

Polymer Solar Cells Based on PEDOT:PSS Electrodes

Subjects: **Materials Science, Coatings & Films**

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Solution-processed polymer solar cells (PSCs) have advantages of low cost, solution processability, light weight, and excellent flexibility. Recent progress in materials synthesis and devices engineering has boosted the power conversion efficiency (PCE) of single-junction PSCs over 17%. Poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) is one of the most promising candidates for electrodes due to its high conductivity ($>4000\text{ S/cm}$), excellent transmittance ($>90\%$), intrinsically high work function ($WF > 5.0\text{ eV}$), and aqueous solution processability. To date, a great number of single-junction PSCs based on PEDOT:PSS electrodes have realized a PCE over 12%.

polymer solar cells

flexible electrodes

PEDOT:PSS

electrical conductivity

1. Introduction

Solution-processed polymer solar cells (PSCs) have attracted dramatically increasing attention over the past few decades owing to their advantages of low cost, easy fabrication, light weight, and good flexibility. Recent achievements in materials development and devices engineering have produced a record power conversion efficiency (PCE) over 17% ^[1], demonstrating an anticipated application prospect. However, a high-quality electrode is another challenge for high performance PSCs, especially the flexible electrode for the corresponding flexible devices.

It is well known that a typical PSC configuration consists of a light absorption layer sandwiched between two electrodes (the top one and the bottom one). At least one of them has to be transparent for light illumination through and then to the light absorption layer. Device performance of PSCs is highly dependent on the quality of the electrodes, and a stricter requirement is necessary in the case of flexible electrodes. A high-quality flexible electrode should possess characteristics such as superior mechanical flexibility, low sheet resistance, high transmittance in the visible-light range, and excellent thermal stability. Among current candidates, indium-tin-oxide (ITO) and vacuum-deposited metals-based flexible electrodes have been commercially used in PSCs. However, high production costs and especially poor mechanical stability strongly hinder their practical applications ^[2]. Therefore, many scientific communities are searching other candidates including silver nanowires (AgNWs), silver-grids, conductive polymers (Poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate); PEDOT:PSS), and carbon-based materials, etc.

The commercially available PEDOT:PSS is one of the best choices for flexible electrodes owing to its high transmittance in the visible range, high and adjustable conductivity, intrinsically high work function, excellent thermal stability, and good film-forming capability as well as superior mechanical flexibility. PEDOT:PSS (**Figure 1**) is composed of positively charged conjugated PEDOT and negatively charged saturated PSS. Its aqueous dispersion can be processed to form a thin film on either rigid or flexible substrates by various solution-processing techniques. The achieved PEDOT:PSS film is smooth and ideal for the application of electrodes in electronics. PEDOT:PSS exhibits a wide range of conductivities from 10^{-4} to 10^3 S/cm⁻¹, determined by synthetic conditions, doping additives, or post-treatment methods. PEDOT:PSS film also possesses a high work function of 5.0–5.2 eV, matching well with the highest occupied molecular orbital (HOMO) level of most of the polymer donors in the absorption layer. The matched energy level ensures that PEDOT:PSS is a suitable electrode for hole collection. Dedoping of PEDOT:PSS can result in reduction of the W_F , endowing potential fabrication of low-work function PEDOT:PSS electrodes for electron collection. In 2002, Zhang et al. [3] first demonstrated that the sorbitol-doped PEDOT:PSS with an appropriated conductivity and work function could be employed as transparent electrodes in PSCs. Since then, more and more scientists have introduced and optimized PEDOT:PSS as a flexible electrode material. A high PCE over 12% for single-junction PSCs based on PEDOT:PSS electrodes has been achieved by several groups [4][5].

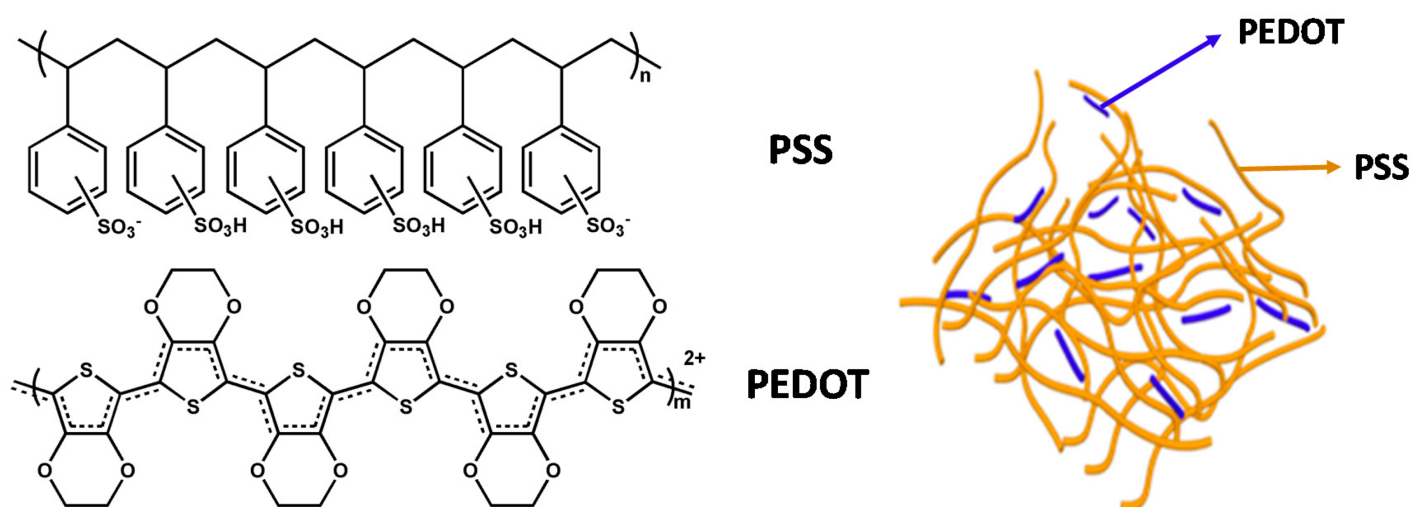


Figure 1. The chemical structure and schematic core-shell structure of Poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS).

2. PEDOT:PSS Electrodes

2.1. Synthesis and Commercialization of PEDOT:PSS Complex

A very feasible way to obtain PEDOT:PSS complex is the oxidative polymerization of EDOT in aqueous dispersion using sodium peroxodisulfate as the oxidant combined with a PSS template polymer. The PSS in the complex has two important roles. One is to act as counterions for charge balancing. A monomolecular oxidation product can be formed without counterions. Another role of PSS is to keep the PEDOT segments dispersed in the aqueous

solution as the PEDOT polycation is hardly soluble in any solvent. The obtained PEDOT:PSS aqueous dispersion is a deep-blue opaque solution. We also need to note that the molecular weight of PEDOT is only among 1000 to 2500 Da (about 6 to 18 repeating units) resembling an oligomeric nature [6]. Research from Inganas, et al. demonstrated this polymer complex is stable due to the ionic PEDOT⁺ and PSS⁻ not being separated by capillary electrophoresis [7].

PEDOT:PSS aqueous dispersion was first commercialized under the trade name of Baytron® by Bayer AG, followed by H.C. Starck and currently by Heraeus under the trade name of Clevios™. The company of Agfa Gevaert N.V. (German) also introduced PEDOT:PSS for large-scale printing applications under the trade name of Orgacon™. The PEDOT:PSS dispersions are classified into different grades based on the solids content and the ratio of PEDOT to PSS. The component strongly influences the particle size and distribution, viscosity, conductivity, and transmittance after film formation of the dispersion [6]. **Table 1** summarizes important properties of the commercial PEDOT:PSS Clevios dispersions.

Table 1. Commercial PEDOT:PSS dispersions in water and their properties. (Data are from the previous summarization [8][9]).

Trade Name	Solids Content in Water (wt.%)	PEDOT:PSS Ratio (w/w)	Viscosity at 20 °C (mPa)	Particle Size d ₅₀ (nm)	Conductivity (S/cm)
Clevios P	1.3	1:2.5	80	80	<10
Clevios PH	1.3	1:2.5	20	30	<10
Clevios PVP Al 4083	1.5	1:6	10	40	10 ⁻³
Clevios PVP CH800	2.8	1:20	15	25	10 ⁻⁵
Clevios PH500	1.1	1:2.5	25	30	500 ^a
Clevios PH750	1.1	1:2.5	25	30	750 ^a

a	Trade Name	Solids Content in Water (wt.%)	PEDOT:PSS Ratio (w/w)	Viscosity at 20 °C (mPa)	Particle Size d ₅₀ (nm)	Conductivity (S/cm)	dimethyl
	Clevios PH1000	1.1	1:2.5	30	30	1000 ^a	

PEDOT:PSS possesses several characteristics, such as high transmittance in the visible range, high and adjustable electrical conductivity, excellent thermal stability, high work function, as well as good film-forming ability by versatile fabrication techniques. These unique features ensure its widespread applications in various photo-electronic devices.

The transmittance of the PEDOT:PSS electrode is highly dependent on the film thickness. Generally, PEDOT:PSS thin films with a thickness around 100 nm possess high transmittance over 90% at 550 nm. The absorption spectrum of PEDOT:PSS is almost identical to that of in situ chemically polymerized PEDOT without PSS. Moreover, the addition of PSS or other additives like high boiling solvents does not influence the optical properties significantly. However, PEDOT:PSS film treated by concentrated sulfuric acid demonstrates a slight reduction in the transmittance [10]. The reason is mainly attributed to a stacking conformational change of the conjugated ionic PEDOT⁺ segments caused by the significant loss of PSS in the processing.

Electrical conductivity is one of the most important parameters considering its applications in various devices such as electrodes. The electrical conductivity is strongly dependent on the film morphology, chemical and physical structure, as well as the composite ratio, which in turn can be strongly modified via a variety of post treatments. In the past few decades, many efforts have been made in improving the electrical conductivity of PEDOT:PSS, and several approaches such as second doping as well as post-treatment have been reported to significantly improve the conductivity of PEDOT:PSS. In 2002, Kim et al. reported the electrical conductivity enhancement of PEDOT:PSS by doping polar organic solvents [11]. They demonstrated that the electrical conductivity of PEDOT:PSS can be enhanced by one order when dimethylformamide (DMF) is added into the aqueous solution, while enhancement over two orders can be achieved by doping dimethyl sulfoxide (DMSO). Conductivity enhancement was also reported by employing other polar organic solvents like ethylene glycol (EG), glycerol, and sorbitol [3][12]. Moreover, anionic surfactant, ionic liquid, as well as nonionic surfactant were found to be very effective in improving the electrical conductivity of PEDOT:PSS [13][14][15]. In addition to doping, film post-treatment is another common and effective method for enhancing the conductivity of PEDOT:PSS. As for post-treatment, a solvent or solution is dropped (or vaped) onto a PEDOT:PSS film, or the film is dipped into a solvent or a solution. The conductivity can be obviously enhanced through a post treatment with an organic solvent like DMSO, EG, methanol, 2-methoxyethanol as well as co-solvent, aqueous or organics solutions of salts and acids [16][17][18][19]. The strong acids such as HCl and H₂SO₄ are the most often used method for acid treatment. Ouyang and colleagues for the first time observed that conductivity can be obviously improved through a post treatment of the PEDOT:PSS film with concentrated H₂SO₄ [20]. As shown in **Figure 2**, the electrical conductivity of PEDOT:PSS is highly related to the concentration of H₂SO₄ and treated temperature. The electrical conductivity of PEDOT:PSS film can be enhanced to 2400 S/cm treated by diluted 1.5 mol/L H₂SO₄ and up to 3100 S/cm by repeating the process. This electrical conductivity is comparable to that of commercialized indium tin oxide (ITO) electrodes.

Later, Lee's group improved the electrical conductivity by optimizing the post-treatment conditions with concentrated H_2SO_4 [16]. They found the conductivity increased as the improvement of PEDOT:PSS crystallinity and a high conductivity up to 4380 S/cm was obtained. Up to now, the highest conductivity has reached 4840 S/cm through this H_2SO_4 post treatment method [21]. The H_2SO_4 -treated PEDOT:PSS demonstrates unique metallic or semimetallic behavior, indicating strong interactions among the PEDOT chains. Although great progress has been made in the improvement of electrical conductivity, its value is still much lower than that of metal electrodes. In addition, the conductive mechanism is still controversial and further efforts are necessary for exploring the origins of this conductivity enhancement.

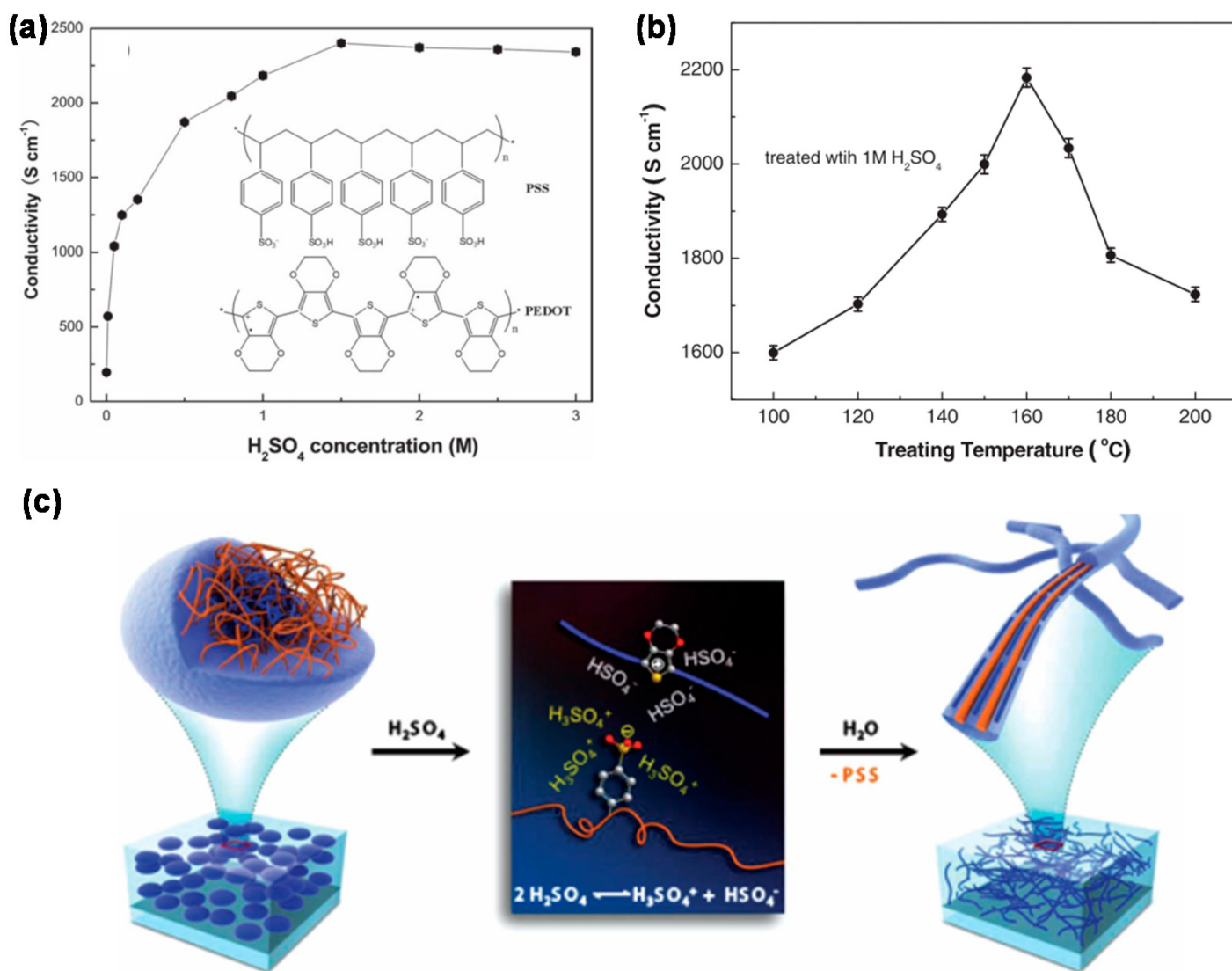


Figure 2. Conductivities of PEDOT:PSS films after treatment with H_2SO_4 solutions [20]. (a) PEDOT:PSS films treated with H_2SO_4 solutions of various concentrations at 160 $^{\circ}\text{C}$. (b) PEDOT:PSS films treated with 1 mol/L H_2SO_4 at various temperatures. The PEDOT:PSS films in (a,b) were treated with 1 mol/L H_2SO_4 only once. (c) Diagram of the structural rearrangement of PEDOT:PSS via a concentrated H_2SO_4 treatment [16].

2.3. Fabrication Technologies of PEDOT:PSS Electrodes

PEDOT:PSS can be deposited and can readily form continuous thin film on either rigid or flexible substrates by all common techniques employed for the deposition of waterborne coatings (such as spin coating, doctor blading, screen printing, inkjet printing; see **Figure 3**) [22]. However, pristine PEDOT:PSS solution possesses hydrophobic nature and cannot be processed directly. One of the effective methods is to employ plasma or ozone cleaning, but this makes the process complex. Another preferable method is to add surfactants (such as Zonyls, Dynols, and Triton X-100) into PEDOT:PSS solution to improve its wetting property [23][24][25]. Previous reports demonstrate that the contact angle of commercially available PEDOT:PSS solution (PH1000) on top of active layer can be reduced from 99.6° to 29.1° by doping 0.4% surfactant PEG-TmDD [14]. The PEDOT:PSS films achieved by the above procedures are smooth, and the thickness can also be controlled from several to hundreds of nanometers, making these films attractive as cladding layer for thin film device applications.

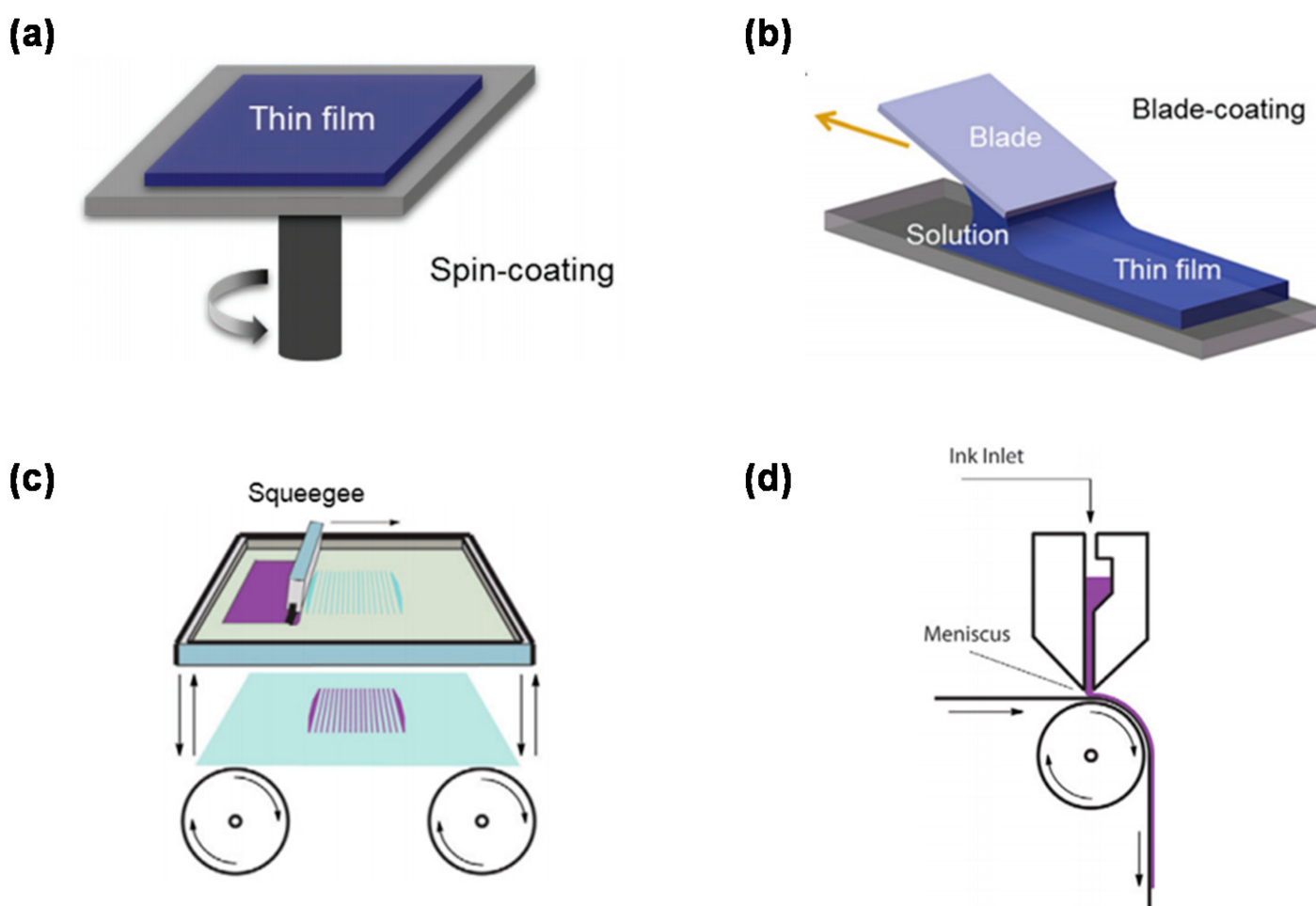


Figure 3. Schematic of (a) spin coating, (b) doctor blade coating, (c) screen printing, and (d) inkjet printing [22].

Spin coating is the most commonly used method in the lab to prepare a highly conductive PEDOT:PSS film as electrodes in the PSCs. During the preparation, the PEDOT:PSS dispersion is dropped onto a substrate followed by a substrate spinning at a selected speed. The thickness mainly depends on the solution concentration and the spinning speed. Normally, the PEDOT:PSS electrodes achieved by spin coating methods possess the film thicknesses of 100–200 nm and transmittances of over 80% in the wavelength range of 350–600 nm, which is

about 10% lower than that of commercialized ITO electrodes beyond 600 nm [8]. However, this technique is unsuited to a device with a large area and not compatible with the industrial roll-to-roll processing. Moreover, more than 80% of the solution/ink flies away during the processing. This motivates intensive research on printing techniques. The doctor blading technique is another coating technique that can be integrated with the roll-to-roll processing. The coating solution is placed in front of a sharp blade, and a thin wet film is formed after the blade moves across the substrate. The distance is typically 10–500 μm [26]. The coating solution is placed in front of a sharp blade, and a thin wet film is formed after the blade moves across the substrate. Film thickness depends on the concentration, viscosity of the solution, and the surface energy of the substrates. The doctor blading technique is not used as widely as the spin-coating method because it is time-consuming and requires a lot of materials to get the right conditions for film processing. Jang et al. propose a simple and fast patterning method based on the doctor blading by adjusting the wetting properties of PEDOT:PSS. They also reveal that the evaporation-induced flow exerts a great influence on the film morphology [27]. Screen printing is also a commonly used method. In this technique, a pre-patterned mesh is used to transfer ink onto a substrate. Then a blade or squeegee is moved across the screen to fill the open mesh apertures with ink followed by a reverse stroke to ensure the screen touches the substrate momentarily along a line of contact. Note that this technique requires a high viscosity of the solution, and the generated films always possess a large thickness. Therefore, this processing technique might not be used for fabricating the active layer but might be suitable for depositing PEDOT:PSS films.

Considering the difference in processing technology and concomitant film-forming mechanism, the requirements for characteristics including viscosity, density, and surface tension of the solution are different. Therefore, rational optimization of the solution composition, such as solvent and additives, is of great importance to obtain a high quality conductive film.

3. Polymer Solar Cells Based on PEDOT:PSS

Polymer solar cells (PSCs) have emerged as an alternative photovoltaic technology and attract tremendous attention owing to the advantages of a wide range of materials source, light weight, high mechanically flexibility, and solution processability. During a device preparation, ITO is always employed as the transparent electrode, and the vacuum-deposited metals (such as Al, Ag, Au) are generally used as top electrodes. Despite its good combination of high optical transmittance and high conductivity, ITO is too expensive and rigid while the vacuum-deposited metals consume high energy and require expensive deposition facilities with high-cost maintenance. Therefore, many scientific communities devote themselves to developing novel conductive materials as an alternative approach. The solution-processable PEDOT and its derivatives have become main candidates and have been functioned as bottom, top, and both electrodes in the PSCs (Table 2).

Table 2. Summary of the device performance of polymer solar cells (PSCs) based on PEDOT:PSS electrodes under AM1.5 illumination.

Device	Thickness (nm)	R (Ω/sq)	T (%)	J_{sc} (mA/cm^2)	V_{oc} (V)	FF	PCE (%)	Ref.
Glass/PEDOT:PSS:S/MEH-PPV/PCBM/Al	150	$\sim 10^3$	80	1.6	0.75	0.30	0.36	[3]
Glass/EG-PEDOT:PSS/MEH-PPV:PCBM/Ca/Al	250	250	-	5.1	0.74	0.39	1.5	[28]
Glass/PH500:5%DMSO/P3HT:PC ₆₁ BM/Ca/Al	100	213	90	9.73	0.63	0.54	3.27	[29]
PET/PH500:5%DMSO/P3HT:PC ₆₁ BM/Ca/Al	100	213	90	9.16	0.61	0.50	2.8	[29]
Glass/Methanol treated PH1000/P3HT:PC ₆₁ BM/Ca/Al	~50	25	85	9.51	0.58	0.67	3.71	[30]
Glass/H ₂ SO ₄ treated PH1000/PEDOT:PSS(4083)/P3HT:PC ₆₁ BM/Ca/Al	70	67	87	9.29	0.59	0.65	3.56	[20]
Glass/PEDOT:PSS:CNTs/PEIE/ZnO/PBDBTTT-C-T:PC ₇₁ BM/V ₂ O ₅ -RGO/Ag	-	40.51	80	15.76	0.77	0.62	7.47	[31]
Glass/Ag grid/PH500 /ZnO/C ₆₀ SAM/P3HT:PC ₆₁ BM/PEDOT:PSS(4083)/Ag	-	9.1	79	9.39	0.60	0.57	3.21	[32]
Glass/CH ₄ SO ₃ treated PH1000/PEDOT:PSS(4083)/PBDB-T:IT-M/PDINO/Al	80	40	-	16.01	0.925	0.72	10.60	[33]
PET/ CH ₄ SO ₃ treated PH1000/PEDOT:PSS(4083)/PBDB-T:IT-M/PDINO/Al	80	40	90	15.49	0.93	0.70	10.12	[34]
Glass/ITO/ZnO/P3HT:PC ₆₁ BM/PPP:PEDOT:PH1000	-	420	-	7.2	0.55	0.58	2.4	[35]
Glass/ITO/PEI/P3HT:ICBA/PH1000:PEG-TmDD	-	526	-	8.70	0.78	0.60	4.1	[14]

Device	Thickness (nm)	R (Ω/sq)	T (%)	J_{sc} (mA/cm^2)	V_{oc} (V)	FF	PCE (%)	Ref.
Glass/ITO/ZnO/PBDB-T:ITIC/MC-PH1000:EG:PEG-TmDD	-	-	-	13.0	0.86	0.66	7.38	[36]
Glass/metal/ZnO/P3HT:PCBM/PH1000 ^T /Ag-busbar	190	-	-	6.96	0.58	0.65	3.08	[37]
Glass/ITO/PEI/P3HT:ICBA/PH1000/PEI/P3HT:ICBA/PH1000:EG:PEG-TmDD ^T	-	-	-	3.10	1.62	0.68	3.60	[38]
Glass/ITO/PEI/P3HT:ICBA/PEDOT:PSS(4083)/HCT-PEDOT:PSS ^T	2780	2.60	-	8.65	0.81	0.66	4.6	[39]
Glass/PH500:5%DMSO/ZnO-NPs/C ₆₀ -SAMP3HT:PCBM/PEDOT:PSS(4083)/PH500:5%DMSO ^T	130	370	-	5.49	0.31	0.28	0.47	[40]
PES/PH1000 5% DMSO/PEI/P3HT:ICBA/PH1000:CPP-PEDOT ^T	130 (bottom) 160 (top)	-	-	7.1	0.80	0.52	3.0	[41]
PES/PH1000:5%DMSO/PEI/P3HT:ICBA ^T /PH1000:5%DMSO ^T	120 (bottom) 150 (top)	-	-	5.6	0.80	0.55	2.4	[42]
Glass/LWF-PEDOT:PSS/P3HT:ICBA/HWF-PH1000:EG:PEG-TmDD ^T	124 (bottom) 150 (top)	-	-	8.10	0.81	0.61	4.0	[43]
PES/H ₃ PO ₄ -PEDOT:PSS/PEI/P3HT:ICBA/EG-PEDOT:PSS	85 (bottom) 150 (top)	120 (bottom)	-	6.6	0.84	0.60	3.3	[44]

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Device	Thickness (nm)	R (Ω /sq)	T (%)	J_{sc} (mA/cm ²)	V_{oc} (V)	FF	PCE (%)	Ref.
PES/hc-PEDOT:PSS/PEI/P3HT:ICBA/PEDOT:PSS/PEI/... P3HT:ICBA/hc-PEDOT:PSS ^T	-	-	-	0.40	5.40	0.40	0.85	[45]

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both electrodes are included in this table. The parameters, such as thickness, sheet resistance (R) and transmittance are referring to the PEDOT:PSS-based bottom or top electrodes in the PSCs device. The superscript

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of the electrode in the device structure depend on the PEDOT:PSS layer deposited via film transfer, lamination

technique, CNTs: Carbon nanotubes, J_{sc} : Short-circuit current density, V_{oc} : open-circuit voltage, FF: Fill factor.

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