Rising advancements in the application of PEDOT:PSS as a prosperous transparent and flexible electrode material for solution-processed organic electronics

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ABSTRACT

An organic conductive polymer, poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS), is an attractive candidate for a low-cost, low-temperature, and solution-processed electrode material for achieving high-performance flexible and stretchable thin-film devices. Unlike most organic materials, this water-soluble conjugated polymer is highly stable against chemical and physical exposure. It exhibits the most superior mechanical flexibility and the highest optical transparency and electrical conductivity among all organic conductors. Therefore, this conductive polymer is among the most promising alternatives to the expensive, rigid, and brittle metal oxide- and even metal-based electrode materials, such as indium tin oxide (ITO) and gold, in the future solution-processed electronic devices. Nevertheless, the intrinsic conductivity of PEDOT:PSS is typically below $1 \text{ S cm}^{-1}$, which is too low for such devices. Fortunately, the material properties of PEDOT:PSS, including its conductivity, are easily tuned by employing a large number of simple approaches. In this paper, the reports on the successful application of PEDOT:PSS to a wide range of solution-processed organic devices, such as organic light-emitting diodes (OLEDs), organic thin-film transistors (OTFTs), and organic photovoltaics (OPVs), are reviewed. The recent progress in the development of highly conductive PEDOT:PSS-based films for electrode applications in the field of organic electronics is the main focus of the discussion herein.

1. Introduction

Conductive polymers have been significantly attractive for a wide range of electronic applications owing to their key advantages, such as their easy handling, solution- and low-cost processability, chemical diversity and tuneability, and biocompatibility, as well as their unique combination of mechanical and optoelectronic properties [1,2]. Unlike the typical polymers mostly used for insulating and packaging purposes in the plastics industry, the conductive polymers allow electricity to pass through owing to the alternating single $\sigma$ and double $\pi$ bonds among the carbon atoms in their structure. These synthetic materials are organic macromolecules with conjugated backbones that contribute delocalized $\pi$ electrons via $sp^2$ hybridization, leading to the actual electrical conductivity through the molecular backbone [3,4]. Their superior features compared to their inorganic counterparts have made them increasingly interesting for both academic and industrial research and engineering to fully realize the newly emerged field of ultra-thin and plastic electronics. Since the discovery of the first-ever conductive polymer, a wide variety of different synthetic conductive polymers have been developed and extensively explored [5]. Depending on the extent to which they conduct electricity, these low-density organic molecules may be utilized both as semiconductors for wide-ranging electronic device applications and as conductors to replace the metal components of the devices. Besides their easy processability and flexibility, polymers also have a number of other important advantages over the vast majority of electronic materials, including metals, such as their light weight and non-corrosive nature [4,6–10].

Among the conductive polymers, poly(3,4-ethylene dioxythiophene) polystyrene sulfonate (PEDOT:PSS) stands out as the most successful and widely studied, which has also been practically and commercially realized [11–13]. This prosperous conductive polymer is a mixture of two polymer ionomers, positively charged...
PEDOT and negatively charged sulfonated polystyrene (PSS), where PSS acts as a template polymer and surfactant [13]. PSS was chosen based on its water solubility, to enable the solution processability of PEDOT in polar solvents, including water, as well as to highly increase and stabilize the conductivity of PEDOT via charge balancing [13–16]. PEDOT:PSS has been widely applied to organic light-emitting diodes (OLEDs), organic thin-film transistors (OTFTs), organic photovoltaics (OPVs) including organic solar cells (OSCs) [13], perovskite solar cells (PSCs) [17], cutting-edge technologies like touch-screens, electronic papers, and next-generation energy storage and conversion devices functioning as capacitors, batteries, thermoelectric devices, etc. [12,13,18], due to its easy synthesis, low cost, and other unique features, such as its low-temperature processing compatible with organic devices [19], easily tunable viscosity [20], high ductility, high transparency in the visible-light range, and high electrical conductivity [12,13,16]. PEDOT:PSS empowers the fabrication of highly transparent, flexible, ultra-thin, and even biodegradable functional films [12,21]. In addition to its chemical and physical durability, the sufficiently stretchable [16,21] conductive polymer is also stable against environmental factors [13]. The feasible control over the material properties to adjust the electrical conductivity and transmittance through a simple PEDOT:PSS ratio adjustment, along with its solution processability and good-film-forming ability [13] and its work function (WF) and film morphology tenability, makes it a ubiquitous substance that can be employed both as an active layer and as an electrode material for reproducible and practical device applications [22–25].

In the following two sections of this review paper, the fundamental techniques reported for the improvement of the electrical conductivity of PEDOT:PSS as well as the preeminent deposition methods for forming PEDOT:PSS films will be summarized. The potential applications of the polymer will also be briefly presented in the fourth section. In the following fifth section and its subsections, the recent advancements in the field of PEDOT:PSS-based electrodes for organic devices like OLEDs, OTFTs, and OPVs will be presented and discussed in detail. Finally, a brief summary and the paper’s conclusions will present an overview of the main points of the article to the reader.

2. Techniques for the enhancement of the electrical conductivity of PEDOT:PSS

Despite its relatively high electrical conductivity, and the commercial interest in low-cost, large-area, and flexible electronics [13], the bare PEDOT:PSS is not sufficiently conductive for practical application for meeting the high-level requirements of industrial development. The pristine PEDOT:PSS usually exhibits less than 1 S cm$^{-1}$ electrical conductivities [16,26–30]. Therefore, various effective methods have been developed to increase its electrical conductivity, with either doping or de-doping the polymer as the target [15,16]. Very frequently, the desired performance of PEDOT:PSS is achieved through the combination of the two [31–34]. Doping aims to increase the concentration of the mobile charge carriers responsible for the conduction through the polymer backbone, by adding extra charges to the whole structure as well as neutralizing some of the PSS$^-$ polyanions. This mechanism is usually referred to as ‘secondary doping’ [28] because the conductive structure of PEDOT:PSS is achieved primarily through the p-type doping of PEDOT with PSS, leading to a very stable ionic bonding between the two counter ions of PEDOT$^+$ and PSS$^-$. De-doping intends to remove the excess of the insulating PSS from the whole structure to improve the electrical conductivity, because the main reason for the low electrical conductivity is the confinement of a significant segment of the conductive PEDOT chains by the insulating PSS chains [14]. Figure 1(b) shows the structural model of PEDOT:PSS proposed by Soleimani-Gorgani to illustrate the role of PSS in the reduction of the conductivity of PEDOT [14].

The approaches for improving the conductivity of PEDOT:PSS can be categorized into two groups: the pre- and post-treatment methods [11,12,15,16]. In this section, these methods will be comprehensively examined.

2.1. Pre-treatment methods

Pre-treatment methods involve the simple addition of polar, protic, and aprotic organic solvents miscible with water, such as N-methyl-2-pyrrolidone (NMP), dimethyl sulfoxide (DMSO), N,N-dimethylformamide (DMF), and ethylene glycol (EG) [32,33,35–37], or of organic compounds that get dissolved in water, such as sorbitol [33,38,39], as well as the incorporation of low-viscosity and transparent ionic liquids like 1-ethyl-3-methylimidazolium tetracyanoborate (EMIM BtI) [16,29] and ionic/non-ionic surfactants like cetrimonium bromide (CTAB), sodium dodecyl sulphate (SDS), and polyethylene glycol p-(1,1,3,3-tetramethylbutyl)-phenyl ethers (Triton X-100) into the aqueous dispersion of PEDOT:PSS [11,40]. The addition of pre-treatment agents (dopants) into the polymer solution will substantially affect the electrical conductivity, transmittance,
and morphology of the formed films. The rate of pre-treatment efficiency is highly dependent on the dopant used [36]. For instance, DMSO has been proven to be so effective that it is added to the commercially available PEDOT:PSS solutions by default [29,37]. A recent research showed that the effects of the addition of organic co-solvents into the aqueous dispersion of PEDOT:PSS are highly dependent on the molecular weight, boiling point, and dipole moment of the utilized co-solvent. That is, the higher the molecular weight, boiling point, and dipole moment of the co-solvent are, the higher the conductivity increase and morphology change rates of the polymer films. This can be explained by the stabilization of the negatively charged PSS$^-$ and the formation of neutral polystyrene sulfonic acid (PSSH), followed by the phase separation of the two segments of the block polymer caused by a proton (H$^+$) donation from the polar solvents with large dipole moments. It was found that the size of the solvent molecules (molecular weight) plays an important role in the effective expansion of the space between the PEDOT and PSS chains, as well as in the particle size of the polymer, whereas the boiling points of the solvents provide the necessary time for the favorable morphological rearrangement and conductivity enhancement of the films [17,39]. Despite its great impact on the film morphology, including increased film thickness and roughness, the addition of co-solvents like EG, diethylene glycol (DEG), and triethylene glycol (TREIG) resulted in only minor changes in the films’ optical properties [14]. The relation between the morphological changes and the increased electrical conductivity after the treatment can also be clearly explained by the characterization of the films through atomic force microscopy (AFM), where it is generally observed as the enlargement of the bright regions assigned to the conductive PEDOT, and as the shrinkage of the dark regions assigned to the insulating PSS (Figure 2) [16,29].

The PEDOT:PSS solution will also improve the crystallinity and molecular ordering of the PEDOT segment in the films [30,41]. Pre-treatment can also imply doping of the dispersion through the addition of specific impurity species [13,42]. This type of uncomplicated doping has been shown to significantly enhance the electrical performance of the polymer.

The PEDOT:PSS solution can also be effectively doped with protonic or sulfonic acids [16,43]. A recent example of this type of research on acid doping, completed in 2017 by F. Wu et al., revealed that the in-situ doping of PEDOT:PSS with a solid acid like chloroplatinic acid (H$_2$PtCl$_6$) can more greatly enhance the electrical conductivity than in-situ doping with organic solvents, including DMSO and EG. The doping occurs via two steps: the reduction of Pt$^{4+}$ to Pt$^{2+}$ through electron transfer from PEDOT, followed by the reduction of the negatively charged PSS$^-$ ions to neutral PSSH through proton transfer from H$_2$PtCl$_6$, which in turn results in the phase separation between the PEDOT and PSS chains, leading to the more favorable conformation of the PEDOT structure and a lower energy barrier for efficient charge hopping [44]. Another group of researchers reported the high electrical conductivity of PEDOT:PSS films resulting from the combination of benzenesulfonic acid doping of the polymer solution with the simple DMSO post-treatment of the polymer films [45].

Some mechanical ways of segregating the excess PSS from the PEDOT:PSS formulation have also been suggested. For instance, S. Kim et al. developed an easy method of purifying the PEDOT:PSS dispersion using an ultrafiltration cell with a 100 nm membrane pore size, which is larger than the free PSS and other impurity species and smaller than the whole PEDOT:PSS structure, to remove all the extra PSS and the unwanted impurities from the polymer solution. The repeated filtration resulted in a significantly higher conductivity of the PEDOT:PSS films. Furthermore, the addition of
DMSO to the purified polymer solutions lowered the films’ sheet resistance ($R_{sh}$) to $37.8 \pm 4.5 \, \Omega \cdot \square^{-1}$, which is sufficiently low for electrode applications [46]. The proposed purification method and the relative conductivity increase are shown in Figure 3.

2.2. Post-treatment methods

The electrical conductivity of the already-deposited PEDOT:PSS films can also be boosted by several orders of magnitude, to make it suitable for replacing the conventional conductors like metals or conductive oxides, including tin-doped indium oxide (ITO) and fluorine-doped tin oxide (FTO) [13]. For this purpose, a number of post-treatment methods have been developed and investigated, which can lead either to doping or de-doping of the polymer films. These post-deposition approaches imply treating the deposited polymer films with various organic and inorganic substances representing a diverse set of compounds, from the simplest alcohol methanol (MeOH) to more complex chemicals like imidazoles [11,47,48], phenol, and other polar organic solvents, or with water co-solvents with high boiling points, such as DMSO, EG, DEG, TRIEG, ethanol (EtOH), isopropyl alcohol, terahydrofuran (THF), and acetone, as well as with different salts [27,37,49,50] and acids [51]. While these methods appear to be simple solvent treatments of the films, there are notable differences in their procedures. The representative techniques are spin coating the ready PEDOT:PSS films with solvents [52], immersing (dipping) the films in solvents, dropping the solvents on the films [11,47], exposure of the films to polar solvent vapor (known as polar-solvent-vapor annealing or PSVA) [53,54], and post-spin/spray rinsing of the films [55,56]. In addition, the effectiveness and suitability of these methods are highly dependent on the materials used. This issue primarily concerns the substrate materials as polymer electrodes are commonly designed for the realization of flexible devices, and while all solvents and acids are applicable for the post-treatment of PEDOT:PSS films deposited on rigid substrates like glass, some of them are not compatible with the flexible substrates. For instance, the post-treatment of PEDOT:PSS films with the strong sulfuric acid $H_2SO_4$ is known to increase the electrical conductivity by several orders of magnitude, up to the level of stable metallic
conduction, without sacrificing the transmittance of the films [57], but it can significantly damage the underlying layer in the case of plastic substrates. The weak phosphoric acid, on the other hand, is an effective and safe post-treatment agent for both rigid and flexible substrates [16,58].

The mechanism of the conductivity enhancement of PEDOT:PSS after treatment has been extensively studied. The main factors leading to the increase in electrical conductivity are said to be the removal of the excess PSS from the films, the extra charges added to stabilize the PSS$^-$ counter ions, and the modification of the films’ morphology as well as the conformational changes of the PEDOT chains, the rearrangement of the whole PEDOT:PSS structure, and the phase separation between the PEDOT and PSS segments [17,28]. Comparing the performance of three different alcohols, MeOH, EtOH, and propanol, as post-treatment agents, Alemu et al. discovered that the conductivity enhancement is highly dependent on the hydrophilicity and dielectric constants of the solvents used, which lead to different conformational and morphology changes in the polymer films. Although all the solvents yielded much higher transmittance and lower resistance of the treated films compared to the pristine one, the solvent with the simplest structure, MeOH, was found to be the most effective one, leading to conductivity increase from 0.3 to 1362 S cm$^{-1}$, almost equal to the conductivity of ITO [26].

The reported achievements in the development of highly conductive PEDOT:PSS films using the above-discussed pre- and post-treatment methods are summarized in Table 1.

The aforementioned post-treatment techniques are pertinent only for the direct exposure of PEDOT:PSS films to solvents. Some progressive studies discovered that the molecules of polar solvents can permeate through organic layers into the underlying PEDOT:PSS layer, and modify its properties. In other words, the removal of the excess PSS from the film bulk, the favorable film morphology, and the increased $W_f$ and conductivity due to solvent treatment are achievable even for PEDOT:PSS films already buried under an organic layer, simply by depositing the solvent on the organic layer, because PEDOT:PSS molecules are very sensitive to the molecules of polar solvents [52]. Previous studies have shown that the $W_f$ of PEDOT:PSS can also be tuned by simply exposing the annealed films to UV-ozone radiation for a short time [22,25].

The innovative methods that have been developed to enhance the electrical conductivity of PEDOT:PSS also include the use of water-soluble and naturally occurring materials like tannic acid or of water-soluble bio-materials like vitamins for the pre- or post-treatment of the polymer [42,59]. The recent reports suggest that PEDOT:PSS can exhibit conductivity values similar to or even surpassing those of ITO in combination with silver nanowires (Ag NWs) [9], graphene, graphene oxide, or carbon nanotubes (CNTs) [60]. Such composite materials for transparent hybrid electrodes are achieved by embedding the mentioned nanostructures into PEDOT:PSS films through the conventional pre- and post-treatment techniques and through novel methods like hot-pressing [48,61–64]. The resultant PEDOT:PSS films exhibit not only high conductivity but also remarkable stretching degrees [12].

Aside from the aforementioned conditions, the film thickness [29,65] and thermal annealing [39,54,66] also affect the electrical conductivity of PEDOT:PSS films.
Table 1. Recent advances in the improvement of the electrical conductivity of the commercially available PEDOT:PSS dispersions.

| Commercial ink name | Range of applications | Treatment agent | Pristine conductivity ($S\,cm^{-1}$) | Improved conductivity ($S\,cm^{-1}$) | Literature ref. no. |
|---------------------|----------------------|-----------------|-------------------------------------|-------------------------------------|---------------------|
| Pre-treatment methods |                      |                 |                                     |                                     |                     |
| Clevios PH1000 (H. C. Starck) | Electrode material | Organic solvent (EG) | ~ 1                                  | 731                                 | 33                  |
| Baytron P (H. C. Starck) | Electrode material | Organic solvent (DMSO) | 0.549                                | 575                                 | 37                  |
| Baytron P (H. C. Starck) | Electrode material | Organic solvent (DMF) | 0.8 ± 0.1                            | 80 ± 30                              | 38                  |
| Baytron P (H. C. Starck) | Electrode material | Organic solvent (THF) | 0.8 ± 0.1                            | 30 ± 10                              | 38                  |
| Baytron P VP A4083 (H. C. Starck) | Hole transport material | Alcohol sorbitol | 24.7 ± 1.3                           | 913 ± 1.3                            | 41                  |
| Clevios PH1000 (H. C. Starck) | Electrode material | Ionic liquid (EMIM TCB) | 0.68                                | 2084                                | 30                  |
| Clevios PH1000 (H. C. Starck) | Electrode material | Ionic liquid (BMIM BF4) | 0.68                                | 287                                 | 30                  |
| Clevios PH1000 (H. C. Starck) | Electrode material | Organic solvent (DMO) | 0.68                                | 575                                 | 30                  |
| Clevios PH1000 (H. C. Starck) | Electrode material | Organic solvent (1,1-dimethyl-2-imidazolidinone, DMI) | 2                                  | 812                                 | 11                  |
| Clevios PH1000 (H. C. Starck) | Electrode material | Tannic acid | 2.33                                | 25.5                                 | 44                  |
| 5 wt% DMSO-doped PEDOT:PSS* | Electrode material | Mechanical purification through ultrafiltration | 1100                                | 2000                                | 47                  |
| Post-treatment methods |                      |                 |                                     |                                     |                     |
| Clevios™ PH1000 | Electrode material | Organic solvent (EtOH) | ~ 0.8                             | 1.8                                  | 48                  |
| Clevios™ PH1000 | Electrode material | 2-Methylimidazole in EtOH | ~ 0.8                             | 930                                  | 48                  |
| Clevios PH1000 | Electrode material | Organic solvent (MeOH) | ~ 0.2                             | 370                                  | 28                  |
| Clevios PH1000 | Electrode material | Organic solvent (DMSO) | ~ 0.2                             | 890                                  | 28                  |
| Clevios PH1000 | Electrode material | Organic solvent (EG) | ~ 0.2                             | 960                                  | 28                  |
| Clevios PH1000 | Electrode material | Salt solution (methylammonium bromide in DMSO) | ~ 0.2                             | 1280                                 | 28                  |
| Clevios PH1000 | Electrode material | Salt solution (methylammonium iodide in DMSO) | ~ 0.2                             | 1660                                 | 28                  |
| Clevios PH1000 | Electrode material | Organic solvent (EG) | 2.5 ± 1.2                           | 971 ± 166                             | 51                  |
| Clevios PH1000 | Electrode material | Inorganic (sulfuric) acid (H$_2$SO$_4$) | 2.5 ± 1.2                           | 2118 ± 102                           | 51                  |
| Clevios PH1000 | Electrode material | Double-treatment with EG and H$_2$SO$_4$ | 2.5 ± 1.2                           | 1968 ± 150                           | 51                  |
| Clevios PH1000 | Electrode material | Inorganic (sulfuric) acid (H$_2$SO$_4$) | 0.3                              | 3065                                 | 57                  |
| Clevios PH1000 | Electrode material | Inorganic (phosphoric) acid (H$_3$PO$_4$) | 1                                | 1460                                 | 58                  |
| Combined pre- and post-treatment methods |                      |                 |                                     |                                     |                     |
| Clevios PH1000 (H. C. Starck) | Electrode material | Pre- and post-treated with EG | ~ 1                                 | 1418                                 | 33                  |
| Clevios PH1000 (H. C. Starck) | Electrode material | Pre-treated with DMI, post-treated with with MeOH | 2                                  | 986.2                                | 11                  |
| Clevios PH1000 (H. C. Starck) | Electrode material | Pre-treated with EG, post-treated with MeOH | 2                                  | 933.5                                | 11                  |

*Synthesized by the authors following the synthesis protocol of Baytron P.

Unlike the a high film thickness, however, a high annealing temperature generally has an adverse effect on the conductivity of PEDOT:PSS. Investigations of the annealing conditions and their influence on the PEDOT:PSS film properties have shown that the optimal heating condition for removing the residual water from the films without affecting their performance is 130°C, whereas heating them to above 200°C significantly degrades the polymer, leading to its critical weight reduction and chemical decomposition [66]. An alternative and ultrafast annealing method for PEDOT:PSS films was revealed by another interesting study, using near-infrared (NIR) curing, where the minimum time of 240 s for drying the polymer in a conventional oven was reduced to 2 s using NIR rays. This kind of radiative thermal annealing was also found to be highly compatible with flexible substrates, and to lead to film $R_{sh}$ values lower than those of the films annealed in a conventional oven for a much longer time. Such effect was attributed to the strong absorbance of PEDOT:PSS in the NIR region, which allows the fast and low-energy drying of the polymer films [67].

3. Deposition techniques for PEDOT:PSS films

Conductive thin and uniform PEDOT:PSS polymer films are formed by a wide range of environment-friendly coating techniques developed to produce high-quality solution-processed films at a low cost [16]. The common techniques include spin coating, dip coating, slot die coating, bar coating, spray coating, doctor blading, and drop casting [46,64,68,69]. Some printing technologies, such as roll-to-roll (R2R), screen, and inkjet printing, are also among the wet-film deposition methods highly suitable for PEDOT:PSS [13,70]. These large-scale and high-volume deposition advantages of PEDOT:PSS, combined with its conductivity enhancement opportunities, lead the way towards the replacement of the high-cost and high-energy-vacuum-deposited or vacuum-sputtered ITO.
[33,37] to take plastic electronics to new extremes. Among these film deposition techniques, inkjet printing is particularly interesting as it enables the creation of the finest details of high-resolution electronic devices at a low cost, by simply transferring the digital images onto plastic, fabric, and other rigid or flexible substrates using the conventional computer printers. This deposition method eliminates material waste and completely prevents pollution by precisely and rapidly transferring the necessary amount to the desired regions, leading to considerably reduced fabrication costs [70–72]. It is also the most preferred technique for the patterning of polymer conductors [19]. From a mechanical viewpoint (the surface tension of the used ink), the water-based ink formation makes PEDOT:PSS highly suitable for inkjet printing [14]. In the beginning of this decade, a facile deposition of PEDOT:PSS as anode electrodes for OLEDs by an ordinary desktop inkjet printer was suggested by Ummartyotin et al. [73]. Recently, Bihar et al. demonstrated PEDOT:PSS electrodes inkjet-printed on paper for medical device applications [74].

4. Range of applications of PEDOT:PSS

The formulation of PEDOT:PSS generally yields an ink dispersion, which is widely used for numerous purposes, ranging from specific protective coatings and planarization layers to the hole transport layers (HTLs) and non-metallic electrodes of various electronic devices [1,15–17]. PEDOT:PSS has the most efficient structure among all organic thermoelectric materials [17,18,43]. As this discussion focuses on identifying the achievements in the sphere of PEDOT:PSS-based electrodes as well as the development directions therein, the alternative applications of PEDOT:PSS will be briefly mentioned. These applications mainly depend on the transparency and flexibility of this polymer as well as on its compatibility with diverse and large-area fabrication methods like R2R manufacturing, inkjet printing, and spin coating. The impressive development directions in this field include antistatic, protective, or even shielding film coatings for several appliances [14,69], stable polymeric electrolytes for capacitors, effective interfacial and buffer layers [13,33], thermally stable surface coating and binder layers for lithium batteries [75,76], and common solution-processed hole injection layers (HILs) and HTLs for OLEDs, OTFTs, OPVs, organic electrochemical transistors (OECTs), etc. [13,15,16,37]. In the recent past, D. H. Yoon et al. proposed that PEDOT:PSS be used as a multifunctional material for composite battery electrodes [77]. The application of PEDOT:PSS as an electrode material for large-area, flexible, and wearable organic electronics has been considerably interesting due to its excellent optomechanical properties, relatively high electrical conductivity, and advantageous processing methods arising from its polymer nature [11,16].

5. PEDOT:PSS-based polymer electrodes

Optoelectronic devices, including OLEDs, OPVs, and liquid-crystal displays (LCDs), require thin films of conducting materials for use as transparent electrodes to emit or absorb light at least from one side. These materials should have higher than 80% optical transmittance in the visible-light region for efficient light emission or absorption, as well as electrical conductivities greater than or equal to $10^3 \text{S cm}^{-1}$ to provide the necessary charge carrier conduction for low operational voltages [26,28,31,33,38,78–80]. A conventional electrode material that combines the above-mentioned two essential parameters is the widely applied ITO with higher than 90% transmittance and conductivity values reaching $10^5 \text{S cm}^{-1}$ [33,66]. Besides the fact, however, that the film thicknesses of the present-day and forthcoming generations of electronics are approaching nanometer levels, such electronics also demand high flexibility and a light weight from the device components. Sadly, the high-performance ITO belongs to the family of ceramic materials, which are naturally too brittle for flexible device application [9,33]. In addition, intrinsic brittleness is not the only demerit of ITO. This transparent conductive oxide (TCO) is usually obtained through high-temperature [37,81,82] and expensive deposition methods using the rare metal indium, which makes its synthesis increasingly expensive [37,66]. Moreover, the patterning of ITO on glass substrates requires wet chemical etching using acids that are highly toxic. There is also a possible diffusion of the tin (Sn) and indium (In) metals as well as of the oxygen ($O_2$) atoms into the organic layers, leading to the device failure and degradation [66,81,83]. Therefore, finding an alternative electrode material to replace ITO is an actual challenge that the optoelectronics field is facing today. Different kinds of materials involving CNTs, graphene, Ag NWs, and conducting polymers have been tested for this purpose [9,33,34]. Although, each one of these materials has merits for replacing ITO, they all have drawbacks incompatible with practical application. In addition to the main issues, such as the critical surface roughness of the electrodes that lead to high leakage currents, thereby limiting the device performance [17,78,79], and undesirable electrical parameters like a low $W_f$ and a high $R_{sh}$, which increase the power consumption and crack down on the benefits of the material [78,79], the synthesis and processing of CNTs, graphene, and Ag NWs are not as cheap and easy as, for instance, those of conductive polymers [13].
These polymers are inherently highly flexible, and their fabrication is environment-friendly and low-cost. Thus, conductive polymers are regarded as among the most promising candidates for replacing ITO, despite the fact that their electrical conductivity is considerably lower. The conductive polymer PEDOT:PSS has been broadly studied as one of the most suitable materials for producing transparent, colorless, and flexible electrodes for capacitors, batteries, thermoelectric devices, liquid-crystal devices, photodiodes, OLEDs, OFETs, and OPVs as is or as a part of a composite [12,18,20,31,46,84].

5.1. PEDOT:PSS-based thin-film electrodes for OLEDs

OLEDs are flat self-light-emitting optoelectronic devices commonly applied for display and solid-state lighting technologies [11,33,65]. The simple structure of OLEDs consists of multilayers formed from organic materials functioning as an HTL, a light-emitting active layer, and an electron transport layer (ETL) stacked between two transparent and reflective electrodes called ‘anode’ and ‘cathode’, respectively [33]. The main parameters defining the efficiency of OLED devices include the external quantum efficiency (EQE), turn-on voltage (\(V_T\)), current density, current efficiency, power efficiency, color quality, and lifetime [33,66]. OLEDs are ultra-thin, ultra-light, flexible, and simple in design, as well as suitable for large-area electronics [11]. OLED displays are more efficient than liquid crystal displays (LCDs), and they deliver better image quality for lower power consumption. In addition, they do not require a backlight, as LCDs do [85]. OLEDs can also be a safe and excellent light source. They are both transparent and color-tunable [86]. Although the first practical OLED appeared a little more than 30 years ago, the flexible OLEDs have been on the market for many years, and are the important components for the realization of the future flexible, wearable, foldable, and even biodegradable optoelectronic applications [85–87].

The conventional OLEDs are mostly bottom-emission devices with ITO anodes patterned on glass substrates [66]. Despite its outstanding optical transparency and electrical conductivity which makes it the main practical transparent conductor in the display industry, ITO is not the ideal electrode material for OLEDs [28,65]. In addition to its high cost, rigidity, and fragility, it is also unstable against electrochemical factors, in addition to the serious issues related to its film quality, such as its surface roughness, morphology irregularities, and other film defects [33]. Besides, it has poor transmittance in the blue and NIR regions. Furthermore, the refractive index of ITO electrodes is higher than that of any other layer of an OLED, including the glass substrate [33]. It is around 1.8–2.2, depending on the ITO fabrication and treatment methods [37,78,82,88], while the refractive indices of the organic layers and the glass substrates of OLEDs are usually around 1.7–1.9 and 1.5, respectively [82,89]. This refractive index mismatch between ITO and the overlying organic layers as well as the underlying glass substrate leads to the total internal reflection (TIR) of light. As a result, only a small portion of the generated light is emitted whereas 80% of it is either trapped between the ITO and the organic layers as well as between the ITO and the glass substrate in the waveguide mode or reflected back inside the OLED in the substrate mode [90]. Therefore, various light extraction methods, including the usage of an additional refractive-index-matching layer, have been developed to limit the total amount of optical loss inside devices. Moreover, the drawbacks of ITO extend to its wettability issues, which can be seen in the poor adhesion between ITO and the overlying organic layers as well as the underlying polymer substrates in the case of flexible devices [28].

Although the conductivity of the untreated PEDOT:PSS is four orders of magnitude lower than that of ITO, PEDOT:PSS is among the main candidates for replacing ITO due to its high transmittance (efficiently emitting the generated light through it) and controllable and lower surface roughness (for better anode/HTL contact) as well as its high (\(W_f\)) [13], which is necessary for effective hole injection from the anode into the organic layers of the device via the formation of ohmic contacts [20,34,91]. In addition, PEDOT:PSS has a 1.5–1.6 refractive index, which is considerably lower than that of ITO and closer to those of the OLED organic layers and glass substrate, which in turn eliminates the necessity for an additional index-matching technique [37,82,91]. PEDOT:PSS’s low electrical conductivity, however, severely hinders its practical use. Another hindrance is its deposition from a water-based solution, as the possible water residual in the electrode layer can lead to the degradation of the overlying organic layers, thus reducing the efficiency and lifetime of the complete OLED devices [21,33,65,66]. Therefore, the approaches to optimizing the performance of PEDOT:PSS and using it as a practical electrode material for ITO-free OLEDs should integrate methods of enhancing the conductivity of the polymers and of eliminating the stability issues of the fabricated devices.

The first transparent OLED based on PEDOT:PSS electrodes with high efficiency and a long lifetime was reported in 2013 by Y. H. Kim et al. Effective structure engineering developed by carefully combining empirical studies with process simulation for optimizing the HTL thickness and doping concentration to provide an ohmic contact between the polymer electrodes and the HTL.
yielded an EQE comparable to that of ITO-based OLEDs, along with an improved lifetime, device efficiency, and a very low $V_T$ [65]. Further, PEDOT:PSS designed as a transparent bottom electrode on top of a low-refractive-index polymer planarization layer and randomly distributed metal oxide nanoclusters was used to realize an internal color-stabilizing and scattering structure for efficient light extraction from white OLEDs (WOLEDs) [92]. A simple vapor treatment of PEDOT:PSS films with organic solvents with low boiling points, such as alcohols like MeOH, EtOH, and 2-propanol, was also found to significantly reduce the sheet resistance of the films to the extent that makes the films suitable for application as anodes in ITO-free OLEDs. This kind of PSVA subnamed ‘alcohol vapour treatment’ (AVT) causes a screening effect that induces phase separation between the PEDOT and PSS segments via the interaction of the hydrophilic molecules of the alcohols and PSS, resulting in longer and better-connected PEDOT chains for improved charge conduction [16,93]. Effective separation of the PEDOT and PSS phases to obtain highly conductive films for OLED transparent anode applications can also be achieved by dipping the films in a heated hydric acid solution [94]. The combination of the pre- and post-treatment methods is also a frequently practiced approach leading to the highly enhanced conductivity of PEDOT:PSS for ITO-free OLED devices [95,96].

ITO-free OLEDs are expected to be applied in the future in wearable electronics like wearable display devices [21]. Over the past decade, several successful approaches were suggested to achieve this. For instance, H. Kim et al. demonstrated OLEDs fabricated via solution processing on soft textile substrates with PEDOT:PSS anodes. Despite the severe limitations originating from the textile substrates, the devices exhibited stable operation under bending stress and angular independency of the electroluminescence (EL) spectra with 9.72 cd A$^{-1}$ and 7.17 lm W$^{-1}$ high luminous and power efficiencies, respectively [97].

As mentioned in the second part of this review article, composite electrodes consisting of PEDOT:PSS and highly conductive and stretchable materials like Ag NWs, CNTs, graphene, and graphene oxide exhibit more stable and superior performance even compared to that of ITO electrodes. The main purposes for the combination of these materials are the integration of their advantageous features, such as the high conductivity and stretching capacity of Ag NWs, CNTs, or graphene with the simple, low-cost, and low-temperature processing of PEDOT:PSS, and the resolution of the material-related issues, such as the surface roughness or stability [60–63].

Another exciting approach to obtaining highly conductive polymer anodes for OLEDs was proposed by Levermore, according to which polymer conductivities as high as 1100 S cm$^{-1}$ are achievable through the vapor phase deposition of 3,4-ethylenedioxythiophene (EDOT) monomers, followed by their in-situ polymerization on a heated substrate without PSS participation [98].

Patterning PEDOT:PSS electrodes for OLEDs is also one of the effective ways of achieving high-performance devices with polymer electrodes. Besides the common inkjet printing, the polymer has also been patterned as anodes for OLEDs through oxygen plasma etching using a stainless-steel mask [99]. Moreover, despite the fact that the traditional photolithography is not compatible with PEDOT:PSS due to the mutual destructive influence of the materials used in photolithography and the polymer, a feasible photolithographic scheme has been suggested for PEDOT:PSS anode patterning. Here, a thin layer of a noble metal like silver (Ag) was used as an etching mask on PEDOT:PSS films to block the interaction of the materials, and was eventually removed with an acidic etchant without affecting the patterned polymer anodes, as PEDOT:PSS is acid-stable. The OLEDs built on the flexible substrates with PEDOT:PSS anodes patterned via the conventional photolithography demonstrated high performance comparable to that of ITO-based OLEDs [100]. The scheme of this photolithographic method and the device performance are shown in Figure 4.

Moreover, H. J. Lee et al. introduced a simple solution-processed method called ‘negative mould transfer printing (nMTP)’ to successfully pattern PEDOT:PSS anodes on flexible substrates for OLED applications. The fabricated flexible OLEDs using doped and nMTP-patterned PEDOT:PSS anodes exhibited electrical performances comparable to those of ITO anode-based devices. Despite its simplicity, nMTP enables the formation of clear micro- and nano-patterns, and is compatible with large-area device manufacturing [101]. Figure 5(a) illustrates the nMTP pattern formation procedure.

Y.-F. Liu et al. achieved highly conductive PEDOT:PSS anodes, thereby improving the efficiency of ITO-free flexible OLEDs. For this purpose, a template stripping process (TSP) was applied, combined with the post-treatment of PEDOT:PSS films, for the enhancement of the surface morphology to obtain ultra-smooth polymer anodes [102]. The surface morphologies obtained after this process are shown in Figure 5(b)-(e).

5.2. PEDOT:PSS-based thin-film electrodes for OPVs

OPVs, particularly OSCs, are especially interesting due to their potential for replacing the conventional silicon-based cells. They are low-cost, easy to fabricate, and economically more practical for mass production and large-scale power generation compared to the inorganic
Figure 4. Photolithographic patterning of PEDOT:PSS anodes using an Ag layer: a) deposition of PEDOT:PSS, Ag, and the photoresist on the substrates; b) UV exposure of the photoresist films through a mask; c) formation of the photoresist patterns on the Ag layer after their development; d) etching of Ag with an acidic etchant; e) etching of PEDOT:PSS with O₂ plasma; f) photoresist stripping; g) PEDOT:PSS patterns on the substrate after Ag layer removal; and h) current density-voltage and i) current efficiency of the OLEDs. The insets in h) and i) show the semi-log plot of the current density and an image of the working ITO-free OLEDs, respectively [100].

Figure 5. a) PEDOT:PSS patterning through nMTP [101]. Surface morphologies of PEDOT:PSS films with and without the template stripping process (TSP): b) post-treated films without TSP; c) untreated template-stripped films; d) untreated films without TSP; and e) post-treated template-stripped films [102].
photovoltaics [13]. In addition, their constituent materials make them the most suitable candidates for thin, lightweight, and flexible applications. Their solution processability is once again their key advantage [48]. These third-generation devices use organic semiconductors to absorb and convert sunlight to electricity by imitating the natural process of photosynthesis [103]. Technically, OSCs are also diodes with a structure similar to that of OLEDs. Their main difference from OLEDs is that they use a reverse process for the organic (donor–acceptor) active layer to transform the solar energy into electrical energy [104]. Figure 6 demonstrates the similarities in the device structures of the two above-mentioned optoelectronic devices.

Similar to OLEDs, the basic operation of OSCs also requires at least one transparent electrode, and despite its severe drawbacks, ITO is also still the most common transparent electrode material for the conventional OPVs [105]. Significant progress has been made, however, towards ITO-free high-performance flexible OSCs, and PEDOT:PSS has been among the most investigated alternative electrode materials due to its great potential for the construction of the future soft electronic applications [16]. Early materials like OSCs based on small molecules were produced on both rigid glass and soft plastic substrates using transparent PEDOT:PSS electrodes, whose conductivity was optimized via the combined influence of solvent pre- and post-treatment, along with the thermal post-annealing of the electrode films. The devices with the optimized polymer electrodes exhibited power conversion efficiencies (PCEs) as high as those reported for ITO-based cells [32,80]. By 2017, the PCE values had been optimized up to 3.74% for the OSCs built on highly flexible substrates with PEDOT:PSS anodes, whose conductivity was optimized through the morphology or conformational modifications of the polymer using effectively combined treatment and deposition methods. These methods included various pre- and post-treatment methods employing a diverse range of chemicals, such as the insertion of metal oxide HTLs between the polymer electrodes and the photoactive layers [23,68,106,107]. As the film surface morphology and chain arrangement of PEDOT:PSS are highly sensitive to the film deposition conditions, the choice of the right technique is crucial to obtain highly conductive and transparent electrodes. Some unique low-cost deposition techniques, such as spray coating and brush painting, have been shown to yield films with remarkably uniform morphologies, which in turn greatly contributed to the PCEs of OSCs when used as transparent electrode layers [108,109]. Y.-J. Noh et al. demonstrated high-performance ITO-free OSCs with Ag NWs/PEDOT:PSS composite electrodes fabricated through a one-step spray-coating method during which the $R_{sh}$, optical transmittance, and thickness of the hybrid electrodes were easily controlled simply by varying the
spray deposition time [110]. These results emphasize the capacity of PEDOT:PSS to replace the brittle and expensive ITO in the future low-cost, easy-to-fabricate, and high-performance plastic devices.

A high $W_f$ is another key parameter defining the performance of the anode material as it is used on the hole collection side of an OSC, and despite the fact that the $W_f$ of PEDOT:PSS is considered high and suitable for such electronic devices as OLEDs, it is relatively lower than what is needed for energy level alignment with the valence band of the active layer in OSCs. This issue, along with PEDOT:PSS’s other shortcomings, severely limits the device efficiency. Fortunately, the $W_f$ of PEDOT:PSS can be increased by simply using organic solvents like alkyl alcohol for the ‘secondary doping’ of the polymer, which enables higher hole extraction rates and leads to enhanced device performance [23].

On the other hand, to fully realize organic and polymer OSCs for more cost-effective and flexible applications, not only ITO-free but also metal-free cells should be achieved, which means that the metal used as a cathode material also has to be replaced with a conductive polymer. The $W_f$ of PEDOT:PSS, however, the most common conductive polymer, is too high to be suitable for electron collection in OSCs, and hence, for the application of PEDOT:PSS as an alternative cathode material in OSCs. Nevertheless, Z. Li et al. suggested a two-step dipping process for PEDOT:PSS treatment to effectively increase its electrical conductivity and easily lower its $W_f$, while also using sulfuric acid and polyethylenimine (PEI) solutions, respectively. As a result, the second dipping in PEI solution aimed to tune the $W_f$ of the polymer reduced it from 4.9 to 4.0 eV, transforming PEDOT:PSS into a low-$W_f$ material while maintaining its high conductivity of 1140 S cm$^{-1}$. These results emphasize the potential of this universal polymer to supersede the conventional rigid conductors of both the top and bottom electrodes in OPVs [24].

Wei et al. also reported all-plastic flexible OSCs using PEDOT:PSS films for both the top and bottom electrodes, whose electrical conductivity was enhanced with different treatment agents. The cells constructed using the PES/H$_3$PO$_4$-treated PEDOT:PSS/PEI/P3HT: ICBA/EG-treated PEDOT:PSS structure exhibited a 3.3% PCE under 100 mW·cm$^{-2}$ white-light illumination, with a high fill factor (FF) of 60% [58]. The device configuration and electrical performance of this flexible cell are shown in Figure 7.

A very recent report suggests the use of solution-processed PEDOT:PSS composites with CNTs or reduced graphene oxide (RGO) as additional conducting electrode layers on the main ITO anodes to significantly enhance the PCEs of OPVs [60]. It should be noted that a similar technique had been suggested by T.-W. Koh et al. to enhance the outcoupling efficiency of OLEDs, by simply coating ITO electrodes with additional highly conductive PEDOT:PSS electrode layers [111]. These patterns are shown and compared in Figure 8.

5.3. PEDOT:PSS-based thin-film electrodes for OTFTs

OTFTs are devices with three electrodes that use organic semiconductors in their active channels. These devices are the basic building blocks for low-cost, thin, lightweight, and flexible electronic applications [112]. A representative example of such applications is the integrated optoelectronic circuit of displays, where every
Figure 8. ITO anodes coated with a highly conductive PEDOT:PSS layer as (a) an additional electrode layer to enhance the PCEs of OSCs [60] and as (b) a low-index layer to enhance the outcoupling efficiency of OLEDs [111].

OLED is driven by an organic field effect transistor (OFET) [113,114]. The device structure and working principles of this transistor are very similar to those of its inorganic counterparts, such as the metal–oxide–semiconductor field effect transistor (MOSFET) and other metal–insulator–semiconductor (MIS) transistors. OTFTs consist of five main elements and one supporting element: a gate electrode (G) to modulate/control the charge carrier concentration in the channel, a source electrode (S) to inject the charge carriers into the channel, a drain electrode (D) to collect/extract the charge carriers from the channel, a gate insulator to separate the G from the channel, an organic semiconductor to provide current flow in the conductive channel, and a substrate to construct and support the whole structure. Depending on the position of the S and D (S/D) electrodes relative to the organic active layer, OTFTs are typically classified as bottom-contact (BC) and top-contact (TC). Further, their structure is defined as top-gate (TG) and bottom-gate (BG) depending on whether the G electrode is disposed on top of the insulating layer or under it. Thus, OTFTs are designed in the four configurations — BCBG, BCTG, TCTG, and TCBG — to assemble two different device geometries: coplanar (BCBG and TCTG) and staggered (BCTG and TCBG) [10,113–115].

PEDOT:PSS has also been suggested as a promising material for the fabrication of G, S, and D polymer electrodes for solution-processed flexible organic transistors like OFETs and OECTs [16,116]. For these devices, the solution processability of PEDOT:PSS is also frequently incorporated with the outstanding electrical and optical properties of Ag NWs, CNTs, and graphene to realize high-performance composite electrodes for OTFTs [64]. It has been empirically confirmed that an Ag NW concentration as low as 0.1 wt% considerably lowers the $R_{sh}$ of PEDOT:PSS S/D electrodes, enabling efficient contact with the active layer from the viewpoint of energy level matching, and resulting in high carrier mobilities of the OTFTs [112]. Besides the incorporation of carbon- or metal-NW-based species into PEDOT:PSS composite electrodes, a number of other common pre- and post-treatment methods are utilized to improve the electrical performance of PEDOT:PSS for OTFT electrode applications. Patterning is the ideal technique of enhancing the device characteristics of organic transistors. Moreover, although solution-based deposition techniques like spin coating or drop casting are currently among the most preferred for organic polymers, micropatterning of the deposited organic layers is a difficult task in the production of the microscale OTFT configurations. Patterning is especially crucial for these organic devices as it considerably enhances the overall performance by preventing the off-state leakage currents and increasing the transconductance [113]. Moreover, despite the fact that solution-processed inkjet printing enables the formation of micropatterns, its resolution is significantly limited compared to that of the conventional photolithographic patterning methods. On the other hand, organic materials are very sensitive to the common photoresist solvents, and get severely damaged during the conventional photolithography. In addition, the highly acidic PEDOT:PSS is especially delicate to be patterned via photolithography as it does not only become damaged itself but also decomposes the used photoresist. Therefore, there is a challenge to develop new lithographic materials compatible with PEDOT:PSS and other organic materials. For this purpose, Taylor et al. synthesized a UV-sensitive and acid-stable polymer photoresist dissolved in nontoxic fluorinated solvents,
hydrofluoroethers (HFEs), which are chemically orthogonal to non-fluorinated organic materials. The novel photoresist facilitated the sub-micrometre patterning of PEDOT:PSS as the S/D electrodes and the organic small molecules of pentacene as the active layer in the fabrication of a BC-OTFT with a high on–off current ratio of $10^4$. It was found that the photoresist is completely processed with HFEs without any extra co-solvent, and that it does not have a destructive impact on the performance or the thickness of PEDOT:PSS films [19]. Another example of patterning similar to the work reported in [87], which was performed on the patterning of PEDOT:PSS OLED

**Figure 9.** a) Schematic steps (above) and digital camera images (below) of PEDOT:PSS patterning with a Cu protection layer: (I) deposition of a PEDOT:PSS film onto the substrate; (II) Cu deposition through a metal shadow mask; (III) oxygen-plasma etching of the unprotected PEDOT:PSS film; (IV) acid etching of the Cu protection layer; and b) transfer and c) output characteristics of the diketopyrrolopyrrole-thieno[3,2-b]thiophene (DPPT-TT)-based OFET with the patterned PEDOT:PSS S/D electrodes [84].

**Figure 10.** a) Schematic demonstration of S/D patterning through spray coating. b) Optical microscopy and scanning electron microscopy (SEM) images of Ag NW/PEDOT:PSS composite S/D electrodes [64].
anodes via the conventional photolithography using an Ag interlayer, was reported by Kostianovskii et al., who employed a much simpler process of oxygen plasma etching instead of photolithography, and a protective layer formed from another noble but less expensive metal, copper (Cu). The patterned PEDOT:PSS films were applied as S/D electrodes for OFETs and yielded a high hole mobility of 0.29 cm²·V⁻¹·s⁻¹ with the contact resistance ($R_c$) values at least 10 times lower than those of gold-based OFETs [84]. Figure 9 summarizes the scheme of this facile patterning technique of PEDOT:PSS electrodes as well as the electrical characteristics of the OFET devices with the patterned polymer bottom electrodes.

Meanwhile, Sanyoto from the research group of Kostianovskii et al. devised a remarkably efficient and solution-processed method for the facile patterning of OTFT electrodes, using a well-known spray-coating technique (see Figure 10). Different types of conductive ink, such as the SWCNT, DMSO-treated PEDOT:PSS, MeOH-treated PEDOT:PSS, and Ag NW/PEDOT:PSS composite ink solutions, were spray-deposited through a metallic shadow mask under ambient conditions, using a commercially available pneumatic spray nozzle, to pattern S/D electrodes [64].

A number of high-performance diketopyrrolopyrrole-thieno[3,2-b]thiophene (DPPT-TT)-based OFETs with polymer and composite electrodes spray-patterned on rigid and flexible substrates have been successfully demonstrated [64]. Figure 11 shows the transfer characteristics of the fabricated transistors.

Another approach to patterning PEDOT:PSS as an ordered thin-film structure for application in organic electronic devices suggests the use of self-assembled monolayers (SAMs) for the modification of the surface
free energy and morphology of the polymer to effectively adjust its $W_f$ for better energy level matching between the polymer electrodes and the active layers of the devices [117,118].

6. Summary

In this review, the recent progress achieved in the field of organic electronics using the poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) conductive polymer was discussed. A multitude of experiment methods defined as pre- and post-treatment methods [119] aiming to enhance the electrical conductivity of the polymer and the theory behind them were presented in depth. Special focus was given to the application of PEDOT:PSS as an electrode material in organic devices due to the increasing demand for flexible, transparent, wearable, and biodegradable device applications. From this viewpoint, PEDOT:PSS is among the materials potentially suitable for use in the fabrication of non-metallic and non-metal–oxide-based electrodes for a wide variety of electronic components. Hence, the realization of polymer electrodes will pave the way for the future design of an astonishing diversity of soft electronics. Notwithstanding the poor performance of PEDOT:PSS, it is intensively being investigated, and comprehensive empirical and theoretical studies are being conducted worldwide for the enhancement of its electrical conductivity to meet the state-of-the-art requirements put forward for flexible and transparent electrode materials. There is no doubt that the prospective research and engineering resources combined with the existing techniques will enable this target to be met.

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References

[1] K. Samuel, N.M. Jizat, N. Ahmad, L.Y. Chiang, and M.I. Sabran, A Low-Cost Thin Film Flexible Plastic Graphene-Conductive Polymer Composite Antenna, AIP Conf. Proc 2030, 020003 (2018).
[2] R. Cherrington, J. Liang, Design and Manufacture of Plastic Components for Multifunctionality: 2 - Materials and Deposition Processes for Multifunctionality, Structural Composites, Injection Molding, and 3D Printing 2016, pp. 19–51.
[3] K. Deshmukh, M. B. Ahamed, R. R. Deshmukh, S. K. K. Pasha, P. R. Bhagat, K. Chidambaram, Biopolymer Composites in Electronics: 3 - Biopolymer Composites with High Dielectric Performance: Interface Engineering, 2017, pp. 27–128.
[4] T.-H. Le, Y. Kim, and H. Yoon, Electrical and Electrochemical Properties of Conducting Polymers, Polymers 9, 150 (2017).

[5] N. Hall, Twenty-five years of conducting polymers, Chem. Commun 7, 1–4 (2003).

[6] N. Yi, M. R. Abidian, Biosynthetic Polymers for Medical Applications: 10 - Conducting Polymers and Their Biomedical Applications, Woodhead Publishing Series in Biomaterials, pp. 243–276, 2016.

[7] Y.V. Yakhmi, V. Saxena, and D.K. Aswal, Functional Materials: 2 - Conducting Polymer Sensors, Actuators and Field-Effect Transistors, Preparation, Processing and Applications, 61–110 (2012).

[8] G. Kaur, R. Adhikari, P. Cass, M. Brown, and P. Gunatilake, Electrically Conducting Polymers and Composites for Biomedical Applications, RSC Adv. 5, 37553 (2015).

[9] J. Lee, P. Lee, H.B. Lee, S. Hong, I. Lee, J. Yeo, S.S. Lee, T.-S. Kim, D. Lee, and S.H. Ko, Room-Temperature Nanosoldering of a Very Long Metal Nanowire Network by Conducting-Polymer-Assisted Joining for a Flexible Touch-Panel Application, Adv. Funct. Mater 23, 4171–4176 (2013).

[10] D. Fichou, G. Horowitz, Molecular and Polymer Semiconductors, Conductors, and Superconductors: Overview, Encyclopaedia of Materials: Science and Technology (Second Edition), pp. 5748–5757, 2001.

[11] J.H. Kim, C.W. Joo, J.Lee, Y.K. Seo, J.W. Han, J.Y. Oh, J.S. Kim, S. Yu, J.H. Lee, J.-I. Lee, C. Yun, B.H. Choi, and Y.H. Kim, Highly Conductive PEDOT:PSS Films with 1,3-Dimethyl-2-imidazolidinone as Transparent Electrodes for Organic Light-Emitting Diodes, Macromol. Rapid Commun 37, 1427–1433 (2016).

[12] L.V. Kayser, and D.J. Lipomi, Stretchable Conductive Polymers and Composites Based on PEDOT and PEDOT:PSS, Adv. Mater 31, 1806133 (2019).

[13] K. Sun, S. Zhang, P. Li, Y. Xia, X. Zhang, D. Du, F.H. Isikgor, and J. Ouyang, Review on Application on PEDOTs and PEDOT:PSS in Energy Conversion and Storage Devices, J. Mater. Sci: Mater. Electron 26 (7), 4438–4462 (2015).

[14] A. Soleimani-Gorgani, Co-Solvents Roles in PEDOT: PSS Ink-Jet Inks, Adv. Nat. Sci.: Nanosci. Nanotechnol 9, 025009 (2018).

[15] T.-W. Lee, and Y. Chung, Control of the Surface Composition of a Conducting-Polymer Complex Film to Tune the Work Function, Adv. Funct. Mater 18, 2246–2252 (2008).

[16] X. Fan, W. Nie, H. Tsai, N. Wang, H. Huang, Y. Cheng, R. Wen, L. Ma, F. Yan, and Y. Xia, PEDOT:PSS for Flexible and Stretchable Electronics: Modifications, Strategies, and Applications, Adv. Sci 1900813 (2019).

[17] K.M. Reza, S. Mabrouk, and Q. Qiao, A Review on Tailoring PEDOT:PSS Layer for Improved Performance of Perovskite Solar Cells, Proc. Nat. Res. Soc 2, 02004 (2018).

[18] Q. Wei, M. Mukaida, K. Kiriha, Y. Naitoh, and T. Ishida, Recent Progress on PEDOT-Based Thermoelectric Materials, Mater 8, 732–750 (2015).

[19] P.G. Taylor, J.-K. Lee, A.A. Zakhidov, M. Chatzichristidi, H.H. Fong, J.A. DeFranco, G.G. Malliaras, and C.K. Ober, Orthogonal Patterning of PEDOT:PSS for Organic Electronics using Hydrofluoroether Solvents, Adv. Mater 21, 2314–2317 (2009).

[20] B. Park, C.H. Park, Y. Yim, and J. Park, Electrical Annealing for Flexible Organic Light-Emitting Diods Having Poly(3, 4-ethylenedioxythiophene):Poly(Styrene Sulfonate) Anodes, J. Appl. Phys 108, 084508 (2010).

[21] M.S. White, M. Kaltenbrunner, E.D. Glowacki, K. Gutnicken, G. Kettlgruber, I. Graz, S. Aazou, C. Ulbricht, D.A.M. Egbe, M.C. Miron, Z. Major, M.C. Scharber, T. Sekitani, T. Someya, S. Bauer, and N.S. Sariciftci, Ultrathin, Highly Flexible and Stretchable PLEDs, Nat. Photonics 7, 811–816 (2013).

[22] A. Benor, S.-Y. Takizawa, C. Perez-Bolivar, and P. Anzenbacher, Jr, Efficiency Improvement of Fluorescent OLEDs by Tuning the Working Function of PEDOT:PSS Using UV-Ozone Exposure, Org. Electron 11, 938–945 (2010).

[23] J. Saghaei, A. Fallahzadeh, and T. Saghaei, ITO-Free Organic Solar Cells Using Highly Conductive Phenol-Treated PEDOT:PSS Anodes, Org. Electron 24 (9@015), 188–194.

[24] Z. Li, Y. Liang, Z. Zhong, J. Qian, G. Liang, K. Zhao, H. Shi, S. Zhong, Y. Yin, and W. Tian, A Low-Work-Function, High-Conductivity PEDOT:PSS Electrode for Organic Solar Cells with a Simple Structure, Synth. Met 210, 363–366 (2015).

[25] T. Nagata, S. Oh, T. Chikyow, and Y. Wakayama, Effect of UV-Ozone Treatment on Electrical Properties of PEDOT:PSS Film, Org. Electron 12, 279–284 (2011).

[26] D. Alemu, H.-Y. Wei, K.-C. Ho, and C.-W. Chu, Highly Conductive PEDOT:PSS Electrode by Simple Film Treatment with Methanol for ITO-Free Polymer Solar Cells, Energy Environ. Sci 5, 9662–9671 (2012).

[27] Z. Yu, Y. Xia, D. Du, and J. Ouyang, PEDOT:PSS Films with Metallic Conductivity through a Treatment with Common Organic Solutions of Organic Salts and Their Application as a Transparent Electrode of Polymer Solar Cells, ACS Appl. Mater. Interfaces 8, 11629–11638 (2016).

[28] J. Ouyang, “Secondary Doping” Methods to Significantly Enhance the Conductivity of PEDOT:PSS for its Application as Transparent Electrode of Optoelectronic Devices, Displays 34, 423–436 (2013).

[29] C. Badre, L. Marquant, A.M. Alsayed, and L.A. Hough, Highly Conductive Poly(3, 4-Ethylenedioxythiophene): Poly(Styrenesulfonate) Films Using 1-Ethyl-3-Methylimidazolium Tetracyanoborate Ionic Liquid, Adv. Funct. Mater 22 (13), 2723–2727 (2012).

[30] A. Keawprajak, W. Koetniyom, P. Piyakulawat, K. Jiramitmongkon, S. Pratontep, and U. Asawapirrom, Effects of Tetramethylene Sulfone Solvent Additives on Conductivity of PEDOT:PSS Film and Performance of Polymer Photovoltaic Cells, Org. Electron 14, 402–410 (2013).

[31] T.-R. Chou, S.-H. Chen, Y.-T. Chiang, T.-T. Chang, C.-W. Lin, and C.-Y. Chao, Highly Conductive PEDOT:PSS Film by Doping P-Toluenesulfonic Acid and Post-Treatment with Dimethyl Sulfoxide for ITO-Free Polymer Dispersed Liquid Crystal Device, Org. Electron 48, 223–229 (2017).

[32] Y.H. Kim, C. Sachse, M.L. Machala, C. May, L. Müller-Meskamp, and K. Leo, Highly Conductive PEDOT:PSS
Electrode with Optimized Solvent and Thermal Post-Treatment for ITO-Free Organic Solar Cells, Adv. Funct. Mater. 21, 1076–1081 (2011). [33] M. Cai, Z. Ye, T. Xiao, R. Liu, Y. Chen, R.W. Mayer, R. Biswas, K.-M. Ho, R. Shinar, and J. Shinar, Extremely Efficient Indium–Tin-Oxide-Free Green Phosphorescent Organic Light-Emitting Diodes, Adv. Mater. 24, 1–6 (2012). [34] Y.K. Seo, C.W. Joo, J. Lee, J.W. Han, N.S. Cho, K.T. Lim, S. Yu, M.H. Kang, C. Yun, B.H. Choi, and Y.H. Kim, Efficient ITO-Free Organic Light-Emitting Diodes Comprising PEDOT:PSS Transparent Electrodes Optimized with 2-Ethoxyethanol and Post Treatment, Org. Electron 42, 348–354 (2017). [35] J. Gasirwoski, R. Menon, K. Hingerl, M. Dachev, and N.S. Sariciftci, Surface Morphology, Optical Properties and Conductivity Changes of Poly(3,4-Ethylenedioxythiophene):Poly[(Styrenesulphonate) by Using Additives, Thin Solid Films 536, 211–215 (2013). [36] J.Y. Kim, J.H. Jung, D.E. Lee, and J. Joo, Enhancement of Electrical Conductivity of Poly(3, 4-Ethylenedioxythiophene)/Poly(4-Styrenesulphonate) by a Change of Solvents, Synt. Met 126, 311–316 (2002). [37] K. Fehse, K. Walzer, K. Leo, W. Lövenich, and A. Elschner, Highly Conductive Polymer Anodes as Replacememts for Inorganic Materials in High-Efficiency Organic Light-Emitting Diodes, Adv. Mater. 19, 441–444 (2007). [38] A.K. Havare, M. Can, S. Demic, M. Kus, and S. Icli, The Performance of OLEDs Based on Sorbitol Doped PEDOT:PSS, Synth. Met 161, 2734–2738 (2012). [39] A.M. Nardes, R.A.J. Janssen, and M. Kemerink, A Morphological Model for the Solvent-Enhanced Conductivity of PEDOT:PSS Thin Films, Adv. Funct. Mater. 18, 865–871 (2008). [40] P. Sakunpongpitiporn, K. Phasuksom, N. Paradeeng, and A. Sirivat, Facile Synthesis of Highly Conductive PEDOT:PSS via Surfactant Templates, RSC Adv. 9, 6363–6378 (2019). [41] Q. Wei, M. Mukaida, Y. Naitoh, and T. Ishida, Morphological Change and Mobility Enhancement in PEDOT: PSS by Adding Co-solvents, Adv. Mater. 25, 2831–2836 (2013). [42] Z. Yi, Y. Zhao, P. Li, K. Ho, N. Blozowski, G. Walker, S. Jaffer, J. Tjong, M. Sain, and Z. Lu, The Effect of Tannic Acids on the Electrical Conductivity of PEDOT:PSS Films, Appl. Surf. Sci 448, 583–588 (2018). [43] C.C. Han, and R.L. Eisenbaumer, Protonic Acids: Generally Applicable Dopants for Conducting Polymers, Synth. Met 30 (1), 123–131 (1989). [44] F. Wu, P. Li, K. Sun, Y. Zhou, W. Chen, J. Fu, M. Li, S. Lu, D. Wei, X. Tang, Z. Zang, L. Sun, X. Liu, and J. Ouyang, Conductivity Enhancement of PEDOT:PSS via Addition of Chloroplatanic Acid and Its Mechanism, Adv. Electron. Mater. 3 (7), 1700047 (2017). [45] C. Wang, K. Sun, J. Fu, R. Chen, M. Li, Z. Zang, X. Liu, B. Li, H. Gong, and J. Ouyang, Enhancement of Conductivity and Thermoelectric Property of PEDOT:PSS via Acid Doping and Single Post-Treatment for Flexible Power Generator, Adv. Sustain. Syst 2, 1800085 (2018). [46] S. Kim, B. Sanyoto, W.-T. Park, S. Kim, S. Mandal, J.-C. Lim, Y.-Y. Noh, and J.-H. Kim, Purification of PEDOT:PSS by Ultrafiltration for Highly Conductive Transparent Electrode of All-Printed Organic Devices, Adv. Mater. 28, 10149–10154 (2016). [47] J. Saghaei, A. Fallahzadeh, and M.H. Yousefi, Improvement of Electrical Conductivity of PEDOT:PSS Films by 2-Methylimidazole Post-Treatment, Org. Electron 19, 70–75 (2015). [48] W. Wei, H. Wang, and Y.H. Hu, A Review on PEDOT-Based Counter Electrodes for Dye-Sensitized Solar Cells, Int. J. Energy Res 38, 1099–1111 (2014). [49] Z. Fan, D. Du, Z. Yu, P. Li, Y. Xia, and J. Ouyang, Significant Enhancement in the Thermoelectric Properties of PEDOT:PSS Films through a Treatment with Organic Solutions of Inorganic Salts, ACS Appl. Mater. Interfaces 8, 23204–23211 (2016). [50] Y. Xia, and J. Ouyang, Anion Effect on Salt-Induced Conductivity Enhancement of Poly(3,4-Ethylenedioxythiophene):Poly[(Styrenesulphonate) Films, Org. Electron 11, 1129–1135 (2010). [51] S. He, M. Mukaida, K. Kirihara, L. Lyu, and Q. Wei, Reversible Protonic Doping in Poly(3,4-Ethylenedioxythiophene), Polymers 10, 1065 (2018). [52] C. Song, Z. Zong, Z. Hu, Y. Luo, L. Wang, J. Wang, and Y. Cao, The Effect of Solvent Treatment on the Buried PEDOT:PSS Layer, Org. Electron 43, 9–14 (2017). [53] J-S. Yeo, J-M. Yun, D-Y. Kim, S. Park, S-S. Kim, M-H. Yoon, T-W. Kim, and S-I. Na, Significant Vertical Phase Separation in Solvent–Vapor-Annealed Poly(3,4-ethylenedioxythiophene):Poly[(styrene sulfonate)] Composite Films Leading to Better Conductivity and Work Function for High-Performance Indium Tin Oxide-Free Optoelectronics, ACS Appl. Mater. Interfaces 4 (5), 2551–2560 (2012). [54] O.P. Dimitriev, D.A. Grinko, Y.V. Noskov, N.A. Ogurtsov, and A.A. Pud, PEDOT:PSS Films – Effect of Organic Solvent Additives and Annealing on the Film Conductivity, Synth. Met 159, 2237–2239 (2009). [55] X. Zhang, J. Wua, J. Wang, J. Zhang, Q. Yang, Y. Fua, and Z. Xie, Highly Conductive PEDOT:PSS Transparent Electrode Prepared by a Post-Spin-Rinsing Method for Efficient ITO-Free Polymer Solar Cells, Sol. Energy Mater. Sol. Cells 144, 143–149 (2016). [56] E.M. Tarmidzi, and S.B. Sasongko, Highly Conductive PEDOT: PSS Flexible Film with Secondary Doping and Spray Pyrolysis Method, IJAER 13 (12), 10234–10239 (2018). [57] Y. Xia, K. Sun, and J. Ouyang, Solution-Processed Metal- Lithic Conducting Polymer Films as Transparent Electrode of Optoelectronic Devices, Adv. Mater. 24, 2436–2440 (2012). [58] W. Meng, R. Ge, Z. Li, J. Tong, T. Liu, Q. Zhao, S. Xiong, F. Jiang, L. Mao, and Y. Zhou, Conductivity Enhancement of PEDOT:PSS Films via Phosphoric Acid Treatment for Flexible All-Plastic Solar Cells, ACS Appl. Mater. Interfaces 7, 14089–14094 (2015). [59] S. Zhang, Y. Xia, and J. Ouyang, Effect of Water-Soluble Vitamins on the Structure and Properties of Poly(3,4- ethylenedioxythiophene):Poly[(Styrenesulphonate), Org. Electron 45, 139–144 (2017). [60] B.V.R.S. Subramanam, P.C. Mahakul, K. Sa, J. Raiguru, I. Alam, S. Das, M. Mondal, S. Subudhi, and P. Mahanandia, Improved Stability and Performance of
Organic Photovoltaic Cells by Application of Carbon Nanostructures and PEDOT:PSS Composites as Additional Transparent Electrodes, Sol. Energy 186, 146–155 (2019).

[61] Y.-F. Liu, J. Feng, Y.-F. Zhang, H.-F. Cui, D. Yin, Y.-G. bi, J.-F. Song, Q.-D. Chen, and H.-B. Sun, Improved Efficiency of Indium-Tin-Oxide-Free Organic Light-Emitting Devices Using PEDOT:PSS/Graphene Oxide Composite Anode, Org. Electron 26, 81–85 (2015).

[62] Y.S. Liu, J. Feng, X.-L. Ou, H.-F. Cui, M. Xu, and H.-B. Sun, Ultrasmooth, Highly Conductive and Transparent PEDOT:PSS/Silver Nanowire Composite Electrode for Flexible Organic Light-Emitting Devices, Org. Electron 31, 247–252 (2016).

[63] B. Wei, X. Wu, L. Lian, S. Yang, D. Dong, D. Feng, and G. He, A Highly Conductive and Smooth Ag NW/PEDOT:PSS Film Treated by Hot-Pressing as Electrode for Organic Light Emitting Diode, Org. Electron 43, 182–188 (2017).

[64] B. Sanyoto, S. Kim, W.-T. Park, Y. Xu, J.-H. Kim, J.-C. Lim, and Y.-Y. Noh, Solution Processable PEDOT:PSS Based Hybrid Electrodes for Organic Field Effect Transistors, Org. Electron 37, 352–357 (2016).

[65] Y.H. Kim, J. Lee, S. Hofmann, M.C. Gather, L. Müller-Meskamp, and K. Leo, Achieving High Efficiency and Improved Stability in ITO-Free Transparent Organic Light-Emitting Diodes with Conductive Polymer Electrodes, Adv. Funct. Mater 23, 3763–3769 (2013).

[66] K. Fehse, R. Meerheim, K. Walzer, K. Leo, W. Lövenich, and A. Elschner, Lifetime of Organic Light Emitting Diodes on Polymer Anodes, Appl. Phys. Lett 93, 083303 (2008).

[67] D. Bryant, I. Mabbett, P. Greenwood, T. Watson, M. Wijdekop, and D. Worsley, Ultrafast Near-Infrared Curing of PEDOT:PSS, Org. Electron 15, 1126–1130 (2014).

[68] S.H. Chang, C.-H. Chiang, F.-S. Kao, C.-L. Tien, and C.-G. Wu, Unraveling the Enhanced Electrical Conductivity of PEDOT:PSS Thin Films for ITO-Free Organic Photovoltaics, IEEE Photonics J 6 (4), (2014).

[69] N.K. Unsworth, I. Hancox, C.A. Dearden, T. Howells, P. Sullivan, R.S. Lilley, J. Sharp, and T.S. Jones, Conduction Spray Deposited Poly(3, 4-ethylenedioxythiophene):Poly(styrenesulfonate) Electrodes for Indium Tin Oxide-Free Small Molecule Organic Photovoltaic Devices, Appl. Phys. Lett 103, 173304 (2013).

[70] B. Andò, S. Baglio, A.R. Bulsara, T. Emery, V. Marletta, and A. Pistorio, Low-Cost Inkjet Printing Technology for the Rapid Prototyping of Transducers, Sensors 17, 748 (2017).

[71] G. Cummins, and M. Desmulliez, Inkjet Printing of Conductive Materials: A Review, Circuit World 38 (4), 193–213 (2012).

[72] J. Alamán, R. Alicante, J.I. Peña, and C. Sánchez-Somolinos, Inkjet Printing of Functional Materials for Opticaland Photonic Applications, Mater 9, 910 (2016).

[73] S. Ummartyotin, J. Juntaro, C. Wu, M. Sain, and H. Manuspiya, Deposition of PEDOT: PSS Nanoparticles as a Conductive Microlayer Anode in OLEDs Device by Desktop Inkjet Printer, J. Nanomater 606714 (2011).

[74] E. Timothée, T. Roberts, M. Saaadoui, T. Hervé, J.B. De Graaf, and G.G. Malliaras, Inkjet-Printed PEDOT:PSS Electrodes on Paper for Electrocardiography, Adv. Healthcare Mater 6 (6), 1601167.

[75] S. Bai, Y. Ma, X. Jiang, Q. Li, Z. Yang, Q. Liu, and D. He, Greatly Improved Cyclability for Li-Ion Batteries with a PEDOT–PSS Coated Nanostructured Ge Anode, Surf. Interfac 8, 214–218 (2017).

[76] P.R. Das, L. Komsisyska, O. Osters, and G. Wittstock, PEDOT: PSS as a Functional Binder for Cathodes in Lithium Ion Batteries, J. Electrochem. Soc 162 (4), A674–A678 (2015).

[77] D.H. Yoon, S.H. Yoon, K.-S. Ryu, and YJ. Park, PEDOT:PSS as Multi-Functional Composite Material for Enhanced Li-Air-Battery Air Electrodes, Scientific Reports 6, 19962 (2016).

[78] T.-H. Han, S.-H. Jeong, Y. Lee, H.-K. Seo, S.-J. Kwon, M.-H. Park, and T.-W. Lee, Flexible Transparent Electrodes for Organic Light-Emitting Diodes, J. Inform. Display 16 (2), (2015).

[79] T.-B. Song, and N. Li, Emerging Transparent Conducting Electrodes for Organic Light Emitting Diodes, Electronics 3 (1), 190–204 (2014).

[80] J. Meiss, C.L. Uhrich, K. Fehe, S. Pfuetzner, M. K. Riede, K. Leo, Transparent Electrode Materials for Solar Cells, Proc. of SPIE, 7002, 700210-1 (2008).

[81] S. Günes (2011) Organic Solar Cells and Their Nanosctructural Improvement. In Energy Efficiency and Renewable Energy Through Nanotechnology. Green Energy and Technology, edited by Zang L. (Springer, London).

[82] Y.-H. Huang, C.-Y. Lu, S.-T. Tsai, Y.-T. Tsai, C.-Y. Chen, W.-L. Tsai, C.-Y. Lin, H.-W. Chang, W.-K. Lee, M. Jiao, and C.-C. Wu, Enhancing Light Out-Coupling of Organic Light-Emitting Devices Using Indium Tin Oxide-Free Low-Index Transparent Electrodes, Appl. Phys. Lett 104, 183302 (2014).

[83] Y. Li, C. Liu, S. Tong, L. Pan, L. Pu, T. Minari, K. Tsukagoshi, and Y. Shi, Metal-Diffusio-Induced ITO Nanoparticles at the Organic/ITO Interface, J. Phys. D: Appl. Phys 45, 165104 (2012).

[84] V. Kostianovskii, B. Sanyoto, and Y.-Y. Noh, A Facile Way to Pattern PEDOT:PSS Film as an Electrode for Organic Devices, Org. Electron 44, 99–105 (2017).

[85] M.-K. Wei, C.-W. Lin, C.-C. Yang, Y.-W. Kiang, J.-H. Lee, and H.-Y. Lin, Emission Characteristics of Organic Light-Emitting Diodes and Organic Thin-Films with Planar and Corrugated Structures, Int. J. Mol. Sci 11, 1527–1545 (2010).

[86] A. Kim, G. Huseynova, J. Lee, and J.-H. Lee, Enhancement of Out-Coupling Efficiency of Flexible Organic Light-Emitting Diodes Fabricated on an MLA-Patterned Parylene Substrate, Org. Electron 71, 246–250 (2019).

[87] C.W. Joo, G. Huseynova, J. Yifei, J.-M. Yood, Y.H. Kim, N.S. Choa, J.-H. Lee, Y.-H. Kim, and J. Lee, Highly Efficient Solution-processed Blue Organic Light-Emitting Diodes Based on Thermally Activated Delayed Fluorescence Emitters with Spiroacridine Donor, J. Ind. Eng. Chem 78, 265–270 (2019).

[88] T. Bockshrocker, F. Maier-Flaig, C. Eschenbaum, and U. Lemmer, Efficient Waveguide Mode Extraction in White Organic Light Emitting Diodes Using ITO-Anodes with Integrated MgF2-Columns, Opt. Express 20 (6), 6100–6107 (2012).
[89] G. Gaertner, H. Greiner, Light Extraction from OLEDs with (High) Index Matched Glass Substrates, Proceedings 6999, Organic Optoelectronics and Photonics III; 69992 T (2008).

[90] S.S. Jeong, and J.-H. Ko, Simulation Study on the Optical Structures for Improving the Outcoupling Efficiency of Organic Light-Emitting Diodes, J. Inform. Display 13 (4), (2012).

[91] M. Cai, T. Xiao, R. Liu, Y. Chen, R. Shinar, and J. Shinar, Indium-Tin-Oxide-Free Tris(8-Hydroxyquinoline) Al Organic Light-Emitting Diodes with 80% Enhanced Power Efficiency, Appl. Phys. Lett 99, 153303 (2011).

[92] Y.H. Kim, J. Lee, W.M. Kim, C. Fuchs, S. Hofmann, H.-W. Chang, M.C. Gather, L. Müller-Meskamp, and K. Leo, We Want Our Photons Back: Simple Nanostructures for White Organic Light-Emitting Diode Outcoupling, Adv. Funct. Mater 24, 2553–2559 (2014).

[93] A. Fallahazardeh, J. Saghaei, and M.H. Yousefi, Effect of Alcohol Vapor Treatment on Electrical and Optical Properties of Poly(3, 4-Ethylene Dioxithiophene):Poly (Styrene Sulfonate) Films for Indium Tin Oxide-Free Organic Light Emitting Diodes, Appl. Surf. Sci 320, 895–900 (2014).

[94] X. Wu, J. Liu, and G. He, A Highly Conductive PEDOT:PSS Film with the Dipping Treatment by Hydroiodic Acid as Anode for Organic Light Emitting Diode, Org. Electron 22, 160–165 (2015).

[95] J. Lee, and Y.H. Kim, High Performance ITO-Free White Organic Light-Emitting Diodes Using Highly Conductive PEDOT:PSS Transparent Electrodes, Synth. Met 242, 99–102 (2018).

[96] Y.K. Seo, C.W. Joo, J. Lee, J.W. Han, D.J. Lee, S.A.N. Entifar, S. Kim, N.S. Cho, and Y.H. Kim, Enhanced Electrical Properties of PEDOT:PSS Films Using Solvent Treatment and Its Application to ITO-Free Organic Light-Emitting Diodes, J. Lumin 187, 221–226 (2017).

[97] H. Kim, S. Kwon, S. Choi, and K.C. Choi, Solution-Processed Bottom-Emitting Polymer Light-Emitting Diodes on a Textile Substrate Towards a Wearable Display, J. Inf. Disp. 16 (4), 179–184.

[98] P.A. Levermore, R. Jin, X. Wang, L. Chen, D.D.C. Bradley, and J.C. de Mello, High Efficiency Organic Light-Emitting Diodes with PEDOT-Based Conducting Polymer Anodes, J. Mater. Chem B 18, 4414–4420 (2008).

[99] B. Riedel, J. Hauss, U. Geyer, J. Guettel, U. Lemmer, and M. Gerken, Enhancing Outcoupling Efficiency of Indium-Tin-Oxide_free Organic Light-Emitting Diodes via Nanostructured High Index Layers, Appl. Phys. Lett 96, 243302 (2010).

[100] S. Ouyang, Y. Xie, D. Zhu, X. Xu, D. Wang, T. Tan, and H.H. Fong, Photolithographic Patterning of PEDOT:PSS with a Silver Interlayer and Its Application in Organic Light Emitting Diodes, Org. Electron 15, 1822–1827 (2014).

[101] H.J. Lee, T.H. Park, J.H. Choi, E.H. Song, S.J. Shin, H. Kim, K.C. Choi, Y.W. Park, and B.-K. Ju, Negative Mold Transfer Patterned Conductive Polymer Electrode for Flexible Organic Light-Emitting Diodes, Org. Electron 14, 416–422 (2013).

[102] Y.-F. Liu, J. Feng, Y.-F. Zhang, H.-F. Cui, D. Yin, Y.-G. Bi, J.-F. Song, Q.-D. Chen, and H.-B. Sun, Improved Efficiency of Indium-Tin-Oxide-Free Flexible Organic Light-Emitting Devices, Org. Electron 15, 478–483 (2014).

[103] S.C. Bhatia, 5 – Solar Photovoltaic Systems, Advanced Renewable Energy Systems 2914, 144–157.

[104] Y. Wu, T. Currier, Y. Li, S.-T. Tsai, Organic Light Emitting Diode and Organic Solar Cell Stack, United States Patent, 2013, Patent No. US8399898B2.

[105] S. Lu, Y. Sun, K. Ren, K. Liu, Z. Wang, and S. Qu, Recent Development in ITO-Free Polymer Solar Cells, Polymers 10 (1), 5 (2018).

[106] M.R. Lenze, N.M. Kronenberg, F. Wührer, and K. Meehrholz, In-Situ Modification of PEDOT:PSS Work Function Using Alkyl Alcohols as Secondary Processing Solvents and Their Impact on Merocyanine Based Bulk Heterojunction Solar Cells, Org. Electron 21, 171–176 (2015).

[107] D.-Y. Lee, S.-P. Cho, S.-I. Na, and S.-S. Kim, ITO-Free Polymer Solar Cells with Vanadium Oxide Hole Transport Layer, J. Ind. Eng. Chem 45, 1–4 (2017).

[108] J.G. Tait, B.J. Worfolk, S.A. Maloney, T.C. Hauger, A.L. Elias, J.M. Buriai, and K.D. Harris, Spray Coated High-Conductivity PEDOT:PSS Transparent Electrodes for Stretchable and Mechanically-Robust Organic Solar Cells, Sol. Energy Mater. Sol. Cells 110, 98–106 (2013).

[109] S.-S. Kim, S.-P. Cho, and S.-I. Na, Two-Step Brush-Painted PEDOT:PSS Electrodes for ITO-Free Organic Solar Cells, J. Ind. Eng. Chem 62, 40–45 (2018).

[110] Y.-J. Noh, S.-S. Kim, T.-W. Kim, and S.-I. Na, Cost-Effective ITO-Free Organic Solar Cells with Silver Nanowire-PEDOT:PSS Composite Electrodes via a One-Step Spray Deposition Method, Sol. Energy Mater. Sol. Cells 120, 226–230 (2014).

[111] T.-W. Koh, J.-M. Choi, S. Lee, and S. Yoo, Optical Outcoupling Enhancement in Organic Light-Emitting Diodes: Highly Conductive Polymer as a Low-Index Layer on Microstructured ITO Electrodes, Adv. Mater 22, 1849–1853 (2010).

[112] L.-H. Chen, P. Lin, M.-C. Chen, P.-Y. Huang, C. Kim, J.-C. Ho, and C.-C. Lee, Silver Nanowire-Polymer Composite Electrode for High-Performance Solution-Processed Thin-Film Transistors, Org. Electron 13, 1881–1886 (2012).

[113] C. Reese, M. Roberts, M.-M. Ling, and Z. Bao, Organic Thin Film Transistors, Mater. Today 7 (9), 20–27 (2004).

[114] H. Siringhaus, 25th Anniversary Article: Organic Field-Effect Transistors: The Path Beyond Amorphous Silicon, Adv. Mater 26 (9), 1319–1335 (2014).

[115] X. Tao, V. Koncar, 25-Textile Electronic Circuits Based on Organic Fibrous Transistors, Smart Textiles and Their Applications, Woodhead Publishing Series in Textiles, pp. 569–598, 2016.

[116] I. Gualandi, M. Marzocchi, E. Scavetta, M. Calienni, A. Bonfiglioc, and B. Fraboni, A Simple All-PEDOT:PSS Electrochemical Transistor for Ascorbic Acid Sensing, J. Mater. Chem. B 3, 6753–6762 (2015).

[117] D. Ohayon, C. Pitsalidis, A.-M. Pappa, A. Hama, Y. Zhang, L. Gallais, and R.M. Owens, Laser Patternning of Self-Assembled Monolayers on PEDOT:PSS Films for Controlled Cell Adhesion, Adv. Mater. Interfaces 1700191 (2017).
[118] P. Dabczynski, M.M. Marzec, L. Pieta, K. Fijalkowski, J. Raczkowska, A. Bernasik, A. Budkowski, and J. Rysz, Engineering a Poly(3,4-ethylenedioxythiophene):(Poly styreneSulfonate) Surface Using Self-Assembling Molecules A ChemicalLibrary Approach, ACS Omega 3, 3631–3639 (2018).

[119] Z. Zhu, C. Liu, F. Jiang, J. Xu, and E. Liu, Effective Treatment Methods on PEDOT:PSS To Enhance Its Thermoelectric Performance, Synth. Met 225, 31–40 (2017).