International Journal of Nanoelectronics and Materials
Volume 14 (Special Issue) December 2021 [341-347]

Optimization of MEH-PPV Based Single and Double-Layer TOLED Structure by Numerical Simulation

T. Kersenan[1], N. F. Zakaria[1,2*], S. Shaari[1], N. Sabani[1], N. Juhari[1,2], M. F. Ahmad[1,2], and A.F.A Rahim[3]

[1] Micro and Nano Electronics (MiNE) Research Group, Faculty of Electronic Engineering Technology, Universiti Malaysia Perlis (UniMAP), Perlis, Malaysia.
[2] Advanced Communication Engineering (ACE) Centre of Excellence, Faculty Electronic Engineering Technology, Universiti Malaysia Perlis, Arau 02600, Perlis, Malaysia
[3] Faculty of Electrical Engineering, Universiti Teknologi MARA, Cawangan Pulau Pinang, Malaysia.

ABSTRACT

In this work, we simulated and characterized Poly [2-methoxy-5-(2'-ethylhexyloxy)-1, 4-phenylene vinylene] (MEH-PPV) based single and double-layer TOLED by using Silvaco ATLAS device simulator to achieve prominent values of electrical and optical properties of the device. MEH-PPV were used as the emitting layer (EML) in the single-layer, while addition of Poly [(3,4-ethylene dioxythiophene)-poly(styrene sulfonate)] (PEDOT-PSS) as the electron transport layer (ETL) were conducted in double-layer TOLED simulation. The EML and ETL thickness in both structures were varied between 10 – 150 nm, respectively, to observe and understand the underlying physics of the relation in the layer thickness to the electrical and optical characteristics. Furthermore, variation of the EML/ETL thickness ratio from 1:1 to 5:1 (with thickness in between 10 to 50 nm) had also been conducted. From this work, it is understood that the thickness of the EML layer plays the most important role in TOLED, and by balancing the carrier injections and recombination rate in appropriate EML/ETL thickness ratio, the electrical and optical properties can be improved. By optimizing the EML/ETL thickness and thickness ratio, an optimal forward current of 1.41 mA and luminescent power of 1.93e-18 W/µm has been achieved with both MEH-PPV and PEDOT-PSS layer thickness of 10 nm (1:1 ratio), respectively. The results from this work will assist the improvement of TOLED device to be implemented widely in low power and transparent electronic appliances.

Keywords: OLED, organic, ATLAS Silvaco, device simulator

1. INTRODUCTION

In recent years, organic materials have replaced traditional semiconductors in many electronic devices, such as organic displays, organic transistors, and organic solar cells. Using organic materials instead of inorganic materials, especially converting light to electricity (photovoltaics) [1–3] and electricity to light (light diodes) [4,5], has obvious economic and ecological advantages. The key explanation on why organic material has been commonly used in the industry is because of the advantage in easy deposition of the substance by evaporation and spin-coating. These techniques of deposition minimize the expense and difficulty of the deposition process. While organic materials were valuable to the microelectronic industry, organic material fits perfectly into the display industries. Organic material has almost 100% of the visible spectrum in internal quantum efficiency [6]. Organic light-emitting diodes (OLED) for example, is generally introduced as an organic electroluminescent (EL) emissions system which is built by utilizing organic materials. An organic polymer such as MEH-PPV which is the PPV derivative, possesses a good optical and electronic properties and is suitable for display manufacturing due to its strong solubility in most popular organic solvents [4], and its capacity

*norfarhani@unimap.edu.my

to generate high-quality optical films. Common researched OLED comprises of a single organic layer placed in between two electrodes; anode and cathode with one-sided transparent surface electrode. Apart from the standard OLED, transparent OLED (TOLED) which implemented double-sided transparent electrode materials is now become one of the focus areas, regarded as high-tech lighting technology and attracted considerable attention from display manufacturers because of the potential to view texts and graphics on a see-through panel.

In this project, simulation and characterization of MEH-PPV based TOLED had been performed to investigate the optimum condition to achieve a functional level of forward current and luminescent power in single-layer and double-layer TOLED. By choosing appropriate thickness and organic material parameters in designing TOLED, carrier recombination inside the emitting layer (EML) can be altered and consequently improve electrical and optical properties which resulted in better performance of the device. The results from this project may assist future development of applications using TOLED.

2. MODELS AND DEVICE PARAMETERS

In this work, we developed TOLED structure which employed ITO-coated glass with work function, ɸ of 4.7 eV, both as anode and cathode, to enable two-way transmission of the emitted light. As shown in Figure 1, the single layer structure was ITO(10 nm)/EML(X nm)/ITO(10 nm), where X=10, 20, 30, … , 150 nm. The EML was Poly [2-methoxy-5-(2'-ethylhexyloxy)-1, 4-phenylene vinylene] (MEH-PPV). Double layer structure consisted of ITO(10nm)/EML(X nm)/ETL(Y nm)/ITO(10 nm), where Poly [(3,4-ethylene dioxythiophene)-poly(styrene sulfonate)] (PEDOT-PSS) were added as the ETL, with variation of thickness Y from 10 to 150 nm (similar to X).

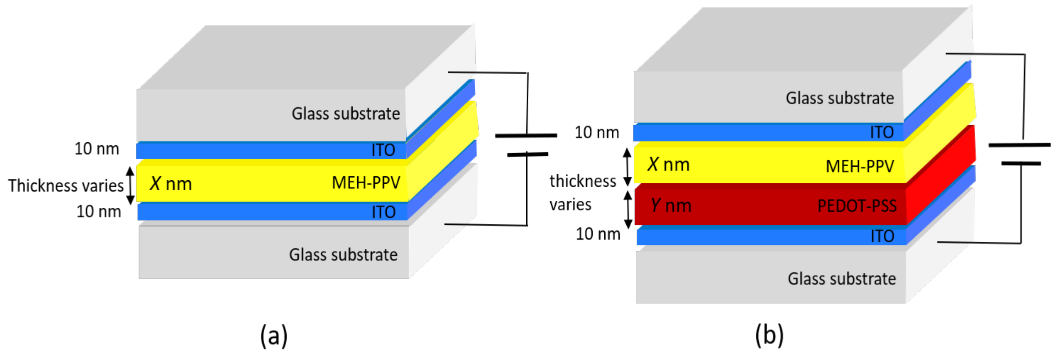


Figure 1. The structure of (a) single-layer and (b) double-layer MEH-PPV based transparent organic light emitting diode.

The simulations were based on time-independent drift-diffusion transport model to model the charge carrier transport in the organic semiconductor. The Poole-Frenkel field-dependent mobility model of

$$\mu_n(E) = \mu_{n0} exp\left(-\frac{DELTAEN.PFMOB}{kT_{neff}} + \left(\frac{BETAN.PFMOB}{kT_{neff}} - GAMMAN.PFMOB\right)\sqrt{|E|}\right) \quad (1)$$

$$\mu_p(E) = \mu_{p0} exp\left(-\frac{DELTAEP.PFMOB}{kT_{peff}} + \left(\frac{BETAN.PFMOB}{kT_{peff}} - GAMMAP.PFMOB\right)\sqrt{|E|}\right) \quad (2)$$

were utilized as the mobility model where μ_n and μ_p are the Poole-Frenkel mobilities for electrons and holes, respectively, E is the magnitude of the electric field, μ_{no} and μ_{po} are the zero field mobilities, and DELTAEN.PFMOB and DELTAEP.PFMOB are the activation energy at zero electric field for electrons and holes, respectively. BETAN.PFMOB is the electron Poole-Frenkel factor, and BETAP.PFMOB is the hole Poole-Frenkel factor. When the applied electric field is large, carrier mobility is not constant, and depends on the temperature and the applied field, and one of the ways to introduce the field effect is to use the Poole-Frenkel effect. In addition, the Langevin bimolecular recombination model of:

$$R_L = \frac{q}{\varepsilon}(\mu_n + \mu_p)np \tag{3}$$

were employed to described the recombination rate of free electrons and holes. Addition of the singlet exciton continuity equations models were also done to infer the radiative rates for luminescence in the TOLEDs simulation [7]. The material parameters for the organic materials used in the simulation are shown in Table 1 [7,8].

Table 1 Material parameters of the organic materials used in TOLED

Parameters	MEH-PPV[7]	PEDOT-PSS[8]
Electron affinity, E_a (eV)	0.897	0.673
Band gap, E_g (eV)	4	3.001
Hole mobility, μ_H ($cm^2V^{-1}s^{-1}$)	0.5×10^{-4}	40
Electron mobility , μ_E ($cm^2V^{-1}s^{-1}$)	0.5×10^{-5}	1
Relative permittivity, ε_r	3.0	2.2
Temperature, *T* (K)	300	300
Richardson constant, *A* ($A\cdot cm^{-2}K^{-2}$)	120	-
Effective density of states for electrons, *Nc* (cm^{-3})	2.5×10^{19}	2.0×10^{21}
Effective density of states for holes, N_v (cm^{-3})	2.5×10^{19}	2.0×10^{21}
The singlet radiative decay lifetime (s)	1.0×10^{-9}	-

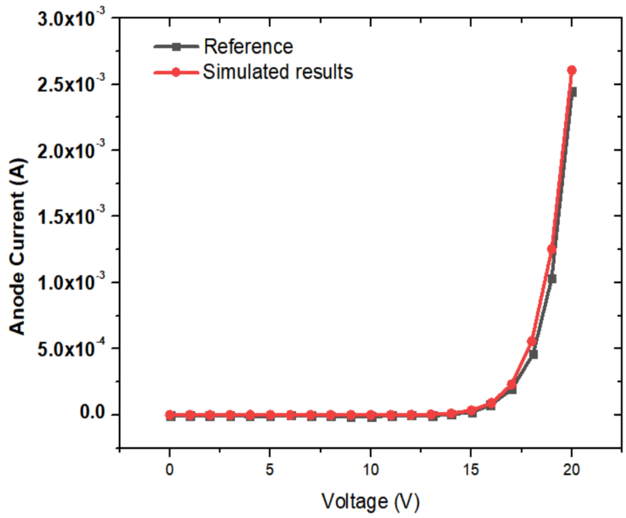


Figure 2. Comparison of simulated I-V characteristics of ITO/MEH-PPV/Ca OLED with reference structure from [7].

To ensure the accuracy of the physical model used in the simulation, we first validated our physical models for OLED characterization with a simulated ITO/MEH-PPV/Ca OLED structure from [7]. The same structural and device parameters were used with additional definition of physical models to simulate the electrical characteristics of the device. The results were then compared. As can be seen in Figure 2, our simulation results are in good agreement with [7] which validated the physical models used in the OLED simulation. Both results showed similar threshold voltage at around 16 V and forward current value of around 2.50 mA and 2.61 mA, respectively with 20 V bias. This validated setting will then be used in the optimization of TOLED using Silvaco ATLAS device simulator. The thickness of the EML and ETL layer of the TOLEDs were varied and their performance in term of electrical and optical properties were observed.

3. MODELS AND DEVICE PARAMETERS

3.1 Electrical and Optical Performance in Single-layer TOLED

Figure 3(a) shows the turn-on voltage, V_{on} in single-layer TOLED with various EML thicknesses. V_{on} of X=10 nm was the lowest at approximately 3.5 V before increased and became constant at 7.5 V from 30 nm onwards. By increasing the EML thickness in TOLED, a raise to direct charge trapping effect can occurred which resulted in poor current efficiency. Effective charge balance was attainable in lower EML thickness as well as reducing direct charge trapping effect [9]. Besides, the efficiency of charge recombination in thick EML could be affected, which results in constant turn-on voltage.

Figure 3(b) shows that the forwards current, I_{fwd} and luminescent power, L_p increased with higher voltage biasing in X=10, 50, 100, and 150 nm, respectively. Furthermore, the maximum L_p and I_{fwd} values at 10 V bias in each thickness were recorded and the values are seen exponentially decreased when X>20 nm (refer Figure 3(c)).

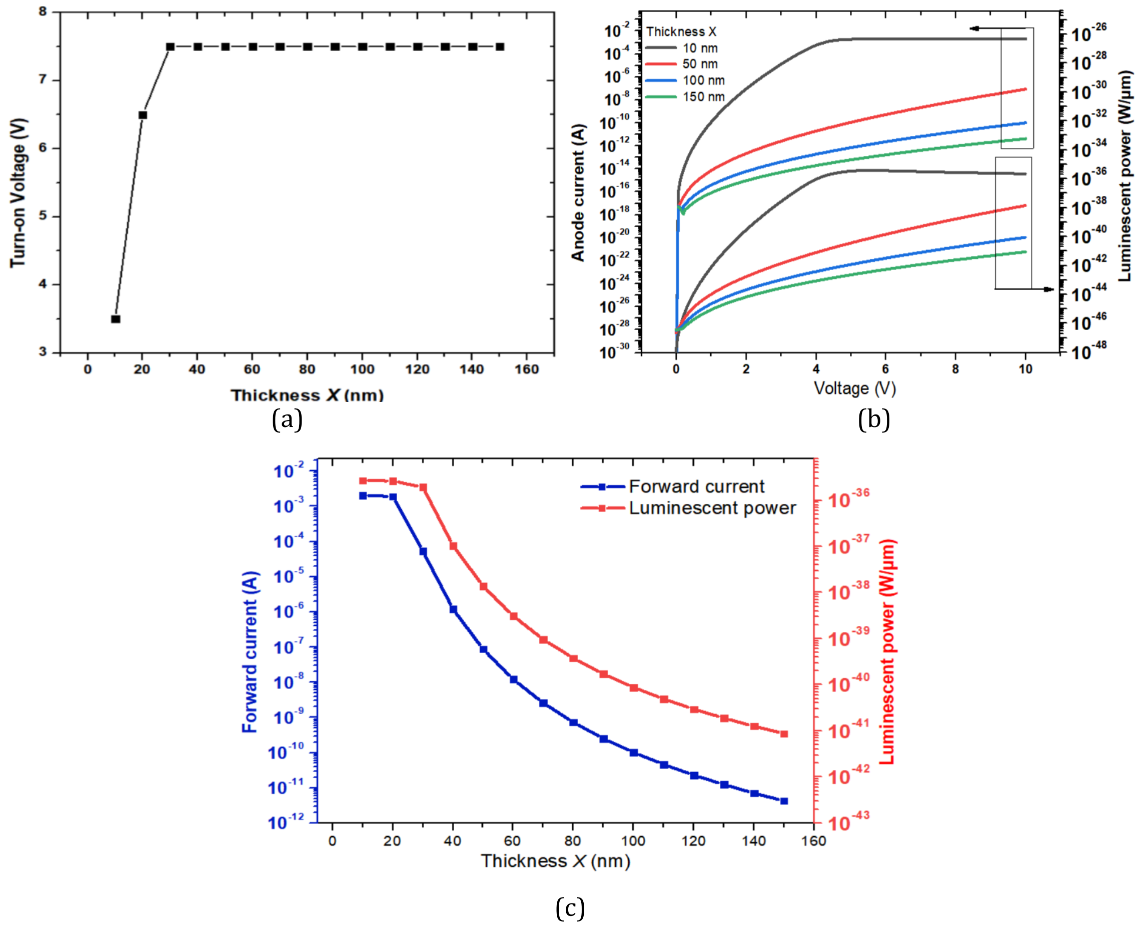


Figure 3. (a) The turn-on voltage, (b) anode current and luminescent power values, and (c) forward-current and luminescent power values in single-layer ITO/MEH-PPV/ITO structure.

Low electrical and optical performance were observed in single-layer TOLED with highest I_{fwd} of ~2.05 mA and L_p of 2.71×10^{-36} W/µm obtained with X=10 nm, respectively. The same trend has been observed in [10] by using Lif/Al as cathode which might due to the exceptional physical properties of TOLED in thinner EML structure as previously researched in [9] where effective charge balance was attained in thin EML thicknes, at the same time reducing direct charge trapping effect.

However, the highest L_p in this single-layer structure shows a very minimal power which is 2.71×10^{-36} W/µm make it unsuitable for most optical characterization. This issue may emerge because of the high energy potential difference between the cathode (ITO) and LUMO of the emitting layer (MEH-PPV) [10] of this single-layer TOLED which is 1.9 eV [as shown in Figure 4(a)], much higher than the optimum potential difference of 1.3 eV to 0.1 eV [7 - 9]. Device with electron injection potential of 1.9 eV contributed to inferior electron injection characteristics which leads to inadequate luminescent power production. To overcome this problem, addition of ETL layer in double-layer TOLED as shown in Figure 4(b) is performed to reduce the energy barrier between cathode and ETL's LUMO to 1.1 eV, to efficiently transport electron into the EML.

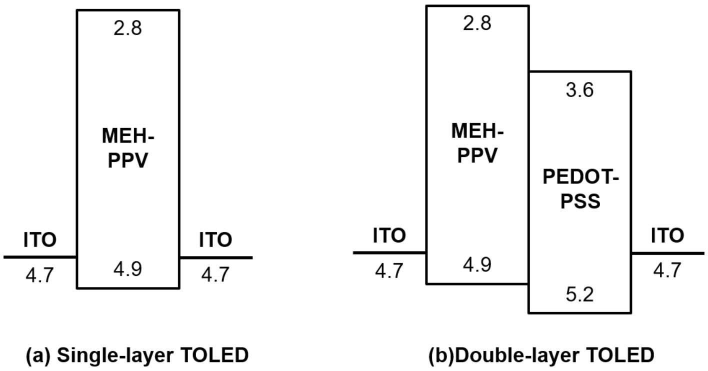


Figure 4. Energy band diagram in Values of anode current and luminescent power in (a) single-layer and (b) double-layer TOLED structure with MEH-PPV and PEDOT-PSS material.

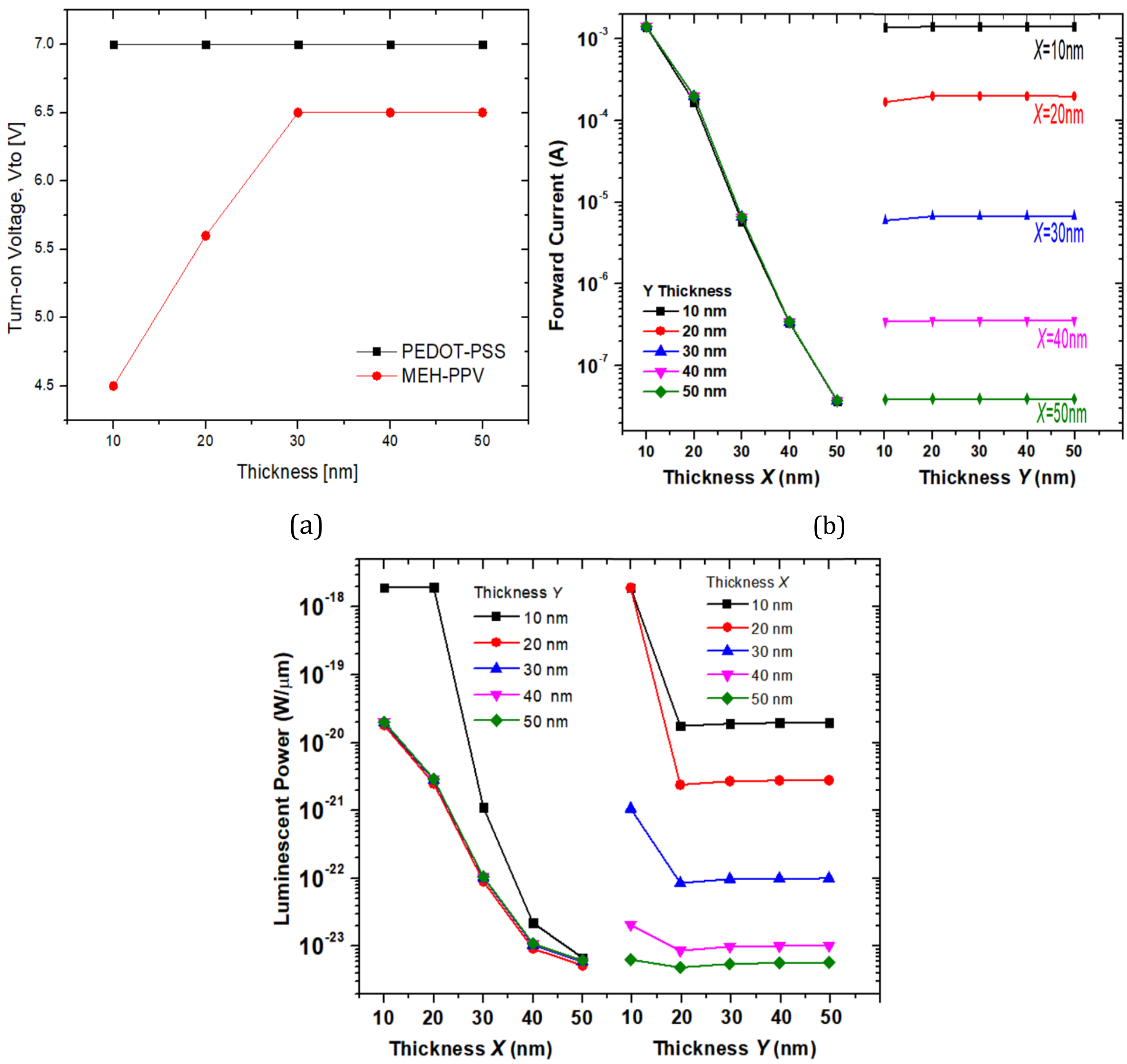
3.2 Electrical and Optical Performance in Double-layer TOLED

From the variation of EML thickness in the single-layer TOLED simulation, it is understandable that the performance of MEH-PPV in smaller thickness structure contributes to better values of TOLED, both in electrical and optical performances. Thus, variation of smaller EML thickness, X of 10 – 50 nm will be considered in the characterization of double-layer TOLED structure.

Figure 5(a) shows the V_{to} for double-layer TOLED in EML and ETL thickness variation between 10 – 50 nm, while the other layer (of ETL or EML) is fixed at 50 nm. As can be seen, the V_{to} increased when the thickness of the EML (X) were increased and ETL (Y) was fixed at 50 nm. The lowest V_{to} has been observed at ~4.5 V with X=10 nm before increased to a constant value of 6.5 V as X approaches 30 nm. Constant V_{to} might arise due to needy hole and electron recombination at particular MEH-PPV layer thickness. While in fixed EML(X) of 50 nm, the V_{to} are seen constant at ~7.0 V for various ETL(Y) thickness. The carrier recombination between hole and electron in EML might be inefficacious, which results in constant electrical and optical properties.

The I_{fwd} values at 10 V biasing voltage in varied EML and ETL thickness from 10 – 50 nm are shown in Figure 5(b). The I_{fwd} were seen linearly decreased with increasing EML thickness(X) with highest I_{fwd} value of ~1.43 mA with X=10 nm. This trend has also been observed in various researches using OLED [7 - 9]. In increased ETL thickness (Y), no significant I_{fwd} changes had been observed when X are fixed [right-side of Figure 5(b)]. Since the mobility of electrons are far smaller than the organic materials holes, the major challenge for the performance of the lighting device is to increase the charge balance. In order to achieve a better charge balance, the capacity to inject electrons and transport must be enhanced. By reducing the thickness of the EML, an improved charge balance can be attained [9] which resulted in increased electrical performance (higher V_{to} and lower I_{fwd}). The same trend has been observed in experimental research in [9].

In term of L_p, the values were seen decreased in increasing EML(X) thickness, as can be seen in the left-side of Figure 5(c). Abrupt changes of the luminescent power values had been observed in 10 to 20 nm thick ETL(Y) layers in all variation of EML thickness, before became nearly constant, independent of the increasing thickness Y. The highest luminescent power has been observed at ~1.9×10^{-18} W/µm with 10 nm ETL thickness with 10 and 20 nm EML thickness, respectively. In order to achieve a better charge balance in a lighting device, the capacity to inject electrons and transport must be enhanced. By reducing the thickness of the EML, an improved charge balance can be attained [9]. The charge balance in thin MEH-PPV layer might improve, resulting in higher electrical and optical properties resulting in better electrical and optical performance which is consistent with the experiment result conducted by Fadavieslam [9]. Meanwhile, higher EML thickness raises the impact of direct charge trapping and decreases current efficiency. In other words, a balanced hole and electron currents can be improved by lowering the MEH-PPV layer thickness.



(c)

Figure 5. (a)The turn-on voltage, (b) forward-current and (c) luminescent power values in double-layer ITO/MEH-PPV/ITO structure with variation of the EML(X) and ETL(Y) thickness.

4. CONCLUSION

Based on simulation of single-layer and double-layer MEH-PPV based TOLED, it can be concluded that the thickness of the EML and ETL layer plays an important part in the electrical and optical performance of TOLED. Addition of ETL layer (PEDOT-PSS) in double-layer TOLED has reduced the energy barrier between cathode and ETL's LUMO to 1.1 eV, which resulted in more efficient transport electron into the EML. And by optimizing the thickness ratio in both layers, it is assumed that the electron injection and charge balanced in the layers can be improved resulting in higher electrical and optical properties. An optimal forward current of 1.41 mA and luminescent power of $1.93e^{-18}$ W/µm has been achieved with both MEH-PPV and PEDOT-PSS layer thickness of 10 nm (1:1 ratio), respectively.

REFERENCES

[1] Jamalullail, N., Mohamad, I. S., Norizan, M. N., Mahmed, N. and Taib, B. N., EPJ Web Conf. vol. **162** (2017) pp. 1–5, 2017.
[2] Norhisamudin, N. A., Sabani, N., Rosli, N. , Ahmad, M.F., Juhari, N., Shaari, S. and Zakaria, N. F. AIP Conf. Proc., Vol. **2203**, No. 1 (2020) p. 020052:1–6.
[3] N. Jamalullail, I. S. Mohamad, M. N. Norizan, N. A. Baharum, and N. Mahmed. "Short review: Natural pigments photosensitizer for dye-sensitized solar cell (DSSC)." in 2017 Proc. IEEE 15th IEEE Student Conf. Res. Dev. Inspiring Technol. Humanit. SCOReD (2017), pp. 344–349.
[4] N. Juhari, N. I. A. Shukri, N. Sabani, S. Shaari, M. F. Ahmad, and N. F. Zakaria, AIP Conf. Proc., Vol. **2203**, No. 1 (2020) p. 020046.
[5] Ismail, N.A.N., Shaari, S., Juhari, N., Sabani, N., Ahmad, M. F. and Zakaria, N. F., Journal of Physics: Conf. Series, Vol. **1755**, No. 1 (2021) p. 012027.
[6] Lee, T., Yasuda, and Adachi, C., Advanced Materials, vol. **27**, no. 12 (2015) pp. 2096–2100
[7] Mohammed, B. S., Sari, N. E. C., and Selma, M. K., Trans. on Electrical and Electronic Mat., vol. **16**, no. 3 (2015) pp. 124–129.
[8] Singh, T. J., Singh, S., Islam, S. M., Get, R., Mahala, P. and Singh, K. J., Optik, vol. **181** (2019) pp. 984–992.
[9] Fadavieslam, M., Optik, vol. **182** (2019) pp. 452–457.
[10] A. Udhiarto, L. M. Haryanto, B. Khoerun, and D. Hartanto, "Effect of anode and cathode workfunction on the operating voltage and luminance of a single emissive layer organic light emitting diode", in 2017 15th Int. Conf. on Quality in Research (QiR) : Int. Symp. on Electrical and Comp. Eng., (2017) pp. 65–68.