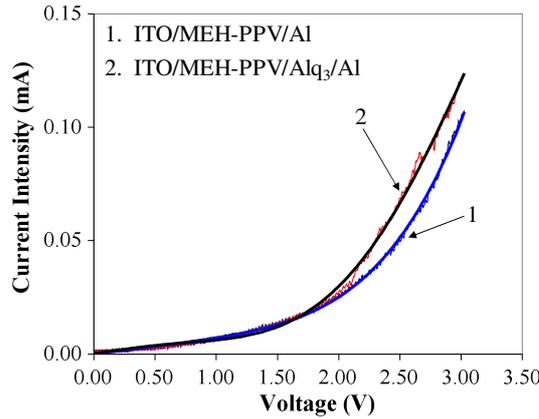


Fig. 5. Current-voltage characteristics of (1) the (ITO/MEH-PPV/Al) monolayer device & (2) the (ITO/MEH-PPV/Alq₃/Al) bilayer device .

Moreover, in the bilayer device, the Alq₃ layer was thicker than the MEH-PPV one, so holes could be accumulated near the MEH-PPV/Alq₃ interface until electrons arrived. Therefore, the I-V characteristics of the MEH-PPV/Alq₃ bilayer device demonstrate a larger value of the turn-on voltage (1.7-1.8 V), and higher current intensity than these values of the MEH-PPV monolayer device. Moreover, the I-V curve of the bilayer device looks more identical to that of a diode in comparison with the I-V curve of the single-layer device (Fig. 5). This might be attributed to increase in the resistor due to the deposition of Alq₃ of the bilayer devices that demanded a larger turn-on voltage. And also, the Alq₃ layer adjacent to the Al cathode acted as an electron-transporting layer that can also prevent excitons quenching close to the cathode. However, a number of holes and electrons that could be recombined at the interface of MEH-PPV/Alq₃ bilayer, considerably increased. It resulted in the enhancement of luminescent efficiency and the lifetime of the ITO/MEH-PPV/Alq₃/Al device.

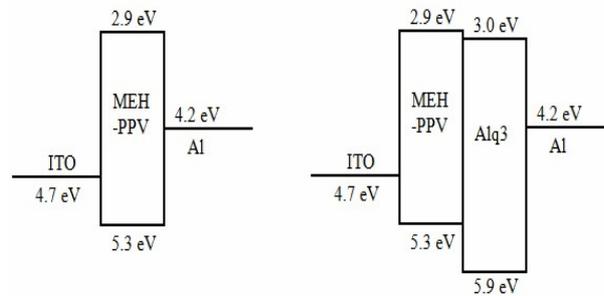


Fig. 6 Barrier energy at HOMO-LUMO level of MEH-PPV and Alq₃.

4. Conclusions

The addition of the Alq₃ electron-transport layer sandwiched between Al-cathode and MEH-PPV by the thermal vacuum evaporation have improved the current-voltage characteristics of OLEDs based on the bilayer structure (ITO/MEH-PPV/Alq₃/Al). The I-V curve of the bilayer device demonstrated diode-behavior while the curve of the monolayer device revealed a resistor behaviour only. This could be ascribed to Alq₃ serving as the hole-blocking that improved the balance of electron and hole current. It resulted in stability and luminescent efficiency of the OLEDs.

Acknowledgments

The author would like to thank the strong support and financial aids of Vietnam national University, Hanoi for the QG.10.42 project in the 2010-2012 academic years. My gratitude is also extended to Professor Nguyen Nang Dinh, Faculty of Engineering Physics & Nanotechnology for useful discussion on charge mobility through hetero-junction of MEH-PPV- and Alq₃-layers. Thanks are also extent to the BSc Van Trong Nghia, a former student of the Faculty Engineering Physics & Nanotechnology, UET-VNU, Hanoi for the sample preparation and providing SEM's micrographs.

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