Augmented behaviours of OLEDs as ETL based on PPV and PEDOT:PSS hybrid organic-inorganic nanocomposites: A review

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ABSTRACT

This work presents recent significant advancements in polymer-based hybrid nanocomposites, as well as a deeper understanding of the fundamental principles that regulate their optical, electrical, and electrochemical properties. Combining nanoparticles with a conjugate polymer matrix yields a polymer hybrid nanocomposite, which is used as an active electron transporting layer between the structures of optoelectronic devices like organic light emitting diodes (OLEDs). The materials PPV (Poly p-phenylene vinylene) and PEDOT:PSS (poly(3,4-ethylenedioxythiophene):polystyrene sulfonate) with various inorganic fillers have excellent electrical, electrochemical, and optical properties, making them appealing for usage as electron transportation layers in OLEDs (ETL). The current communication gives a quick overview of OLEDs and how they are made using with various hybrid polymeric nanocomposites. The comparative data for optical, chemical and electrical properties of PPV and PEDOT:PSS with various inorganic fillers reflects the better performance of OLEDs as electron transporting layer material.

Keywords

Hybrid nanocomposites, Multi-layered OLEDs, Poly p-phenylene vinylene, Poly(3,4-ethylenedioxythiophene:poly (styrene sulfonate), Optical-electrical properties.

1. Introduction

Polymer nanocomposites (PNCs) are one of the most prominent areas for current study and development in nanotechnology, and the investigation field encompasses a wide range of topics. Nanoelectronics, polymeric bionanomaterials, reinforced PNCs, nanocomposite-based drug delivery devices, and so on are examples. The term "nanocomposite" refers to composite or hybrid materials in which one component's size is in the nanometre range (1 nm=10-9 m). [1-3]. Polymer nanocomposites are a type of material with unique physicochemical features that are impossible to achieve with individual components functioning alone. Because of their great potential for a wide range of applications in environmental remediation and treating various environmental challenges, these nanocomposites have recently gained a lot of research interest. Organic (small molecule/polymer) light-emitting diodes (OLEDs) have recently made significant advances in full-color flat panel displays and other applications. However, OLEDs are currently dealing with high manufacturing costs, which severely limit the size of OLED panels, as well as life duration, particularly colour differential ageing. We believe solution-processing would be an appealing option to be more cost-effective for producing OLEDs due to its simplicity and low equipment costs [2-6]. Conjugated polymers have a number of features that make them appealing as emissive materials in organic light-emitting diode (OLED) devices. Low operating voltage, low fabrication cost, ease of processing and manufacturing, flexibility, ability to manufacture large-area devices with good solubility in common organic solvents, and photothermal stability are some of these advantages [7-10]. Several techniques can be used to create OLEDs with various colours, including: building bilayers in a tandem diode structure, using a single polymer with multiple functional groups, using quantum dots, blending of conjugated polymers, and mixing polymers with nanostructured materials. As previously stated, in addition to expense, OLEDs face the issue of colour disparity ageing. Small molecules and polymers are both examples of organic emitting materials. OLEDs not only emit a wide range of colours, but they also age at variable rates. Emissive colloidal quantum dots (QDs) are well-known for their excellent colour purity, differential stability, and tunability, as well as their inherent robustness. Due to its applications in flexible electronics, light-emitting displays [11], and photovoltaics [12-13], hybrid nanocomposites based on quantum dots (QDs) and conjugated polymers have recently gained a lot of attention. The employment of organic nanoparticles in organic devices causes them to fail even at room temperature, therefore the strong interfacial contacts between the polymer and inorganic fillers are to blame for the improved stability compared to the inorganic phase into the polymer matrix [14]. However, due to the disadvantages mentioned above, the durability of organic devices is still inferior to that of conventional semiconductors [15-17]. Because of their outstanding processability and high charge carrier mobility combined with efficient electroluminescence, PEDOT:PSS (poly(3,4-ethylenedioxythiophene):polystyrene sulfonate), polyfluorene (PF) and polyphenylene vinylene (PPV) derivatives have emerged as an interesting class of conjugated polymers for display applications [18,19]. OLEDs based on hybrids of poly(9,9-di-n-octylfluorenyl-2,7-diyl) (PFO), which serves as a donor material and has a high band gap, and poly(2-methoxy-5-(2-ethoxy)-1,4-phenylene-vinylene) (MEH-PPV), which acts as an acceptor material, have sparked interest in the scientific community.

In the current study reflects the enhancement of OLED performance based by incorporation of various inorganic filler in bare conducting conjugated polymers. The optical and electrical of PPV (Poly p-phenylene vinylene) and PEDOT:PSS (poly(3,4-

ethylenedioxythiophene):polystyrene sulfonate) with various inorganic fillers in terms of OLED application are also showed in comparative manner.

2. Employment of different Polymers in OLEDs

Polymer organic light emitting diodes (OLEDs) are highly efficient and only consume a small percentage of the light they generate [20-22]. By choosing the right combination of polymer and device changes, device efficiency can be increased. While devices can be enhanced by adding a thin polymeric layer between the hole transport layer and LEP, the light emitting polymer (LEP) can be tweaked to boost its photoluminescence and quantum yield (PLQY). To improve device performance, two types of non-conjugated polymer OLEDs, the pendant type and the host-guest blend type, have been created. Functional units are connected to the polymer's non-conjugated backbone in the former, whereas functional guests are doped into the host polymer in the latter. Polyvinylcarbazole (PVCz) is one of the most common non-conjugated polymers, and its optical and electrical properties were thoroughly investigated during the early stages of OLED research [23], when PVCz was widely utilised as a host material. Because the needed functions are inserted into polymer chains by co-polymerization technology, the conjugated polymer system works without a complex multilayer device construction. Functional backbone units, electron affinity units, hole affinity units, and emissive units are all components of a conjugated polymer. One of the most essential aspects is the colour of the emission, which is often adjusted by adding an emissive unit to the polymer chains. Backbone units are typically chosen from structures with a broad band gap.

"Intrinsically conducting polymer" (ICP) or "synthetic metal" is an organic polymer with the electrical, electronic, magnetic, and optical properties of a metal/metal oxide (i.e., nano-rods, -tubes, -wires, and -fibers). Polyaniline (PANI), polypyrrole (PPY), poly n-methylpyrrole (PNMPY), polythiophene (PTH), and poly ethylenedioxy thiophene (PEDOT) in the form of nano-arrays, nano-tubes, and nano-fibres) are examples of conjugated polymer nanotubes and nano-wires synthesised chemically and electrochemically. The major class of polymers that are used in OLEDs have been shown below along with its chemical structures as shown in Fig.1.

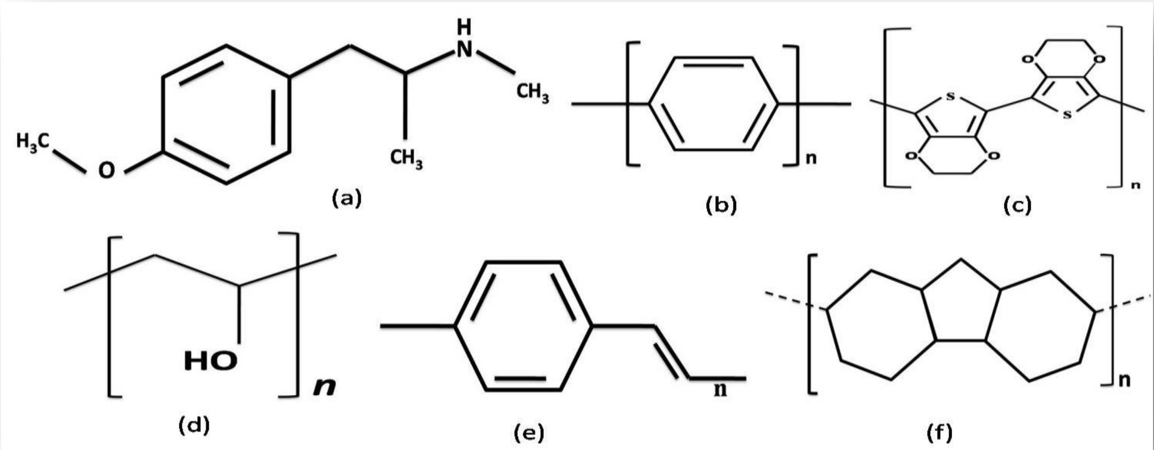

Fig.1- Chemical structures of (a) PMMA, (b) PPP, (c) PEDOT:PSS, (d) PVA, (e) PANI, (f) PPV [13]

3. Working principle and operation in OLED

An OLED device is made up of a number of functional organic layers, such as a hole transport layer, a light-emitting layer (Emissive layer), and an electron transport layer, all of which are sandwiched between an anode and a cathode. The recombination energy of electrons from the cathode and holes from the anode excites the light-emitting layer in an OLED device, and the light-emitting layer generates light when it returns to the ground state [24-25] as shown in Fig.2. To extract light from the light emitting layer, one of the electrodes is made of transparent material.

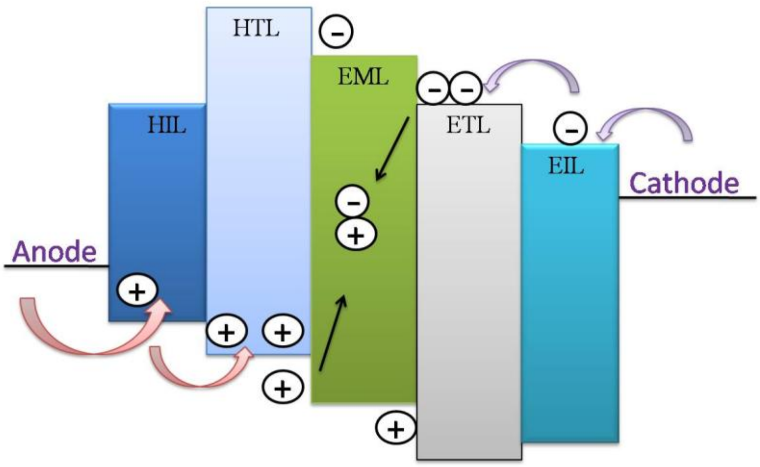

Fig. 2 – Schematic diagram of light emission mechanism in OLEDs

At the time of operation, it just connects a voltage (potential difference) across the anode and cathode to make an OLED light up. The cathode collects electrons from the power source as the electricity flows, whereas the anode loses them (or it "receives holes," if you prefer to look at it that way). The emissive layer is now negatively charged (similar to the n-type layer in a junction diode), while the conductive layer is getting positively charged as a result of the extra electrons (similar to p-type material). Because positive holes are more mobile than negative electrons, they can jump from the conductive to the emissive layer. When a hole (a lack of electron) collides with an electron, the two cancel each other out and unleash a brief burst of energy in the form of a photon. This is known as recombination, and because it occurs hundreds of times per second, the OLED produces continuous light as long as the current flows [26-32].

4. Materials for Electron Transport Layer (ETL):

The electron transport layer is employed in OLEDs from the metal cathode to achieve effective electron injection, transport, and further recombination. Metal cathodes, such as calcium, magnesium, and aluminium, are used in OLEDs with low work functions. Various types of electron transport materials have been employed to improve performance [33-35].

Many extrinsic variables, such as layer thicknesses, influence the performance of small molecule and polymer-based OLEDs, including quantum efficiency, brightness, luminance yield, and turn-on or drive voltage [36-42]. There have only been a few useful ETMs (Electron Transport Materials) created with higher electron mobility. Oxadiazole, for example, is an organic compound with good electron-transport characteristics and is one of the most extensively utilised ETMs [43-50]. The oxadiazole molecule 2-(4-biphenyl)-5-(4-tert-butylphenyl)-1, 3, 4-oxadiazole (PBD, 3) was the first employed as an electron transport material in a bilayer OLED based on a triphenylamine derivative as the emissive material, with an electron affinity of 2.16 eV and an ionisation potential of 6.06 eV [51-53]. It is found that BAlq (common electron-transport materials) has been applied as a smooth and uniform layer in all coated blue fluorescent OLEDs [54-56]. The paramount material for ETMs and their physical, chemical and optical properties have been shown in Table 1.

Table 1- Physical, chemical and optical properties of principal electron transport materials

Materials	Formula	Molecular weight(g/mole)	TGA (0.5% weight loss)	Absorption wavelength(nm)	Emission wavelength(nm)
PBD	$C_{24}H_{22}N_{20}$	354.44	>210 ^{O}C	305 nm	364,380 nm
Liq	C_9H_6LiNO	151.09	>310 ^{O}C	261 nm	331 nm
Bphen	$C_{24}H_{16}N_2$	332.4	>240 ^{O}C	272 nm	379 nm
TAZ	$C_{30}H_{27}N_3$	429.56	>250 ^{O}C	290 nm	370 nm
BAlq	C_{32} H_{25} AlN_2O_3	512.53	>230 ^{O}C	259 nm	334,477 nm
TPBi	$C_{45}H_{30}N_6$	654.76	>350 ^{O}C	305 nm	359,370 nm
BCP	C_{26} H_{20} N_2	360.45	>240	277 nm	386 nm

PBD-*2-(4-Biphenyl)-5-(4-tert-butylphenyl)-1,3,4- oxadiazole*, **Liq**- 8-*Hydroxyquinolinolato-lithium*, **Bphen**- *4,7-Diphenyl-1,10-phenanthroline*, **TAZ**- *3-(4-Biphenyl)-4-phenyl-5-tert-butylphenyl-1,2,4-triazole*, **BAlq**-*Bis(2-methyl-8-quinolinolate)-(phenylphenolato)aluminium*, **TPBi**- *2,20,2"-(1,3,5-Benzinetriyl)-tris(1-phenyl-1- H-benzimidazole)* **BCP**- *2,9-Dimethyl-4,7-diphenyl-1,10- phenanthroline*, **TGA**- *Thermogravimetric analysis*

5. Optical and electrical properties of PPV (Poly p-phenylene vinylene) and PEDOT:PSS (poly(3,4-ethylenedioxythiophene):polystyrene sulfonate) with various inorganic fillers for OLED application

There are some collected data of PPV (Poly p-phenylene vinylene) PEDOT:PSS (poly(3,4-ethylenedioxythiophene):polystyrene sulfonate) with various inorganic fillers and these hybrid nanocomposites shows better performance in OLED applications as electron transporting layer. It is seen that, by the use of composite in OLEDs that enhances the efficiency and increases operation lifetime also [57-59]. Optical and electrical properties of OLED also modified by these nanocomposites which helps to increase high brightness as well as efficiency [60-61], which is shown in Table 1 & 2.

Table 1- Data showing the electrical and optical properties of PPV nanocomposites.

SL No.	**Polymer Nanocomposites (PPV Nanocomposites)**	**Properties**						**Ref.**
		V_{on}	**J**	**L**	**LE**	**PE**	**EQE**	
1	Titanium Dioxide (TiO_2)	8.0	***	1,542	0.49	***	***	[62]
2	Silver Nanoparticles (Ag NPs)	8.1	***	1,317	0.636	***	***	[62]

3	Gold (Au)-capped TiO_2	2.0	2,600	11,630 (6.0v)	0.45	***	***	[63]
4	Cupper Indium disulfide ($CuInS_2$)	2.1	471	2701	0.89	0.200	***	[64]

Table 2- Data showing the electrical and optical properties of PEDOT nanocomposites.

SL No.	Polymeric nanocomposites (*PEDOT:PSS*)	Properties						Ref.
		V_{on}	*J*	*L*	*LE*	*PE*	*EQE*	
1	Titanium Dioxide (*TiO_2*)	9.6	***	100	7.30 (6.5v)	3.15 (3.6)	***	[65]
2	Titanium Dioxide (*TiO_2*)	3.3	***	32,291	24.5	11.2	17.1	[66]
3	Titanium Dioxide (*TiO_2*)	9.0	***	760	0.391	***	***	[66]
4	Nickel (***Ni***) Nanoparticle	4	***	26,374	5.5 (9v)	***	***	[67]
5	Copper (***Cu***) Nanoparticle	5	***	1,814	1.2 (21)	***	***	[67]
6	Nickel Oxide Nanoparticle (***NiO***)	6	***	439.2	1.4 (7V)	***	***	[67]
7	Graphene	2.75	***	25,800	2.09	0.79	***	[68]
8	Cadmium Sulphide Selenide (***CdSSe***)	4.7	67	***	1.42	0.92	***	[69]
9	Silver Nanoparticles (*Ag NPs*)	8.4	***	1,456	0.637	_	***	[70]
10	Multi-wall carbon nanotubes (*MWCNTs*)	4.5	***	2,870	***	***	***	[71]
11	Multi-wall carbon nanotubes (*MWCNTs*)	3.4	***	47,933 (10v)	***	2.42	2.33	[72]

12	Graphene Oxide	2.0	***	106700 (10.8v)	21.74 (6.4v)	13.38 (4.0v)	7.48 (6.4v)	[73]
13	Bio-Gold Nano Particles (Bio-GNPs)	6.8	***	1520	1.58	***	1.09	[74]
14	Ultra-Thin Silver Layer (UTSL)	5.6	***	5480	2.11	***	1.30	[74]

Where, **V_{on}(v)**- *Turn on voltage*, **J(mA/cm^2)**- *Current density*, **L(cd/m^2)**- *Luminanace*, **LE(cd/A)**- *Luminance Efficiency*, **PE(Im/W)**- Power Efficiency, **EQE**- External Quantum Efficiency.

6. Conclusion

Organic materials with tailored functionality can be added to well-preserved thin films, as OLEDs are constantly evolving and highlighting. As a result, the specifications for the materials are varied, spanning from processability and film formulation to electrical transport, emission, and optical qualities. More significant is the availability of effective and dependable light emitters across the whole visible spectral band. A detailed analysis of optical and electrochemical behaviours of organic -inorganic nanocomposite makes electron transport layer (ETL) material as one of the most significant factors in determining the efficiency and suitability of the OLED devices as a whole. The most important issue in producing high-performance ETL material is to provide reduced optical band gap, high electrical conductivity, reversible electrochemical reduction with high reduction potential, high specific capacitance with high dielectric merit. BAlq showed the optimum characteristics for suitable electron transporting material. The numerous forms of OLEDs that have been introduced to date in order to expand their functionality and fulfil diverse objectives have been discussed in this review paper. OLEDs are discussed in terms of their operation and mechanics. OLEDs' output is determined by their layers, with the emissive layer being particularly important. The emergence of emitters based on heavy-metal orientated metal–organic complexes, as well as their continued development. To improve the characteristics and efficiency of OLEDs, PPV (Poly p-phenylene vinylene), PEDOT:PSS (poly(3,4-ethylenedioxythiophene):polystyrene sulfonate) and its hybrid nanocomposites especially with Titanium Dioxide (TiO_2) is employed in different layers. Because of the strong spin-orbit coupling in these compounds, singlet and triplet states are combined even more than in pure hydrocarbons, making phosphorescence a permissible transfer. Meanwhile, encouraging efficiency data for OLEDs made from these materials has been reported; nonetheless, the supply and stability of deep-blue phosphorescent emitters remain a hurdle. The comparative tabular studies showed that the embodiment of inorganic filler in PEDOT:PSS (poly(3,4-ethylenedioxythiophene):polystyrene sulfonate) provides the enhanced optical and electrical properties.

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