Solvent Effect on the Electrical and Structural Properties for MEH-PPV Organic Light Emitting Diodes (OLED)

N A N Ismail1, S Shaari1, N Juhari1,2, N Sabani1, M F Ahmad1 and N F Zakaria1

1 School of Microelectronic Engineering, Pauh Putra Campus, 02600 Arau, Perlis, MALAYSIA.
2 Institute of Nano Electronic Engineering, Universiti Malaysia Perlis, Lot 106, 108 & 110, Blok A, Taman Pertwi Indah, Jalan Kangar-Alor Setar, Seria, 01000 Kangar, Perlis, MALAYSIA.
nurulafiqahnorismail@yahoo.com, safizan@unimap.edu.my

Abstract. In this paper, the performance of the electrical properties (J-V) and surface roughness of MEH-PPV based organic light-emitting diodes (OLED) towards solvent effect was investigated. The MEH-PPV layer was deposited using spin coating technique at fixed spin speed of 3000 rpm. Two different solvents, toluene and chloroform (CHCl3) and mixture toluene:CHCl3 with the ratio of 4:1 and 1:4 were used to dilute MEH-PPV at fixed concentration of 5 mg ml-1. Apparently, the mixture of solvent makes the surface roughness of the MEH-PPV films reduced to 0.15 nm and 3.59 nm under the ratio 4:1 and 1:4 respectively. Besides, the mixture solvents makes the value of turn on voltage was dropped to ~7.2 V and ~9 V respectively compared to non-mixture solvent. The combination of different solvent apparently gives an effect on the electrical and structural properties of organic light-emitting diode.

1. Introduction

Organic light-emitting diode (OLED) that attracts the attention of engineers and scientist around the world [1] have been developed successfully and entered the marketplace. Despite of OLED great performance in display technologies, OLED can be made entirely by solution process which suitable for white lighting panels and large area flexible display panel [2]. However, most of the highly efficient OLED are fabricated by thermal evaporation rather than solution process. This is because thin films fabricated by solution process has few disadvantages such as phase separation [3,4] between different materials, low packing density [5,6] and poor surface morphology [5, 7-8]. Despite that, due to its low manufacturing cost and simple fabricating process, solution process is still a hot topic in the research world [9].

Based on our previous research, surface roughness can be reduced when the annealing temperature of thin films increased. However, organic material mostly have low glass transition temperature, such as poly [2-methoxy-5(2’–ethylhexyloxy)-1, 4-phenylenevinylene], MEH-PPV (65 °C) [10]. Thus the annealing temperature must be higher than the glass transition temperature but not exceed the melting point of the material (250 °C – 300 °C) [11]. In addition, selection of solvent have an effect on the thin films formation. Solvent that evaporate too fast may lead to rough surface morphology [5], if too slow may cause solvent residue [6]. Thus, in this experiment we used mixture of toluene:CHCl3 solvents to increase the electrical performance and to improve the surface roughness.
Previous research by Nguyen et al [12] investigated the performance of conjugated polymer based devices where MEH-PPV was diluted in THF and chlorobenzene. It is reported that MEH-PPV diluted in chlorobenzene produced low turn on voltage. Low quantum efficiency and higher current injection compared to MEH-PPV diluted in THF. The results in the research observed the changes on the performance of the devices based on the current-voltage and brightness-voltage characteristics where some changes were reported as the solvent varied. Therefore, it is necessary to investigate the electrical and structural properties of MEH-PPV thin films in organic light-emitting diode.

In this paper, the investigation of \( J-V \) characteristic of OLED device with the configuration of ITO/MEH-PPV/Al was carried out when two different solvents and mixture of them were used in this experiment. The mixture of solvent apparently reduced the turn on voltage compared when the device was diluted in the original solvent. Interestingly, the surface roughness also reduced to 14% with the mixture of 4:1 meanwhile 6% for 1:4 ratio of solvent. The orientation of the solvent at the ITO surface need to be understood in order to investigate the decreasing value of surface roughness and turn on voltage.

2. Experimental

2.1. Fabrication processes

In this experiment, the MEH-PPV used was purchased from Sigma Aldrich with the molecular weight of \( \sim 40,000 – 70,000 \) Mw. The concentration used for MEH-PPV solution is 5 mg/ml, where 5 mg of MEH-PPV was diluted in three solvents, 1 ml toluene, 1 ml chloroform and 1 ml mixed solvents of toluene:CHCl\(_3\) (4:1 and 1:4 ratio), respectively. Before the fabrication process started, the indium-tin-oxide (ITO) glass substrate were cleaned in ultrasonic bath with deionized (DI) water, ethanol and acetone for 10 minutes each. After the cleaning process, the ITO glass substrate were dried for 2 minutes with temperature of 150 °C for 15 minutes. Lastly, the aluminium (Al) were deposited as cathode by thermal evaporation. This OLED device active area is 0.5 cm\(^2\). The device configuration of ITO/MEH-PPV/Al are shown in Figure 1.

2.2. Device characterization

The surface roughness of the thin films were analyzed by using atomic for microscopic (AFM) with the scanning area of 5 \( \mu m \). The \( J-V \) measurement were measured using semiconductor parameter analyzer (SPA) (Keithley 4200-SCS from Tektonik Company). The voltage applied for the device to operate was varied from 0 V to 20 V.

![Figure 1. Schematic illustration of structure of ITO/MEH-PPV/Al device.](image)
3. Result and Discussion

3.1. Surface morphology of ITO/MEH-PPV

The structural properties of MEH-PPV thin films diluted in four different solvents were comparatively investigated in Figure 2. Figure 2(c) has the lowest surface roughness with 1.254 nm while Figure 2(b) has the highest surface roughness with 58.72 nm. This shows that mixed solvent with 4:1 ratio has smooth surface compared to the other solvents. In order for OLED to have a better performance, smooth surface of thin films are required in the making of OLED devices. Previous research done by Reshak et al showed thin films with smooth surface produced higher current in the $I-V$ characteristics [13]. The other device with rougher surface of thin films has low performance. Both thin films where MEH-PPV diluted with high ratio of toluene produced low surface roughness compared to thin films where MEH-PPV diluted with high ratio of chloroform. The surface roughness of thin films with 100% of toluene was reduced to 0.2 nm while thin films with 100% of chloroform was reduced to 3.6 nm. The analysis proved that thin films fabricated with mixed solvent for 4:1 and 1:4 ratio can improve the film morphology and smoothen the thin films surface. Smooth thin films surface helps in forming a better interface with the electron transport layer (ETL), which gives advantages to electron injections [8].

Chloroform, a solvent that is easy to vaporize because of lower boiling point (62 °C) which result in poor quality thin films [14]. This explained the results where the surface roughness of MEH-PPV diluted in chloroform and 1:4 ratio has high surface roughness. Toluene has higher boiling point at 110 °C where the solvent is not easy to vaporize. Surface roughness of MEH-PPV diluted in mixed solvent with 4:1 ratio produced the lowest surface roughness among all four. This is because of the value of toluene mixed with chloroform present in the solvent for MEH-PPV where it helps in improving the thin film uniformity.

![Figure 2. The AFM images of ITO/MEH-PPV with (a) toluene (Ra = 1.404 nm) (b) chloroform (Ra = 58.72 nm) (c) mixed solvent with 4:1 ratio (Ra = 1.254 nm) (d) mixed solvent with 1:4 ratio (Ra = 55.13 nm).](image-url)
3.2. J-V characteristic of OLED

Figure 3(a) and 3(b) shows the J-V characteristic of OLED device with the configuration of glass substrate/ITO/MEH-PPV/Al. Based on the figures, both devices with high ratio of toluene produced higher current density compared to devices with high ratio of chloroform. The turn on voltage for both devices with high ratio of toluene also different where turn on voltage for device with 100% of toluene is 12 V and device with 4:1 ratio of mixed solvent is 7.2 V. for the devices with high ratio of chloroform, device with 100% of chloroform turn on voltage is 14 V and device with 1:4 ratio of mixed solvent is 9 V. Based on the overall performance of J-V characteristics, devices with high ratio of toluene have better performance. When compared in terms of the surface roughness, it shows that the devices with low surface roughness produce low surface roughness and high current density with low turn on voltage. It is clear that devices with MEH-PPV thin films diluted in 100% toluene and mixed solvent with 4:1 ratio produced higher electron and hole current which result in low turn on voltage. This is correlated with the improved surface roughness in the MEH-PPV thin films.

Previous research by Xiao et al reported that devices with toluene as solvent for thin films dilution produced low turn on voltage and has better performance [15]. Additionally, low turn on voltage correspond to the higher current density values [16].

![Figure 3. J-V measurement of devices of glass substrate/ITO/MEH-PPV/Al with (a) comparison between toluene and chloroform (b) comparison between mixed solvent (4:1 and 1:4 ratio).](image-url)
4. Conclusion
The electrical and structural properties of solution-processed OLEDs formed by diluting the organic material for emissive layer with four different solvents were investigated. The mixing ratio of MEH-PPV were optimized and good performances were obtained. MEH-PPV thin films with high ratio of toluene as solvent produced better film uniformity, low surface roughness and low turn on voltage which result in further improvement of solution-processed OLEDs. This can be one of the possible alternative route in fabrication of OLEDs with low devices cost for lighting applications.

References
[1] Andreopoulou, A. K., Gioti, M., & Kallitsis, J. K. 2019 Solution-Processable Components for Organic Electronic Devices, 413-482.
[2] Zhong, C., Duan, C., Huang, F., Wu, H., & Coa, Y. 2011 Chemistry of materials, 23(3), 326-340.
[3] Ngome, F. O. O., Kim, Y. T., Lee, H. D., Kim, Y. H., Lee, T. W., & Park, C. G. 2017 Journal of Materials Chemistry C, 5(37), 9761-9769.
[4] Yao, B., Lin, X., Zhang, B., Wang, H., Liu, X., & Xie, Z. 2018 Journal of Materials Chemistry C, 6(16), 4409-4417.
[5] Mao, G., Wu, Z., He, Q., Jiao, B., Xu, G., Hou, X. & Gong, Q. 2011 Applied surface science, 257(17), 7394-7398.
[6] Lee, T. W., Noh, T., Shin, H. W., Kwon, O., Park, J. J., Choi, B. K. & Kim, Y. R. 2009 Advanced Functional Materials, 19(10), 1625-1630.
[7] Strawhecker, K. E., Kumar, S. K., Douglas, J. F., & Karim, A. 2001 Macromolecules, 34(14) 4669-4672.
[8] Yang, J., Song, D., Zhao, S., Qiao, B., Xu, Z., Wang, P., & Wei, P. 2019 Organic Electronics, 71, 1-6.
[9] Zhou, L., Xu, Z., Song, D., Zhao, S., Qiao, B., Chen, J., & Zheng, W. 2020 Synthetic Metals, 261, 116322.
[10] Chou, H. L., Lin, K. F., & Wang D. C., 2006 Journal of Polymer Research, 13(1), 79-84.
[11] Chi, S. H., Hales, J. M., Cozzoul, M, Ochoa, C., Fitzpatrick, M., & Perry, J. W. 2009 Optics express, 17(24), 22062-22072.
[12] Nguyen, T. Q., Kwong, R C., Thompson, M. E., & Schwartz, B. J. 2000 Applied Physics Letters, 76(17), 2454-2456.
[13] Reshak, A. H., Shahimin, M. M., Juhari, N., & Suppiah, S. 2013 Progress in biophysics and molecular biology, 113(2), 289-294.
[14] Wang, Z., Naka, S., & Okada, H. 2011 Japanese Journal of Applied Physics, 50(1S2), 01BC06.
[15] Xiao, S., Qiu, C., Jin, E., Chen, Y., Louis, P., Qiu, S., & Shih, I. 2005 Material Letters,59(6), 694-696.
[16] Liu, J., Shi, Y., Ma, L., & Yang, Y. 2000 Journal of Applied Physics, 88(2), 605-609.