

COLOR-TUNABLE OF ORGANIC LIGHT EMITTING DIODE MEH-PPV: CNTS

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Organic light emitting diodes (OLEDs) were fabricated containing host molecule of conjugated polymer MEH-PPV poly (2-methoxy-5-(2'-ethylhexyloxy)-1,4-phenylenevinylene) which was dissolved in chloroform solvent and doped with guest molecules MWCNTs that were dissolved in DMF solvent with different volume ratios 0.05 and 0.15, nanocomposite system which acts as active layer in OLEDs device . The Poly(3,4-ethylenedioxythiophene):poly-(styrenesulfonate) "PEDOT:PSS" and Tris98-hydroxyquinoline aluminum "Alq₃" as hole transport layer HTL and electron transport layer ETL were used respectively, to enhanced the injection charge from the electrodes to the active layer. Spin coating method used for achieving facile and low cost OLED. The absorption spectrum for active layer examined by UV-Vis . It explained that the absorbance decreases with increasing volume ratio of CNTs : ; PL spectrum detected two curves quenching and recombination process results of doping with CNTs and AFM measurement shows increasing of roughness average from (0.168 to 0.668) nm with increasing the volume ratio of CNTs. Current – voltage " I-V " characteristics were determined for OLED, it has MEH-PPV:MWCNTs as active layer ,it is found that the threshold voltage for MEH-PPV diode is 2.5 V and decrease with increase CNT, while the current density increases due to tunneling majority and minority charge at interface between hole transport layer HTL , electron transport layer ETL and active emission layer EML. The specific nanocomposite thin film at 0.85:0.15 volume ratio of (MEH-PPV:MWCNTs) as active mission layer EML emitted tunable light color at specific voltage.

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1. Introduction

The electroluminescent of organic materials are consider as new category of compounds have a large interesting since they obey to the emission with extend spectral range, began from violet to the red light, several different material can be used as combined forms to obtain "white light" .The conjugated polymers are one of the organic category that have electroluminescence properties[1].Organic light-emitting diodes (OLEDs) have more attractive due to multiplications for rigid state lighting, with considerable advances being made in the last two decades [2,3]. The conjugated polymer is define as a hole transport material (HTM) [4] and it can be unique excited generator in optoelectronic material [5].Normally, conductive polymer MEH-PPV which defined as "poly (2-methoxy-5-(2'-ethylhexyloxy)-1,4-phenylenevinylene)" was used in organic diode ,sensor and organic light emitting diodes because of many perfect properties as ecological stability , easy control of conductivity and low-priced in manufacture of OLED. The conjugated polymer such as MEH-PPV have electronic and optical properties of semiconducting [6]. The organic semiconductors, oligomers, polymers and dendrimers, all of them are based on conjugated" π - electrons". The system of conjugated is represented by an alternation between single and double bonds. The π -electrons are more mobile than σ electrons.

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Thus, π -electrons transport from position to another one by hopping. The light absorption in organic solar cell and light emission in organic light emitting diode, the responsible of these process is π -electrons [7]. The researchers were attracted to the discovery of carbon nanotube CNTs and the probability of development carbon based nonmaterial [8]. Many application like electronic structure [9], biological systems [10], mechanical and transport properties. Whatever ideal one dimensional system depends on the long length and the small diameter of carbon nanotube lead to such large width to length ratio depends on the perfect properties of carbon nanotubes [11]. Composites of conducting polymers with carbon nanotubes CNTs [12–14] have attracted considerable attention in last few years because of their probability applications in photovoltaic cells and photodiodes [15], polymer light emitting diodes [16,17], sensors [18] optical limiting devices [19], super capacitors [20].

Despite many stressful attempts has been dedicated to the optimized condition of organic light emitting diode devices performance, carrier injection from the electrodes to the organic materials still restricting factor. To lower these barriers is to use low work function cathode materials for electron injection and high work function anode materials for hole injection [21]. The operating voltage and luminescence efficiency of the OLED devices were strongly depends on charge transport in the organic material and charge injection from the electrode to the organic material. To obtain ohmic interface between the organic material and the charge injection contact and to maximize the drift mobility of both electron and hole carrier, it should be lowering voltage [22].

The positive electrode of choice in OLEDs that has generally been used was indium tin oxide ITO due to large conductivity and high transparence properties. The chemically and physically properties is consider as weak properties of ITO, which reduced the performance of the electrode in organic light emitting diode with time [23,24]. Different hole injection layers have been united between ITO and organic material to support hole injection and match the energy level such as copper phthalocyanine (CuPc) [25], polymer such as polyethylene dioxythiophene: polystyrene sulfonate "PEDOT:PSS" [26], as Hole transfer layer (HTL) [27] and to improve electron injection layer used, Alq₃ as Electron transfer layer (ETL) [28]. Chemical structure of PEDOT:PSS and Alq₃ shown in Figure 1. In this work we demonstrated the conductive polymer and nanocomposite materials of MEH-PPV:MWCNT as OLED device to obtain tunable color and enhance the emission spectrum of the organic polymer.

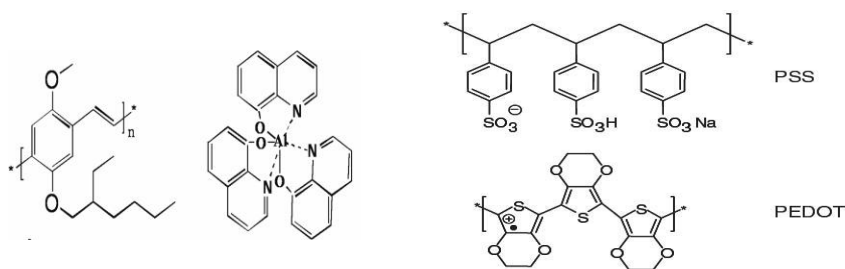


Fig. 1. Molecular structure of organic molecules. MEH:PPV polymer at the left, Alq₃ molecule in the middle and PEDOT:PSS molecule at the right

2. Experimental

2.1. Preparation of Multi Wall Carbon Nano Tube (MWNT) and MEH-PPV solutions:

The powder of MWCNT was first weighted of 10 mg and dissolved in 10 ml of (N,N-dimethylformamide DMF) to obtain the concentration 1 mg/ml, the solution was put on the magnetic stirrer for 6 hour and on sonication for 8 hours to obtain homogeneous solution. This solution is diluted to the concentration 0.01 mg/ml of MWCNT.

The concentration of polymer MEH-PPV, Sigma-Aldrich, $M_n = (40.000-70.000)$ g/mol, was prepared by weighting 10 mg of the polymer and dissolved in 10 ml of chloroform (CHCl₃) solvent, to obtain the concentration 1 mg/ml. Next, the MEH-PPV solution was doped with

MWCNT solution and it was sonicated to enable the MWCNT mixed well with the MEH-PPV solution with volume ratio 0.05:0.95 and 0.15:0.85. PEDOT:PSS is a polymer incorporated both of conducting polycation "PEDOT" and polyanion "PSS", The advantage from this incorporated are high conductivity that larger than 200 S/cm, high transmission in visible range and water soluble property with ratio (3.0-4.0)% of H_2O [27]. Tris (8-hydroxyquinoline) aluminum (Alq_3), possessing superior film stability and thermal endurance [28], were purchased from Sigma-Aldrich, Alq_3 was prepared by weighting 10 mg and dissolved in 10 ml of Methanol solvent (CH_3OH) to obtain 1 mg/ml concentration. Spin coating method is used to fabricate the OLED device ITO/PEDOT:PSS(HTL)/MEH-PPV:MWCNT(EML)/ Alq_3 (ETL)/Al, began with the ITO –coated glass substrate (200nm, $10\Omega/\square$), the surface of the ITO was cleaned by sequence steps in acetone, ethanol, methanol and de-ionized water in ultrasonic bath for 10 minute each. PEDOT:PSS was coated by using spin coating method at 4000 rpm, 1 minute and dried at 150 °C. The nanocomposite solution MEH-PPV:MWCNTs was deposited at 1500 rpm, 1 minute and then dried at 100°C to remove any residual of solvent and deposited the Alq_3 solution at 1500 rpm, 1 minute and then dried at 100°C. Finally Aluminum metal was deposited by thermal evaporation technique, the structure of OLED device is shown in Figure 2a. Figure 2b illustrate energy diagram of these layers. Spin coating system (ACE 200) was used in this study, surface of the thin films was carried out using Atomic Force Microscope(SPM, Model AA3000), tip NSC35/AIBS from Angstrom Advanced Inc (USA), UV-Vis measurements SP – 8001 spectrophotometer over (Meterrech) with wavelength range from 190 to 1100 nm, PL measurement by Agilent Tech. Fluorescence spectrophotometer. The EL spectrum was measured using the SPECTROFLUOROMETER DETECTOR (RF-551) ver 2.3A programmable & scanning connected with I-V forward bias circuit measurement.

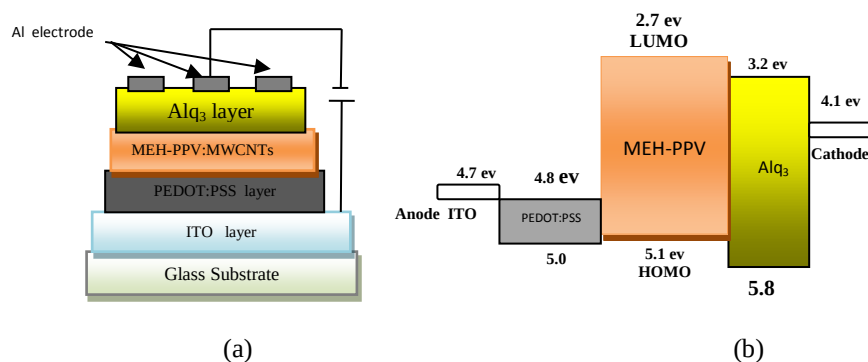


Fig. 2. (a) Scheme diagram of the fabrication OLED device and (b) Energy diagram of the doped OLED.

3. Results and discussion

3.1. Atomic Force Microscope analysis

The morphology investigated of dispersion CNTs in organic polymer as nanocomposite material accomplished by the analysis of surface morphology, using AFM images obtained in the tapping mode with approximately area $2.05 \times 2.05 \mu m^2$, as shown in Figure 3. For the pure MEH-PPV and MEH-PPV:MWCNT thin films, CNTs with different volume ratios 0.05, 0.15 respectively.

(2D) and (3D) image of surface morphologies as shown in Figure 3 illustrate that more grain polymer dispersion in homogenous way, this is due to the well distribution by choice speed 1500 rpm of spin coating and suitable evaporate solvent. Sequentially, the dispersion of MWCNTs increase with increasing the volume ratio of CNTs in nanocomposite thin films as shown in Fig. 3c -(2D) image at volume ratio 0.15 that distributed at the surface more than 0.05 ratio as shown in Fig. 3b-(2D), and this is make 0.15 ratio more suitable in OLED device,

MWCNT bundles can be observed as a shadow bundles, which shows that a polymer layer has wrapped around the nanotube indicated by blue arrows. This is due to the spider network of interconnected tube. That results in efficient charge transport between the interconnected conductive paths of MWCNTs and high optical transparency level due to evolution structure [29]. The average roughness of "MEH-PPV"(pure) and "MEH-PPV:MWCNT" thin films with different volume ratios 0.05,0.15 are 0.168,0.221 and 0.668 nm, respectively. The increase of the surface roughness is due to the formation of aggregation of CNTs, when increase the volume ratio of CNTs. The AFM study display as mighty dependence on the nature of MWCNTs that doped in the matrix of the polymer.

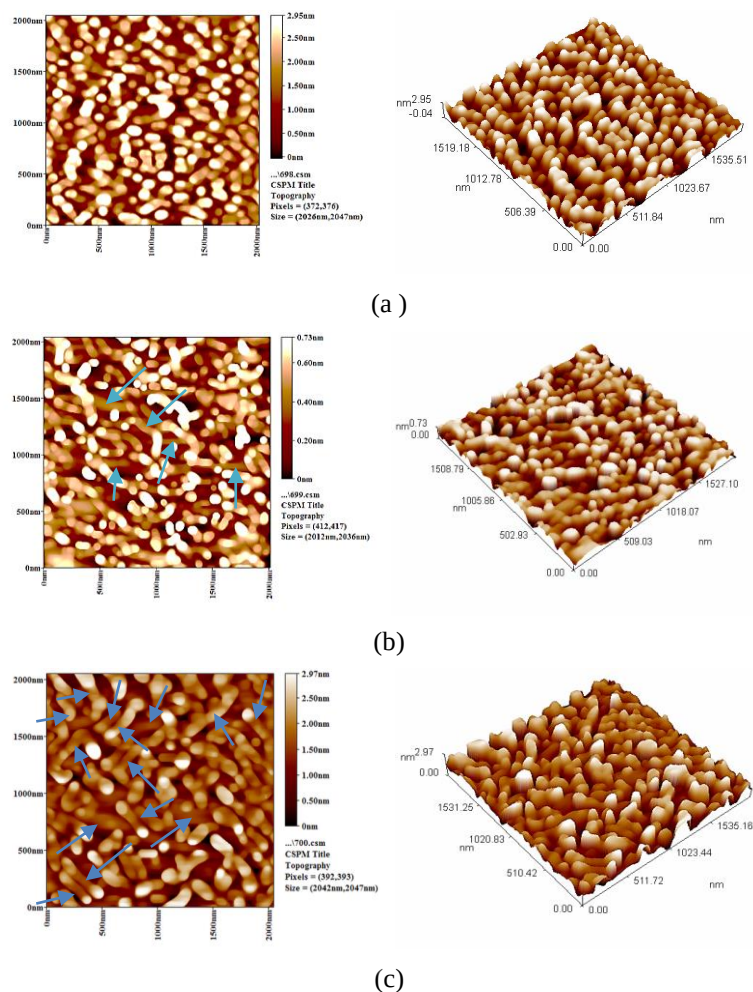


Fig. 3: AFM images (tapping mode) 2D to the left side and 3D to the right side of "MEH-PPV" and "MEH-PPV:MWCNT" nanocomposite thin films, (a) "MEH-PPV" (pure), (b) "MEH-PPV:MWCNTs" (0.05) and (c) "MEH-PPV:MWCNTs" (0.15)

3.2. optical characterization

Absorption spectrum of "MEH-PPV" and "MEH-PPV:MWCNT" nanocomposite thin films with different volume ratios 0.05 and 0.15 CNT were carried out as shown in Fig.4. All spectra show two peaks at 338 nm (B-Band) and 510 nm (Q-Band). Spectra of all volume ratios of MWCNTs detected a linear dependence between spectra of both molecules MEH-PPV and MWCNT. This is indicating that CNT is incorporate into MEH-PPV matrix without modifying the chemical structure of polymer. Therefore, it is likely that molecular orbital hybridization between the HOMO of MEH-PPV and LUMO of MWCNTs is achieved. Upon addition of MWCNTs, the

absorption peak at 338nm and 510 nm gradually decrease. It may be due to the reduction of optical volume of MEH-PPV polymer, and this result is in agreement with ref.[30,31].

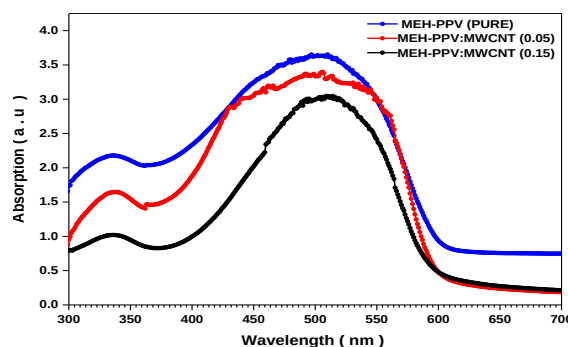


Fig. 4: Absorption spectra of MEH-PPV and MEH-PPV:MWCNT thin films with different volume ratios.

The PL quenching takes place by charge separation growing from polymer to the carbon nanotube, the MWCNT were represented as exciton dissociated centers, to get a great efficient charge, and provide pathway for electrons then increase the effective mobility of the carrier in active organic layer, produced by percolation network of CNT, thus the high electric field causes splitting up the exciton and the charge transfer then follows by transport of electron through the nanotube length[32,33].

The photoluminescence spectra of organic polymer MEH-PPV and "MEH-PPV:MWCNT" nanocomposite were between (530-750) nm at 300 nm excitation wavelength, as shown in Fig.5. The decrease in photoluminescence at 0.05 volume ratio of CNT is due to the excited fluorophore in polymer backbone which is quenched by electronic interaction at the interface between "MEH-PPV" and "MWCNTs", and the quenching PL emission at this ratio is according to the fast charge separating and this suitable in organic photo voltaic (OPV) application, while when the PL intensity increase this is due to slow charge recombination and this is useful in OLED application, this is result in agreement with ref.[30,39].

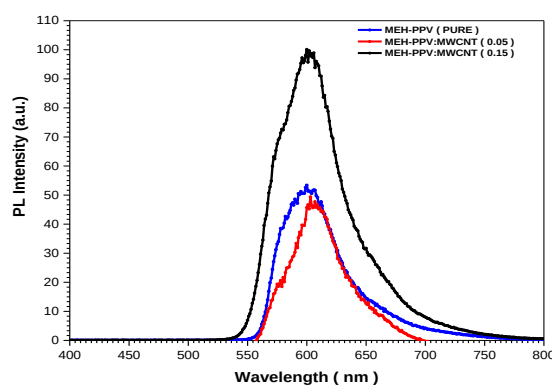


Fig. 5. Photoluminescence spectra of "MEH-PPV" and "MEH-PPV:MWCNT" composite with different volume ratios.

3.3. Electrical characterization

I-V characteristics of MEH-PPV and MEH-PPV:MWCNT nanocomposite were measured using the schematic sandwich geometry, as shown in Fig.2a. From I-V characteristics, we can observe when the volume ratio of CNT increase from 0 to 0.15, the threshold voltage reduces considerable from 2.5 to 1 volt, also the current density increase compared to the pure case from 0.0015 to 1347 $\mu\text{A}/\text{mm}^2$, as shown in Fig.6 b. The effect of doping CNT with organic active layer explained by the J-V characterizing for both pure polymer and doped with MWCNT

nanocomposite. The current increase between 10^3 and 10^5 times with modifying by CNT of 0.05 and 0.15 volume ratio respectively, especially at low voltage. The relationship of J-V undergoes an Ohmic conduction, which is due to trap charge limited (TCL) at low voltage and space charge limited (SCL) current at high voltage for nanocomposite case, in pure polymer case, the current is injection limited by contact impact, and it undergoes power law with a large value of l in the power of law eq(1) and this is due to deep trap at the interface. The reduction of empty traps results quickly increase the current. In the high applied bias, the traps are filled and called (traps filled-limit, TFL) [34], in cases of typical SCL and TCL currents at very low electric fields, the I-V-behavior typically exhibits first linear ohmic ($\alpha = 1$) or super-ohmic ($1 < \alpha < 1.5$) conduction in power of law ($I \propto V^\alpha$) [35,36], the more appropriate notation of this relation is:

$$J_{TLC} \propto \mu \frac{V^{l+1}}{d^{2l+1}} \quad (1)$$

where $l = T_t/T$ with being the absolute temperature and T_t being the characterizes temperature of trap distribution, μ charge mobility, V is applied voltage and d is the active layer thickness[33].

We assume the formation of HTL as PEDOT:PSS and ETL as Alq₃ during anode and cathode electrode, lowering the barrier or creating a tunneling path for majority and minority carrier, the injection barrier height of holes is small, around 0.1 eV, holes can be injection in to active layer at thresholds voltage 2.5, 0.25 and 1 volt at the volume ratio 0, 0.05 and 0.15 of CNT, consider as dominate charge carrier, while the barrier height of electron is 0.5 eV, which is much higher than the holes. Therefore the device with different volume ratio of CNT have to be biased at higher voltage in order to accomplish the injection of minority carrier as well as the recombination of opposite charge carrier. The low threshold voltage and a large current density and SCLC conductance in "MEH-PPV: MWCNT" nanocomposite are explained by reduced the barrier height for hole carrier, the effect of CNT act as exciton dissociated centers and the increase of current density. according to the Fowler – Nordheim tunneling theory [37]. While in organic polymer MEH-PPV OLED, the current is limited by contact impact can be explained the higher threshold voltage for hole injection at the interface between PEDOT:PSS and MEH-PPV.

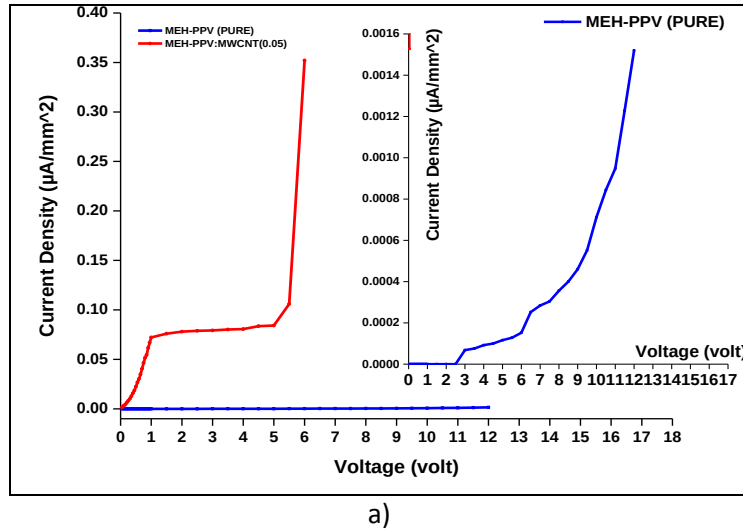


Fig. 6 a). (J-V) characteristics J-V characterize of pure organic polymer MEH-PPV and blend MEH-PPV:MWCNT(0.05) nanocomposite OLEDs

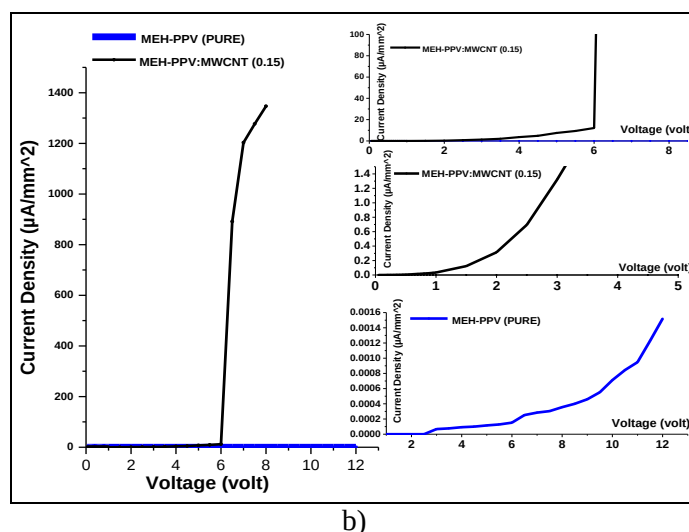


Fig. 6 b) J-V characterize of MEH-PPV and blend MEH-PPV:MWCNT(0.15) nanocomposite OLEDs devices.

3.4. Electroluminescence spectrum

Fig. 7 shows the EL spectra, which studies the effect of doping CNT into organic polymer "MEH-PPV" as active layer in geometric structure of bottom organic light emitting diode (OLED). The emitting light consists of both orange and red light color based on "MEH-PPV" and nanocomposite "MEH-PPV:MWCNTs" OLEDs, where the orange light color dominates red light color in both "MEH-PPV"(pure) and "MEH-PPV:MWCNT" nanocomposite OLED with volume ratio 0.05 of CNT as shown in Fig. 7 a and b, it is evident that EL spectra of orange and red light emitting color are independent on the applied voltage, we selected low and high voltage according to the J-V curve to compare between MEH-PPV and MEH-PPV:MWCNTs OLEDs device, with different volume ratios of MWCNT (0.05,0.15), the intensity level of the emission light at the volume ratio of 0.05 CNT is shown in Table 1, and this agrees with PL spectrum, where the CNT plays as a quencher of emission intensity, while with the volume ratio 0.15 of CNT as shown in Fig. 7 c, and in low voltage, the red light color dominates and at specified and larger voltage like 6.5 volt, the orange light color is dominant as shown in Fig. 7d and e. The tune color and the red shift in all EL spectrum, in addition to the narrow band in all blend curve, is due to microcavity effect. Microcavity effect is changed with the length of light path, refractive index, and detection angle, and can be used to tune the light color of OLEDs besides its efficiency [38].

Table 1. comparable voltage between pure MEH-PPV and blending MEH-PPV:MWCNT nanocomposite OLEDs according to light color position and dominate color

Voltage bias	Volume ratio of CNT	(Position, Intensity) of orange light	(Position, Intensity) of red light	Dominant light
3 volt	0	(580,197)	(620,43)	orange
	0.05	(586,146)	(620,64)	orange
6 volt	0	(580,203)	(620,46)	orange
	0.05	(586,198)	(623,66)	Orange
3 volt	0	(580,197)	(620,43)	orange
	0.15	(586,288)	(626,488)	red
6.5 volt	0	(584,270)	(624,24)	orange
	0.15	(585,329)	(625,220)	orange
8 volt	0	(581,208)	(623,52)	orange
	0.15	(585,431)	(626,223)	orange

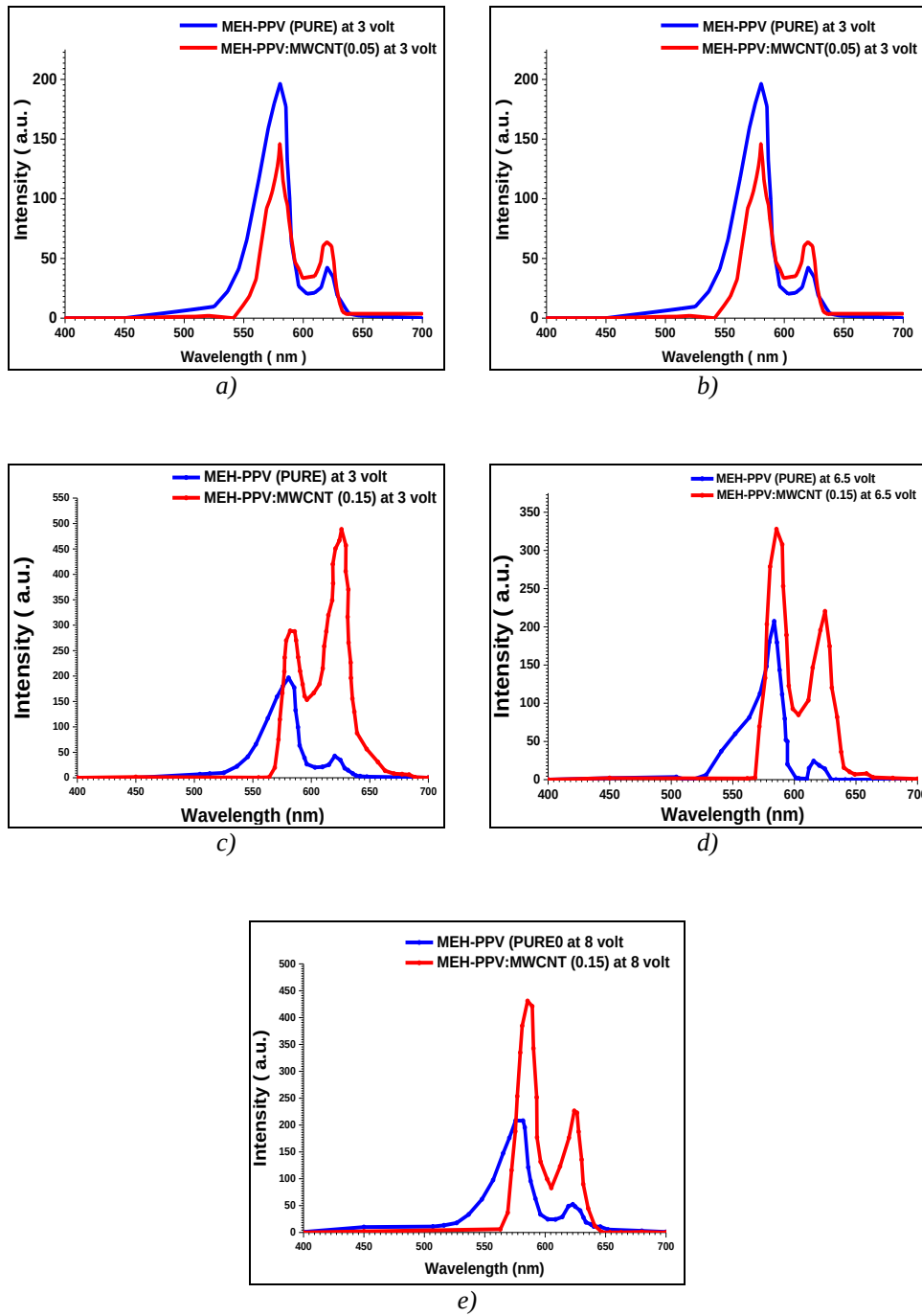


Fig. 7. EL spectra of organic color-tunable OLEDs doped with CNT for (a) MEH-PPV:MWCNT nanocomposite with volume ratio (0 and 0.05CNT) at 3 volt ;(b) at 6 volt; (c) volume ratio (0.15CNT) at 3 volt(d) at 6.5 volt(e) at 8 volt

4. Conclusions

Composites based on MEH-PPV:MWCNTs (0,0.05,0.15) volume ratios have been prepared successfully using spin coating technique. PL spectrum was quenched and recombination behavior and decrease in UV-Vis spectrum with increase in volume ratio of MWCNTs. There is reduce in voltage threshold and increase of current density , special in 0.15 volume ratio, at this ratio obtained tunable OLED from red to orange light color. All of these attributes are desirable with respect to the design OLEDs system as active layer.

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