Comparison of different organic solvents used in the luminescent material for OLED devices

Comparação de diferentes solventes orgânicos usados em material luminescente para dispositivos OLEDs

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ABSTRACT

In this work, organic light-emitting diode (OLED) devices were mounted using the structure: glass (as substrate)/indium tin oxide (ITO) (as anode)/poly(3,4-ethylenedioxythiophene) doped with poly(styrenesulfonate) (PEDOT:PSS) (as hole transport layer)/poly[9,9-dioctifluorene-alt-bis-tienilene(benzotiadiazole)] (PFTB) (as luminescent material)/aluminum-doped zinc oxide (AZO) (as electron transport layer)/aluminum (as cathode). The PFTB was synthetized at laboratory and diluted in different organic solvents as chloroform and trichlorobenzene. The I-V curves of OLED devices showed that the trichlorobenzene used to dillute the PFTB improved the performance for OLED devices promoting the highest electrical current of ≈50 mA and the lowest range of thresold voltage from ≈2.5 to 5 volts, while the device OLEDs mounted with PFTB dilutted in chloroform presented maximum electrical current of ≈23 mA and range of thresold voltage from ≈5 to 8 volts. A hypothesis that explain these results can be attributed to the boiling point of the organic solvent of trichlorobenzene (≈214.4ºC) to be higher than the one of the chloroform (≈61.1ºC), favoring better rearrangement of the polymer chains of PFTB and interfaces between thin films PFTB/PEDOT:PSS and PFTB/AZO improving the injection of charges (holes and electrons) inside the OLEDs devices.

KEYWORDS: Organic light-emitting diode, Poly[9,9-dioctifluorene-alt-bis-tienilene(benzotiadiazole)], Chloroform, Trichlorobenzene, Electrical current, Threshold voltage.

RESUMO

Neste trabalho, dispositivos de diodo orgânico emissor de luz (OLEDs) foram montados utilizando a estrutura: vidro (como substrato)/óxido de índio e estanho (ITO) (como anodo)/poli(3,4-etilenodioxitiofeno) dopado com poli(estirenosulfonado) (PEDOT:PSS) (como camada transportadora de lacunas)/poli[9,9-dioctifluoreno-alt-bis-tienileno(benzotiadiazol)] (PFTB) (como material luminescente)/óxido de zinco dopado com alumínio (AZO) (como camada transportadora de elétrons)/alumínio (como catodo). O PFTB foi sintetizado em laboratório e diluído em diferentes solventes orgânicos como clorofórmio e triclorobenzeno. As curvas I-V dos dispositivos OLEDs mostraram que o triclorobenzeno utilizado para diluir o PFTB melhorou o desempenho dos dispositivos OLEDs promovendo mais alta corrente elétrica de ≈50 mA e mais baixa faixa de tensão de limiar de 2,5 até 5 volts, enquanto os dispositivos OLEDs montados com PFTB diluído em clorofórmio apresentaram corrente elétrica máxima de ≈23 mA e faixa da tensão de limiar de ≈5 a 8 volts. Uma hipótese que explica esses resultados pode ser atribuída ao ponto de ebulição do solvente orgânico triclorobenzeno (≈214,4ºC) ser mais elevado do que o do clorofórmio (≈61,1ºC), favorecendo melhor acomodação das cadeias poliméricas do PFTB e interfaces entre os filmes finos de PFTB/PEDOT:PSS e PFTB/AZO melhorando a injeção de cargas (lacunas e elétrons) no interior dos dispositivos OLEDs.

PALAVRAS-CHAVE: Diodo orgânico emissor de luz, Poli[9,9-dioctifluoreno-alt-bis-tienileno (benzotiadiazol)], Clorofórmio, Triclorobenzeno, Corrente elétrica, Tensão de limiar.

INTRODUCTION

Organic light-emitting diode (OLED) is a solid-state semiconductor device composed by superposition of different thin films to emission of light in different colors[1,2]. Basically, the light-emitting layer is composed by organic material located between two inorganic thin films, the anode (transparent and semiconductor material deposited on transparent substrate) and the cathode (metallic conductor)[3,4]. Researches have been carried out with the objective to find materials that present efficient performance as low threshold voltage and high luminance, but the operating voltage is still elevated if compared with common inorganic light-emitting diode (LED) devices[5,6]. Another problem is related to the prolonged use in OLED technology, because the organic materials have short lifetime, caused principally by water and oxygen in the ambient air[7], leading to dark spots or bur-in deffects[8,9].

The OLED devices are assembled using the concept of multilayers and they present electrical characteristics similar to the ones of the diode devices[10,11]. When a voltage is apllied in the electrodes, the electron charges flow through the cathode, while hole charges flow through the anode, and inside the luminescent organic material occurs the recombination of both charges generating energy in the photons form[12,13]. These devices can be divided in two different segments: polychromatic light, used as displays in smartwatches, smartphones and TVs[14,15]; and monochromatic light, used in ilumination or sinalization[16,17] (as foccus of this work).

In the comparison of displays between OLEDs and other devices as liquid crystal displays (LCDs), plasma or LEDs, the organic devices have presented some advantages, such as[18-20]: better colours and constrast; assembly on flexible substrates; view angle of 180º; green technology (no heavy metals); fast response time; easy to assembly; and no infrared and ultraviolet rays' emission. For this reason, several companies of consumer electronics and lighting products have showed great interest to develop these type of devices.

Structure of OLED device

Assembly of a basic OLED device uses a organic material as luminescent layer on the anode and cathode on top, but this simple structure offers insignificant luminance and elevated threshold voltage. Then, in the literature there are other structures of devices involving more than three multilayers that offer better performance to the OLED devices[21-23]. For example, structures containing five layers have been studied and presented good results as electrical and optical performance using the materials[24-26]:

- Substrate: rigid material used to support the multilayers of the OLED device. In this case, glass is very used, because it has good properties as mechanical resistance and transparency and offers good barrier to degradation of oxygen and moisture from ambient;
- Anode: transparent conductive oxide (or TCO) thin film. This inorganic material is generally commercial and very used as anode. It is deposited on the glass substrates offering different optical and electrical properties, such as: sheet resistances, roughnesses, transmittances and thicknesses. The TCO promotes the injection of holes inside OLED device;
- Hole transport layer (HTL): layer formed by organic material with semiconductor and transparent properties. Generally, this material is commercialized and dilluted in water ready to be used and deposited on the TCO surface. This material contributes with good transport of holes to be injected into the luminescent material;
- Luminescent material: this thin film has insulating electrical property and deposited on the HTL surface. Inside this material, there are the recombination charges (hole-electron pair) to light emission, when the OLED device is polarized. Luminescent material can be evaporated or diluted in organic solvents (method very used by some research laboratories) to thin film's formation;
- Electron transport layer (ETL): layer formed by organic material with semiconductor property, it is deposited on the luminescent material surface. This material offers good transport of electrons to be injected into the luminescent material;
- Cathode: this mettalic layer is deposited on top. The metal promotes the injection of electrons inside the OLED device.

The last procedure is the OLED device encapsulation to maintain the intrinsic chemical properties of all organic layers used and to avoid the degradation caused by moisture, oxygen and ultraviolet rays from ambient[27]. In the OLED device encapsulated, it is possible to obtain several polarizations with stable performance (maintaining the same values for: electrical current, luminance and threshold voltage).

The objective of this work was to know the influence of different organic solvents used to dilute the PFTB material used as luminescent material developed at laboratory to assembly of OLEDs devices.

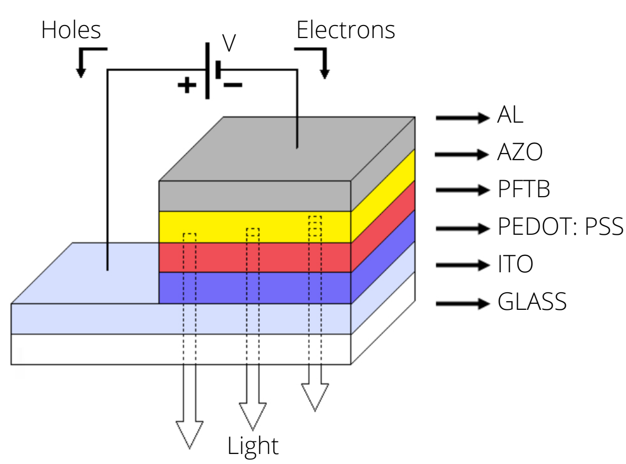
MATERIALS AND METHODS

In the assembly of the OLED devices, the complete multi-layer structure was used with materials:

- Indium tin oxide (ITO)/glass: commercial ITO used as anode electrode and deposited on glass substrates supplied by Diamond Coatings Limited with sheet resistance of 15 Ω/▯ and transmittance above 70% at the range of visible light;
- Poly(3,4-ethylenedioxythiophene) doped with poly(styrenesulfonate) (PEDOT:PSS) (used as HTL): supplied by Sigma-Aldrich Corporation (reference: 483095-250G). It is composed by PEDOT with 0.5 wt. %, PSS with 0.8 wt. % and 1.3 wt. % dispersion in H2O and it was used as received;
- Poly[9,9-dioctifluorene-alt-bis-tienilene(benzotiadiazole)] (PFTB) (used as luminescent layer): this material was synthesized in Eric Tsuneki Yoshiura Ono's work. Previously, PFTB was diluted in chloroform and 1,2,4-trichlorobenzene, both supplied by Tedia Company and used with concentration of 10 mg/mL;
- AZO (aluminium-doped zinc nanoparticle ink, used as ETL): obtained from Sigma-Aldrich Corporation (reference: 793388-5mL). This material is available with 2.5 wt. % crystalline Al-doped ZnO, 98 wt. % ZnO and 2 wt. % Al2O3 in 2-propanol alcohol and it was used as received;
- Al (aluminum used as cathode electrode) with purity of 99.999%. It was thermally evaporated on top.

Partial method of assembly was mounted into glove box under nitrogen atmosphere and relative humidity below 20%. In this case, the deposition of PEDOT:PSS, PFTB and AZO to the thin layers formation used the spin-coating technique with 3,000 rpm by 60 seconds, and each thin film was dried at 55°C by 5 minutes. Four OLEDs were mounted at the same time on each sample with geometry of 2.5 × 2.5 cm and active area of 3 × 3 mm.

The OLED devices were analyzed without encapsulation and polarized using source power Keithley, model 2400, and LabTracer software, version 2.0, to obtain the I-V curves. Figure 1 shows the complete structure with five layers used in the assembly of OLED devices.

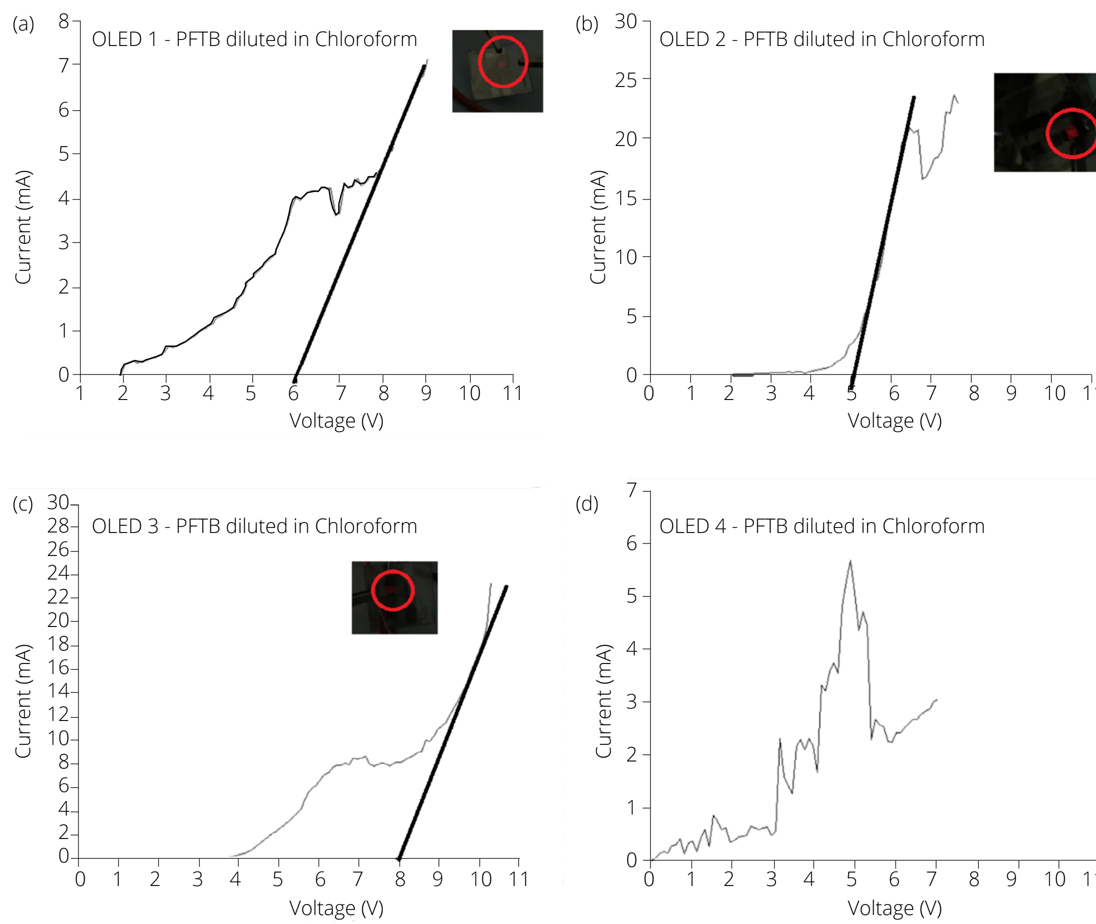


Al: aluminum; AZO: aluminum-doped zinc oxide ; PFTB: poly[9,9-dioctifluorene-alt-bis-tienilene(benzotiadiazole)]; PEDOT:PSS: poly(3,4-ethylenedioxythiophene) doped with poly(styrenesulfonate); ITO: indium tin oxide.

Figure 1: Structure with five layers used in the assembly of organic light-emitting diode (OLED) devices.

RESULTS

The comparison of I-V curves between OLEDs mounted with PFTB emissive polymer diluted in chloroform and trichlorobenzene showed a significant difference in the performance of devices. In this case, all four OLEDs obtained with chloroform revealed lower current level than the devices mounted with PFTB using trichlorobenzene as organic solvent. The I-V curves also revealed that the OLEDs with PFTB diluted in trichlorobenzene presented most reproducible results with aspect of diode curve. This fact was not completely observed in the electrical behavior of I-V curves for OLED devices mounted with PFTB diluted in chloroform. Another observation is related by tangent lines to the I-V curves for OLEDs mounted with PFTB diluted in trichlorobenzene, that revealed an easy method to obtain the threshold voltages[28]. In this case, lower threshold voltages can be observed for OLEDs devices mounted with PFTB diluted in trichlorobenzene presenting narrow values from ≈2.5 to 5 volts, while the OLED devices mounted with chloroform presented broad range from ≈5 to 8 volts. Figures 2a to 2d show the I-V curves for devices mounted with PFTB diluted in chloroform with respectively OLED turned-on (red circle).

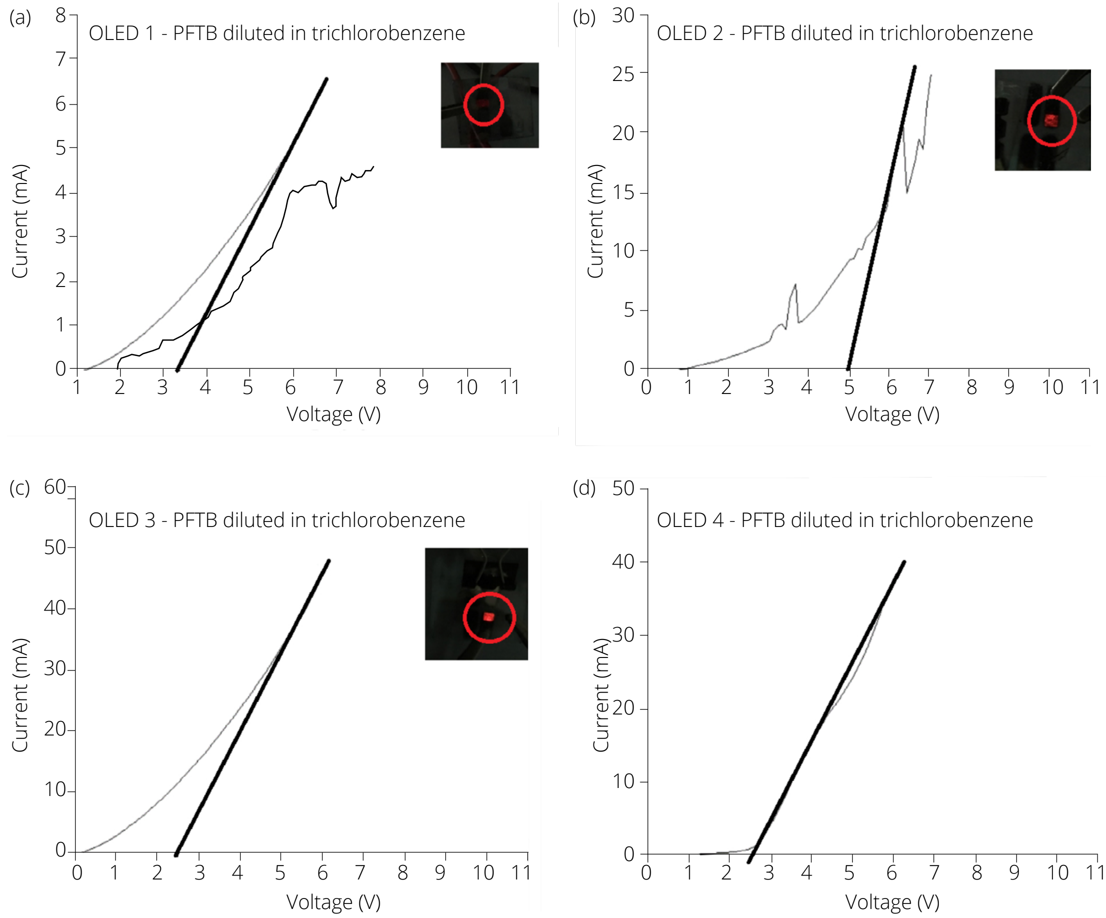


OLED: organic light-emitting diode; PFTB: poly[9,9-dioctifluorene-alt-bis-tienilene(benzotiadiazole)].

Figure 2: (a) OLED 1: PFTB diluted in chloroform; (b) OLED 2: PFTB diluted in chloroform; (c) OLED 3: PFTB diluted in chloroform; (d) OLED 4: PFTB diluted in chloroform.

Due to the bad performance presented by OLED 4 in Fig. 2D, the threshold voltage was not obtained. Moreover, this device presented no luminance. The highest electrical current achieved for OLEDs mounted with PFTB diluted in chloroform was ≈23 mA, while the OLED devices mounted with trichlorobenzene was ≈50 mA. Figures 3a to 3d show the I-V curves for devices mounted with PFTB diluted in trichlorobenzene with OLED turned-on (red circle). The results showed that the OLED device of Fig. 3D presented no luminance, although it presented a similar diode curve (no oscilations). The behavior of I-V curves for OLED devices mounted with PFTB diluted in trichlorobenzene presented similarity compared with common diode curves showing the influence caused by trichlorobenzene in the PFTB as luminescent material. The hypothesis to obtain these results can be attributed to the boiling point of the organic solvent trichlorobenzene (≈214.4°C), that is higher than the one of the chloroform (≈61.1°C), favoring a better rearrangement of the polymer chains alignment and also the interface of layers PFTB-PEDOT:PSS and PFTB-AZO, facilitating the charge carriers mobility and formation of electron-holes recombination inside the luminescent material.

The change of the organic solvent is most evidenced in the threshold voltage of OLED devices, showing better performance for PFTB diluted in trichlorobenzene.



OLED: organic light-emitting diode; PFTB: poly[9,9-dioctifluorene-alt-bis-tienilene(benzotiadiazole)].

Figure 3: (a) OLED 1: PFTB diluted in trichlorobenzene; (b) OLED 2: PFTB diluted in trichlorobenzene; (b) OLED 3: PFTB diluted in trichlorobenzene; (d) OLED 4: PFTB diluted in trichlorobenzene.

CONCLUSION

The study involving the PFTB as luminescent material diluted in different organic solvents such as chloroform and trichlorobenzene revealed better performance of the I-V curves for OLEDs devices using the trichlorobenzene, because it provides the highest electrical current and the lowest threshold voltage.

A hypothesis to obtain better results for OLEDs mounted with PFTB diluted with trichlorobenzene can be related to the boiling point of each organic solvent. The trichlorobenzene's boiling point is ≈214.4°C, that is much higher than the one of the chloroform, ≈61.1°C, and this fact causes better rearrangement of the polymer chains, faciliting the better locomotion of the charge carriers inside the OLED devices, increasing the electrical current and decreasing the threshold voltage considerable.

AUTHORS' CONTRIBUTION

Conceptualization: Santos ER, Ono ETY e Shu Hui W; **Data Curation:** Santos ER, Ono ETY e Shu Hui W; **Formal Analysis:** Santos ER, Ono ETY; **Funding Acquisition:** Santos ER, Ono ETY, Onmori RK e Shu Hui W; **Methodology:** Santos ER, Ono ETY, Onmori RK e Wang; **Resources:** Onmori RK e Shu Hui W; **Software:** Shu Hui W; **Supervision:** Santos ER, Ono ETY, Onmori RK e Shu Hui W; **Validation:** Santos ER e Ono ETY; **Visualization:** Santos ER e Ono ETY; **Writing – Original:** Ono ETY e Shu Hui W; **Draft Preparation:** Santos ER, e Ono ETY; **Writing – Review & Editing:** Santos ER.

DATA AVAILABILITY STATEMENT

Data are available in a data repository.

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