

Review

Emerging Transparent Conducting Electrodes for Organic Light Emitting Diodes

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Abstract: Organic light emitting diodes (OLEDs) have attracted much attention in recent years as next generation lighting and displays, due to their many advantages, including superb performance, mechanical flexibility, ease of fabrication, chemical versatility, *etc.* In order to fully realize the highly flexible features, reduce the cost and further improve the performance of OLED devices, replacing the conventional indium tin oxide with better alternative transparent conducting electrodes (TCEs) is a crucial step. In this review, we focus on the emerging alternative TCE materials for OLED applications, including carbon nanotubes (CNTs), metallic nanowires, conductive polymers and graphene. These materials are selected, because they have been applied as transparent electrodes for OLED devices and achieved reasonably good performance or even higher device performance than that of indium tin oxide (ITO) glass. Various electrode modification techniques and their effects on the device performance are presented. The effects of new TCEs on light extraction, device performance and reliability are discussed. Highly flexible, stretchable and efficient OLED devices are achieved based on these alternative TCEs. These results are summarized for each material. The advantages and current challenges of these TCE materials are also identified.

Keywords: transparent electrode; organic light emitting diode; carbon nanotube; metallic nanowire; graphene; conductive polymer

1. Introduction

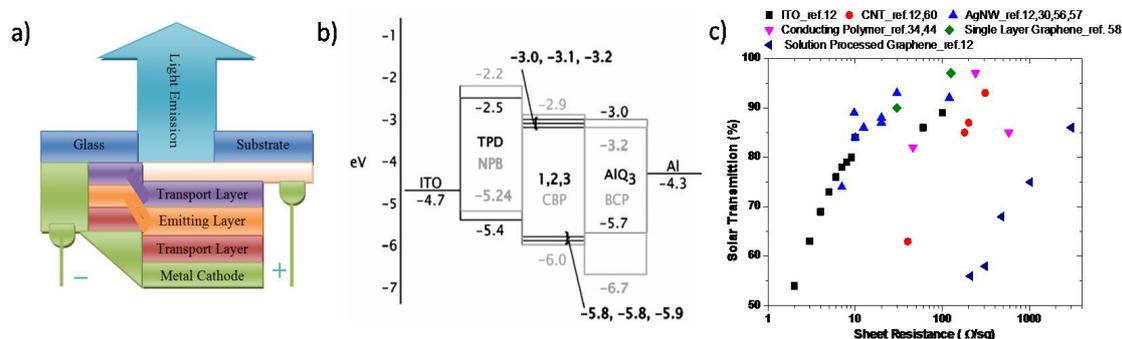
Organic light emitting diode (OLED) has emerged as a potential candidate for next generation flexible, large-area, high resolution display and solid state lighting panels, because of its high color quality, attractive appearance, ease of fabrication, low manufacturing and materials cost, *etc* [1]. With great efforts from both academia and industry, the OLED has been developed based on small molecule and polymer materials and also fabricated with both vacuum deposition and solution processes [2–5]. In the past few decades, researchers have been focusing on improving the device efficiency and lowering the manufacturing cost. Today, OLED displays are becoming dominant in the high-end market. OLED lighting is also on the verge of becoming widely commercially available, and its performance is competitive with that of its inorganic counterparts.

The basic OLED structure is composed of a stack of several layers: anode/hole transport layer (HTL)/emission layer (EL)/electron transport layer (ETL)/cathode, as shown in Figure 1a. [6] The first OLED was developed by Tang and VanSlyke in 1987 with the structure of an indium tin oxide (ITO)/aromatic diamine/8-hydroxyquinoline aluminum (Alq_3)/Mg-Al metal electrode [7]. Since then, ITO glass has been commonly used as the anode for OLEDs, because ITO simultaneously provides good transparency and conductivity [8]. Moreover, the work function of ITO is around 4.7 eV, which forms a low barrier for hole injection into the emission layer made of commonly used organic materials (Figure 1b) [9]. Despite these advantages, ITO is far from being a perfect candidate for OLED applications for the following reasons. First, it is not ideal for highly flexible electronics, due to its brittleness. Under mechanical bending or stretching, crack generation in the ITO film leads to much deteriorated electrical properties [10]. Second, the sputtering deposition of high quality ITO is a low throughput process and requires elevated temperature. Solution processed ITO also requires high temperature annealing to achieve a good conductivity [11]. It is vital therefore to only use substrates that are stable at high temperatures, which means an increased substrate cost and much reduced performance on plastic substrates. Furthermore, due to the widespread application of ITO as the transparent conducting electrode (TCE) for various optical devices and the limited global reserve of indium, the price of ITO will rise dramatically and further raise the cost of OLEDs. In addition, ITO does not offer ideal performance for OLEDs. It has significant light reflection and also traps the light in the waveguide mode. Its conductivity needs to be further improved, as well, for large area devices.

Considering all these factors, there has been increasing interest and an urgent need to look for alternative TCE materials to replace the conventional ITO. These TCE materials should be highly conductive and optically transparent; meanwhile, they should also be low cost and enable new attractive features. Here, we review the recent progress on the promising next generation TCEs. We focus our attention on the following materials: carbon nanotubes (CNTs), metallic nanowires, conducting polymers and graphene. These materials have shown the potential to fulfill standard requirements on the sheet resistance and transmission values of TCE and can be formed by low-cost processes, such as spin coating, spray coating and even roll-to-roll processes [12]. The sheet resistance and transmittance of these materials is summarized in Figure 1c using the solar spectrum as a reference for transmission evaluation. Moreover, OLED devices demonstrated with these techniques show great potential for future highly flexible, foldable and wearable opto-electronics. We summarize the progress

of each of these TCE materials with the device performance achieved and give comparisons between these techniques.

Figure 1. (a) Schematic diagram of the organic light emitting diode (OLED) structure; (b) energy level diagram of a simple OLED device consisting of *N,N'*-Bis(3-methylphenyl)-*N,N'*-diphenylbenzidine (TPD), *N,N'*-Di(1-naphthyl)-*N,N'*-diphenyl-(1,1'-biphenyl)-4,4'-diamine (NPB), 4,4'-Bis(*N*-carbazolyl)-1,1'-biphenyl (CBP), Bathocuproine (BCP), and Tris-(8-hydroxyquinoline)aluminum (Alq3); (c) sheet resistance and transmission chart for various types of transparent conducting electrode (TCE) materials including carbon nanotube (CNT), silver nanowire (AgNW), conductive polymers, and graphene.



2. Carbon Nanotubes

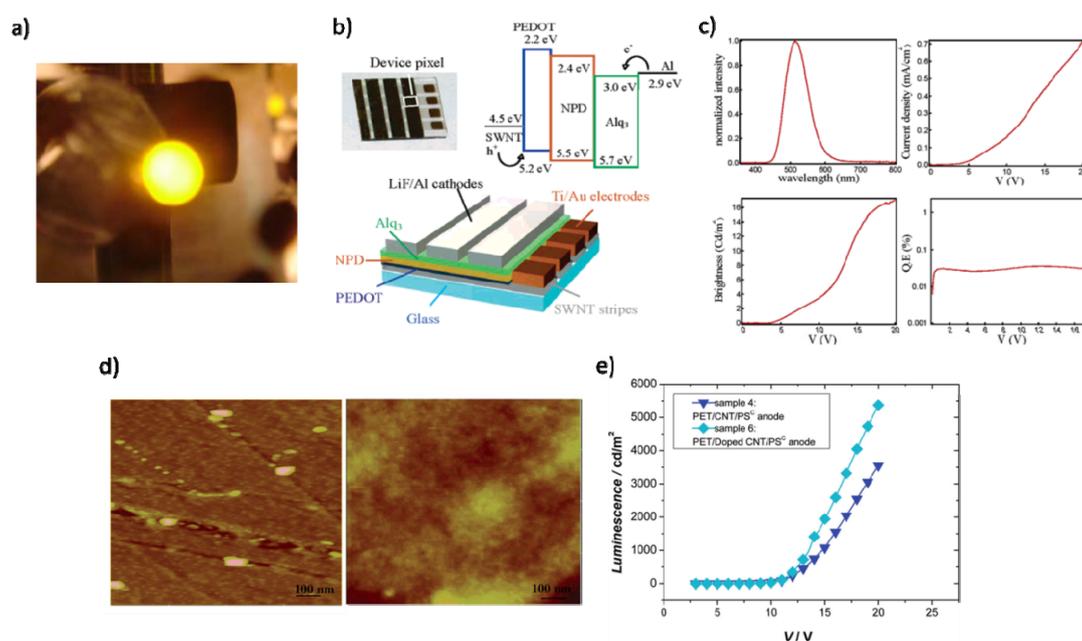
The carbon nanotube (CNT) network is the first nanostructured TCE investigated for OLEDs, leading to a boom of interest in this decade [13]. CNTs exhibit a unique electrical property in that they can be both metallic and semiconducting [14]. Because of this, they are widely applied as high-performance flexible transparent transistors, optical modulators, flexible emitters, as well as TCEs [15–17]. Metallic CNTs have a suitable work function (4.7–5.2 eV) for the application as anodes in OLEDs [18,19]. In addition, the high stability, flexibility and mobility of CNTs make the CNT network a potential candidate to replace the rigid ITO substrate, while avoiding the contamination of the organic layers from the oxygen atoms in ITO.

Zhang *et al.* first developed large area CNT sheets (meter long, 5 cm-wide) as the electrode for OLED, and this nanotube sheet was reported to be as strong as the steel (Figure 2a) [13]. This report demonstrated the huge potential of CNT networks' application in optical electronics and opened a new direction for the nanostructured TCE. Zhang *et al.* tested various CNTs from different growth methods [20]. The arc discharge nanotubes showed better performance compared to high-pressure CO conversion (HiPCO) nanotubes in surface roughness, sheet resistance and transparency. A sheet resistance of ~160 ohm/sq at 87% transparency can be achieved when the CNTs network is treated with SOCl_2 , as shown in Figure 2b,c. Li *et al.* demonstrated that a poly(3,4-ethylenedioxythiophene) poly(styrenesulfonate) (PEDOT:PSS) layer could play an important role in planarizing the roughness from CNT networks (Figure 2d) and decreasing the hole injection barrier from the CNTs to a polymer blend hole transporting layer poly(9,9-dioctylfluorene-co-*N*-(4-butylphenyl)diphenylamine) (TFB)+4,4'-bis[*p*-trichlorosilylpropylphenyl]phenylamino]biphenyl (TPD-Si₂), which could reduce the

leakage current [21]. With a PEDOT:PSS layer modified with methanol, a surface roughness less than 1 nm can be achieved, due to better PEDOT:PSS wetting onto the CNT network.

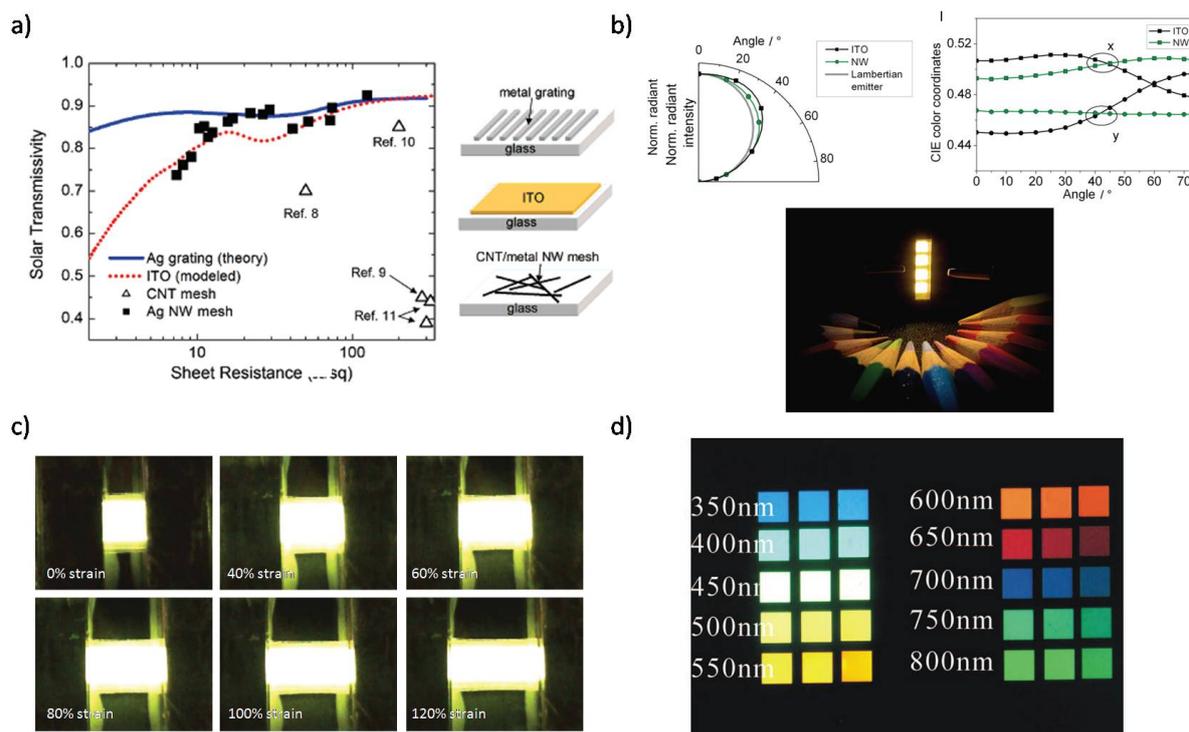
Ou *et al.* further modified the surface of the carbon nanotube network with PEDOT:PSS composite (PS^C) coating, which contained polyethylene glycol (PEG) additive in Baytron P500 and used HNO₃ acid treatment to improve the conductivity of the CNT network and the band alignment for hole injection [22]. Outstanding performance with a maximum luminance of ~9000 cd/m² and a luminance efficiency (LE) of ~10 cd/A at 1000 cd/m² was achieved, which was comparable to devices on ITO substrates. Yu *et al.* explored the capability of CNT transparent electrodes as the cathode and anode for flexible and transparent organic light emitting diodes by a lamination method [23]. Furthermore, stretchable OLEDs based on a CNT network as the TCE was built. The electroluminescent efficiency of the devices can be sustained under a 45% strain, which cannot be achieved for traditional ITO substrates [24]. The device stability with the CNT electrode exhibited comparable lifetime with that of the ITO electrode, and the acid resistivity of the CNT electrode to PEDOT:PSS is better than that of ITO electrode for long-term operation.

Figure 2. (a) Photograph of an OLED that uses an multi-wall nanotube (MWNT) sheet as the anode and poly(3,4-ethylenedioxythiophene) poly(styrenesulfonate) (PEDOT:PSS)/Poly[2-methoxy-5-(2-ethylhexyloxy)-1,4-phenylenevinylene] (MEH-PPV) /Ca/Al [13]; (b) patterned multilayer single wall CNT/PEDOT:PSS/NPB/Alq₃/LiF/Al [20]; (c) device performance: photoluminescence spectrum, current density vs. voltage bias curve, brightness vs. voltage bias and quantum efficiency as a function of current density [20]; (d) surface roughness of PEDOT:PSS ~4 nm and methanol-modified PEDOT:PSS ~0.96 nm on a CNT network [21]; (e) the luminescence vs. the voltage of an OLED with PEDOT:PSS composite (PS^C)-modified CNT on Polyethylene terephthalate (PET) substrate [22].



Reproduced with permission from Zhang *et al.*, Science, published by the American Association for the Advancement of Science, 2005 [13]; Zhang *et al.*, Nano Letter, published by the American Chemical Society, 2006 [20]; Li *et al.*, Nano Letter, published by the American Chemical Society, 2006 [21]; and Ou *et al.*, ACS Nano, published by the American Chemistry Society, 2009 [22].

Figure 3. (a) Solar photon flux-weighted transmissivity vs. sheet resistance for Ag gratings (blue line), ITO (red dotted line), CNT meshes (Δ) and Ag nanowire meshes (\blacksquare) deposited on a glass substrate. The Ag line width is a 40 nm and a 400 nm grating period [26]. (b) Normalized radiant intensity, color coordinates vs. viewing angle and photographic image of four operating nanowire (NW)-OLEDs [28]. (c) Photographs of a polymer light-emitting electrochemical cell (PLEC) (original emission area, $5.0 \times 4.5 \text{ mm}^2$) biased at 14 V at specified strains [29]. (d) Colors from blue to red can be selected by different period nanowire arrays [32].



Reproduced with permission from Lee *et al.*, Nano Letter, published by the American Chemical Society, 2008 [26]; Gaynor *et al.*, Advanced Materials, published by John Wiley & Sons, Inc., 2013 [28]; Liang *et al.* Nature Photonics, published by Macmillan Publishers Limited, 2013 [29]; and Hsu *et al.*, Apply Physics Letter, published by the American Institute of Physics, 2008 [32].

3. Metallic Nanowires

Recent studies on the metallic nanowire’s application in optical electronics have attracted a lot of attention. Similar to CNTs, high conductivity from the metal material and high transmittance from the open space between nanowires make the metallic nanowire a potential candidate as the TCE. Compared with CNTs, the metallic nanowire network shows better sheet resistance and transmission values, because the wire to wire contact resistance can be reduced by thermal treatment [25]. The low contact resistance between nanowires can significantly reduce the power loss on the electrodes. However, the metallic nanowire network requires PEDOT:PSS or other hole transport materials to ensure efficient hole injection as an anode, which slightly restricts the fabrication process. Lee *et al.* demonstrated the potential of a silver nanowire network TCE on a glass substrate with a sheet resistance of 16 ohm/sq and an average transmittance of 86% between the wavelengths of 400 and 800 nm, which is comparable to commercial ITO substrates (Figure 3a) [26]. Yu *et al.* first

demonstrated a composite electrode in which the silver nanowires were embedded in cross-linkable polyacrylate substrate, which could successfully replace the traditional rigid glass substrate [27]. This result opened up the possibility of realizing the high flexibility and high performance OLEDs by incorporating a solution processed metallic nanowire network.

Gaynor *et al.* investigated the angular dependence of white OLEDs using silver nanowires embedded in poly(methyl methacrylate) (PMMA) as the electrode [28]. The scattering of the silver nanowire network kept a stabilized viewing angle characteristic with reduced color shift and better Lambertian emission for the OLED. By further incorporation of light outcoupling techniques, a power efficiency of 54 lm/W was achieved, as shown in Figure 3b. Liang *et al.* reported an elastomeric polymer (polyurethane acrylate (PUA)) -based silver nanowire substrate with yellow light-emitting polymers consisting of ethoxylated trimethylolpropane triacrylate (ETPTA), polyethylene oxide (PEO) and lithium trifluoromethanesulphonate (LiTf), and the efficiency was kept at 2.5 cd/A under 120% strain (Figure 3c) [29]. The concern for metallic nanowire electrode is the instability, due to Rayleigh instability and contact ripening, resulting in the loss of the conductive path during operation. These might be the challenges for having long lifetime OLED devices [30,31].

The improvement of the silver nanowire TCE provided a platform for OLEDs to reach wider applications on display and lighting. Furthermore, the dimension of the metallic nanowire could affect the light scattering, light coupling and sheet resistance to transmission values of the TCE, providing us with an additional degree of freedom in improving the device performance. Aligned metal nanowire fabricated by a vacuum process was reported to have improved light outcoupling of the OLEDs (Figure 3d) [32]. The optical effect of the metallic nanowire on OLED and the alignment control of the nanowire through fabrication are still under investigation.

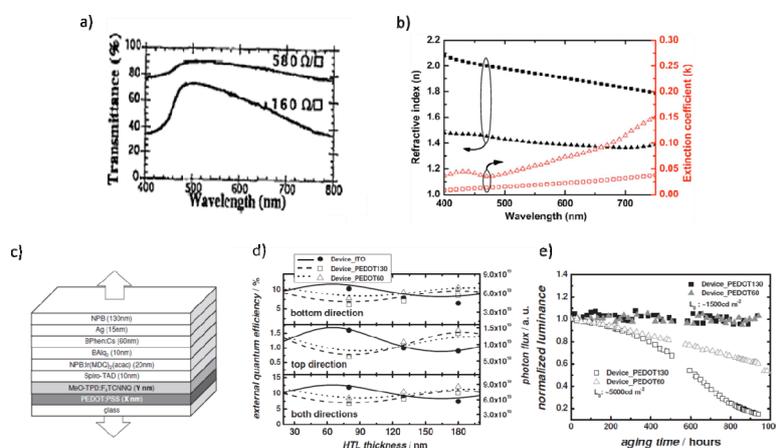
4. Conductive Polymers

Among various types of conductive polymers, PEDOT:PSS and polyaniline (PANI) are currently the most popular materials to replace the conventional ITO electrode. These two materials are well-studied, conjugated polymers with excellent mechanical stability, flexibility and, more importantly, they can achieve a high conductivity and transparency.

It was shown that PANI has the potential as a solution-processable TCE by Cao *et al.* [33]. They discovered that the camphor-sulfonic acid (CSA) doped PANI (PANI:CSA), which is soluble in *m*-cresol or chloroform, is conductive (<10 S/cm) and optically transparent. Gustafsson *et al.* successfully fabricated the PANI:CSA film on Polyethylene terephthalate (PET) substrate as the anode and demonstrated the first flexible polymer light emitting diode (PLED) device [5]. However, PANI:CSA film needs to be thicker than 250 nm to achieve 160 ohm/sq, while the transparency is about 70% under such a thickness (Figure 4a) [34]. Fehse *et al.* reported a new dispersion of PANI (D1033), which has a conductivity of 200 S/cm and a lower absorbance at 750 nm in the visible region than that of PANI:CSA (Figure 4b) [35]. Even though the improvement of the PANI properties resulted in better device performance, it is still lower than that of the commercial ITO substrate. PEDOT:PSS was invented in 1991 by Bayer [36] as an antistatic coating material. Cao *et al.* first introduced PEDOT:PSS into the PLED as a TCE with 500 ohm/sq and 75% transmission within the visible light region [37]. After that, PEDOT:PSS became commonly used as the hole injection layer for OLEDs, as

well. In order to improve the properties of the PEDOT:PSS, many treatments have been applied to improve the conductivity without losing the transparency. Kim *et al.* investigated different organic solvents, including dimethyl sulfoxide (DMSO), *N,N*-dimethyl formamide (DMF) and tetrahydrofuran (THF), and the addition of the high boiling polar solvent DMSO produced the highest conductivity among the three [38]. Ouyang *et al.* reported using ethylene glycol (EG) as a polar additive and acquired PEDOT:PSS thin films with a conductivity up to 160 S/cm, demonstrating a comparable performance to ITO/PEDOT:PSS on Poly[2-methoxy-5-(2-ethylhexyloxy)-1,4-phenylenevinylene] (MEH-PPV) PLED devices [39]. Fehse *et al.* used a commercially available high conductivity PEDOT:PSS Baytron PH500 (conductivity \sim 500 S/cm) as the TCE in small molecular OLED devices and achieved comparable or even superior performance (on green and blue emitting devices) than ITO anode devices [40]. Various treatments, like using zwitterionic surfactants, co-solvent system or exposing the films to dichloroacetic acid, were reported to achieve better conductivity in recent years [41–43]. Kim *et al.* demonstrated that with the addition of EG and a solvent post-treatment method, they could remarkably increase the conductivity up to 1418 S/cm, around 65 ohm/sq at 80% transmission [44]. Vosgueritchian *et al.* reported using a combination of DMSO and the fluorosurfactant Zonyl-FS300 (Zonyl) to achieve 46 ohm/sq at 82% transmission [45]. Recently, Xia *et al.* reported a H_2SO_4 treatment to reach 2400 S/cm and 3065 S/cm from multiple treatments [46]. These values are close to that of commercially available ITO substrates.

Figure 4. (a) Transmittance and sheet resistance of different polyaniline (PANI) thicknesses [34]; (b) the wavelength dependence of the refractive index and the extinction coefficient of ITO (squares) and PANI (triangles) films [35]; (c) the device structure of ITO-free transparent OLEDs based on a PEDOT:PSS TCE; (d) the comparison of external quantum efficiencies (EQEs) and corresponding photon fluxes for bottom and top emission and the sum of both (experiment (symbols) and optical simulation (lines) results); (e) aging characteristics of PEDOT:PSS-based OLEDs with a different PEDOT:PSS thickness. The encapsulated devices are aged over around 900 hours with an initial luminance of around 1500 cd/m^2 . Constant currents are applied for each sample according to the corresponding luminance [47]. HTL, hole transport layer.



Reproduced with permission from Yang *et al.*, Apply Physics Letter, published by the American Institute of Physics, 1994 [34]; Fehse *et al.*, Journal of Apply Physics, published by the American Institute of Physics, 2007 [35]; and Kim *et al.* Advanced Materials, published by John Wiley & Sons, Inc., 2013 [47].

Kim *et al.* demonstrated long lifetime (over 900 h under 1500 cd/m^2) and high performance (over 12% external quantum efficiency (EQE) under a driving current density of 10 mA/cm^2) OLEDs on polymer TCE. The results are shown in Figure 4c–d [47], comparing with those made from the ITO substrate. Moreover, from the optical study from Cai *et al.*, the PEDOT:PSS anode could be more beneficial to the light outcoupling than ITO substrates, due to the match of the refractive index [48]. At the current stage, however, these polymers still exhibit a lower conductivity and transmission than CNTs and metallic nanowire networks. In order to achieve a better device performance compared with ITO and other new materials, the properties and stability of the conductive polymers need to be further improved before they can be mass produced as the next generation TCE.

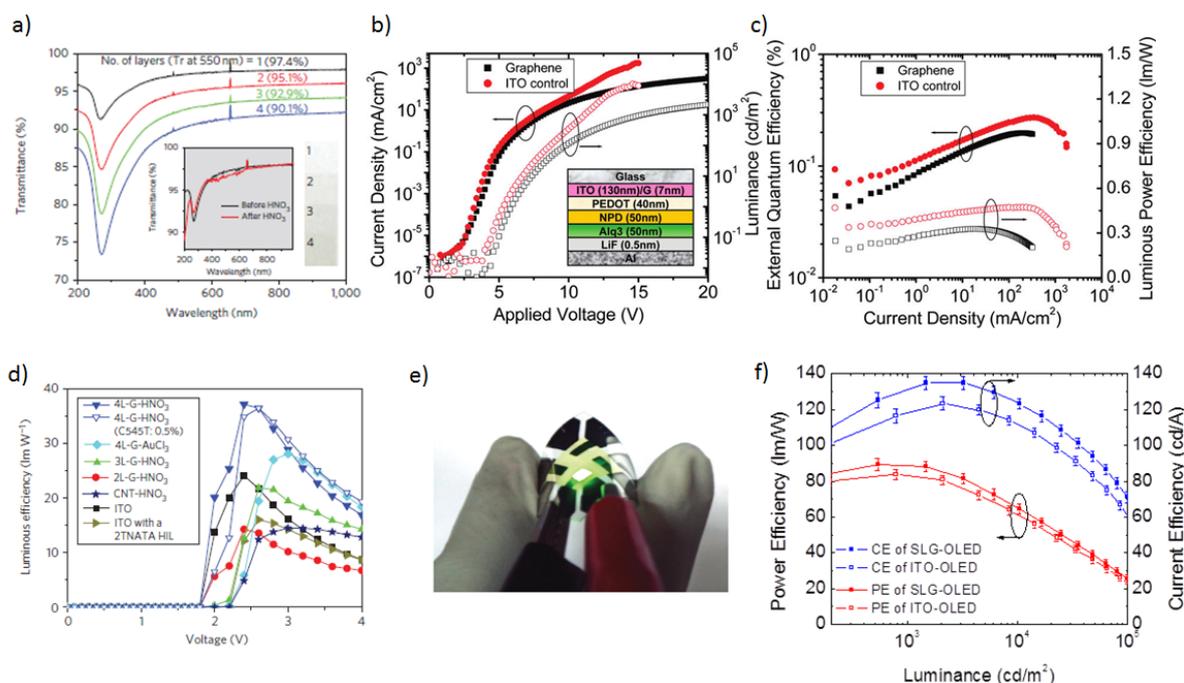
5. Graphene

Graphene is another promising candidate as a TCE in OLEDs. A flexible two-dimensional sheet of sp^2 -hybridized carbon atoms has very high conductivity and is nearly transparent. A single layer of graphene showed a sheet resistance of 125 ohm/sq and 97.4% transmission at the 550 nm wavelength, which is superior to the ITO substrate and other reported TCEs (Figure 5a) [49].

Wu *et al.* demonstrated solution processed graphene oxide thin films fabricated using Hummers' method and the ability to further reduce them to graphene thin films [12,50]. The graphene films were used as the TCE for OLEDs with a film thickness of about 7 nm, a sheet resistance of about 800 ohm/sq and 82% transmission at 550 nm, which is lower than the theoretical value, due to the existence of multiple grain boundaries, lattice defects and oxidative traps formed during the fabrication process. The device performance with a solution processed graphene electrode showed a turn-on voltage of 4.5 V and a luminance of 300 cd/m^2 at 11.7 V, which is comparable to those of ITO substrates, which had a turn-on voltage of 3.8 V and a luminance of 300 cd/m^2 at 9.9 V bias (Figure 5b,c). Han *et al.* recently reported an extremely efficient flexible OLED by introducing a work function tunable layer [51]. The mismatch of the electrical band alignment between graphene (work function $\sim 4.4 \text{ eV}$) and the hole transport layer, like *N,N'*-Di(1-naphthyl)-*N,N'*-diphenyl-(1,1'-biphenyl)-4,4'-diamine (NPB) (work function $\sim 5.4 \text{ eV}$), needs to be reduced for efficient hole injection. The authors incorporated a self-organized gradient hole injection layer (GraHIL), which was composed of PEDOT:PSS and tetrafluoroethylene-perfluoro-3,6-dioxo-4-methyl-7-octenesulphonic acid copolymer, one of the perfluorinated ionomers (PFIs). The GraHIL provided a work function gradient throughout the hole injection layer and, in turn, improved the charge injection efficiency. Based on a graphene/GraHIL/NPB/Alq₃/LiF/Al structure, the luminance efficiencies with doped graphene TCE as the anode (37.2 lm/W in fluorescent OLEDs, 102.7 lm/W in phosphorescent OLEDs) is higher than the devices using conventional ITO as the TCE (24.1 lm/W in fluorescent OLEDs, 85.6 lm/W in phosphorescent OLEDs, shown in the Figure 5d,e). Li *et al.* showed that white OLEDs (WOLEDs) on graphene electrode can indeed exhibit performance satisfying general lighting requirements [52]. WOLEDs on graphene with a power efficiency of 80 lm/W at a high brightness of 3000 cd/m^2 are demonstrated (Figure 5f). It is also found that graphene electrodes have the advantage of light extraction over ITO. White ITO has significant light reflection at both the top and bottom interface and also has significant light trapping. In the waveguide mode, Graphene, on the other hand, is so thin that

it is optically negligible. There is almost no light reflection and trapping in the graphene layer. Ultimately, more light can be coupled out of the graphene OLED than the ITO OLED [52].

Figure 5. (a) Transmittance of the roll-to-roll layer-by-layer transferred graphene films on quartz substrates. The inset shows the transmittance spectra of graphene films with and without HNO₃ doping and the optical images for the corresponding number of transferred layers (1 × 1 cm²) [49]; (b) Current density (filled symbols) and luminance (open symbols) vs. applied forward bias for an OLED on graphene (squares) and ITO (circles), with an OLED device structure of anode/PEDOT:PSS/NPD/Alq₃/LiF/Al; (c) External quantum efficiency (EQE) (filled symbols) and luminous power efficiency (open symbols) for an OLED on graphene film (squares) and ITO glass (circles) [12]; (d) Power efficiencies of OLED devices using various graphene layers (doped with HNO₃ or AuCl₃) and ITO as the anode; (e) Photograph of a flexible fluorescent green OLED with a four-layered graphene anode (4L-G) doped with HNO₃ (4L-G-HNO₃) [51]; (f) Power efficiency and current efficiency of white OLED (WOLED) based on ITO and single layer graphene electrode [52].



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The above high-performance OLEDs on graphene were all demonstrated using polymer interface layers. In contrast, OLEDs on graphene only electrodes without a polymer interface layers were also reported. In the early work, Sun *et al.* applied multilayered graphene from chemical vapor deposition as the anode for an OLED device and showed working devices without using the polymer interface layer, but with much less performance compared with the ITO reference device. The high leakage current and low efficiency were attributed to the surface roughness and work function mismatch [53].

Hwang *et al.* found that using weak plasma treatment to multilayered graphene can improve the hole injection efficiency and demonstrated high OLED device performance on graphene without polymer interface layers [54]. These works clearly indicate graphene's capability of charge conduction and injection. After optimization, graphene can serve as an excellent electrode by itself or together with a polymer layer.

Besides the high device performance, the stability of graphene is also excellent. Due to the inert material properties, it is not reactive with most chemicals. In addition, it does not have the problem of device degradation caused by electrode material diffusion into OLEDs, which can happen for Ag and ITO electrodes. The upcoming challenge for the graphene electrode is to further improve the electrical and optical properties and to make the fabrication process faster, easier and more cost effective.

6. Conclusions

Four different types of TCE materials are discussed in this review in terms of their electrical and optical performance as a TCE and the resultant OLED device performance. A comparison of their current status is summarized in Table 1. The performance of CNT TCEs is mainly limited by the contact resistance between adjacent wires, while it provides excellent mechanical properties and stability. The purity and type of CNT will also significantly affect the device performance, due to the difference in conductivity and surface roughness. Metallic nanowire TCEs exhibit better sheet resistance and transmission compared to their CNT counterparts, which resulted in highly efficient and stretchable OLED devices. Optical enhancement by the metallic nanowires is another major advantage for their future application and needs further investigation. Better control of the dimension and alignment of the metallic nanowires also needs to be further investigated. Conducting polymers have a longer developing history comparing with the other mentioned techniques. Their properties are becoming more and more competitive with traditional ITO glass showing similar conductivity values. They can also be tuned to provide optimal refractive index matching for efficient light outcoupling in OLEDs. The search for methods to precisely control the morphologies of the conducting polymers and to further improve the electrical and optical properties is ongoing. Graphene is the latest material to be applied as the TCE for OLEDs. The high transparency and the absence of light trapping from the ultrathin graphene sheet, the high in-plane conductivity and its ability to be solution processed make it a promising candidate as the next generation TCE. Its potential as the TCE in OLEDs has been proven. Outstanding OLED performance comparable to those on ITO glass has been achieved. Surface modification, the reduction of grain boundaries and defects for roll-to-roll processed graphene are some of the key routes towards further improvement.

Although most of these new TCEs still have many challenges in order to replace ITO completely, their performance is improving very rapidly. Even in the current stage, very low sheet resistance can be obtained by combining the new TCEs with a metal grid for large area applications [55]. Light extraction is an aspect that new TCEs can improve over ITO. The light reflection and trapping in most new TCEs are reduced comparing with ITO, due to the porous nature and scattering effect of CNTs and metal nanowires, the tunable index of the polymer, and the ultra-thin thickness of graphene, respectively. Because of this, light-extraction methods can be much simplified, and more light can be coupled out ultimately. In addition, new TCEs do enable many new attractive features, such as high

mechanical flexibility and stretchability. High efficiency or reasonably good efficiency devices are demonstrated for most of these TCEs, indicating that the process integration of new TCEs into high performance OLED devices will not be a fundamental hurdle for adopting these new materials. Low cost, high throughput and reliable process development is the key to commercial viability. Once a successful process is established, the impact of a new TCE is not only limited to the field of OLED devices, but can be readily applied to various other opto electronic devices. A myriad of applications can result from the development of one alternative material. These materials and related techniques are very likely to have a significant impact on optoelectronic research and the industry in the next few decades.

Table 1. Comparison of the TCE materials.

TCE materials for OLEDs	ITO	CNTs	Metal Nanowires	Conductive Polymers	Graphene
Conductivity/ Transparency (at 550 nm)	10 Ω /sq at 90%	180 Ω /sq at 85%	9.7 Ω /sq at 89% 30 Ω /sq at 93%	42 Ω /sq at 82% 240 Ω /sq at 97%	125 Ω /sq at 97% 30 Ω /sq at 90%
Light Reflection	High	Low	Low	Low	No reflection
Light Trapping	High	Low	Low	Low	No Trapping
Material Cost	High	Low	Medium	Low	Low
Process Cost	High	Potentially Low	Potentially Low	Low	Potentially Low
Stability	Good	Excellent	Medium	Medium	Excellent
Flexibility	Poor	Flexible	Flexible	Highly flexible	Highly flexible
Key OLED Performance Demonstrated	>100 lm/W (white)	10 cd/A at 1000 cd/m (green)	54 lm/W similar to ITO control device (white)	12% EQE 900 hours (green)	103 lm/W (green) 80 lm/W at 3000 cd/m ² (white)
Advantages over ITO		Solution process Flexible Stretchable	Solution process Flexible Stretchable Angle uniformity	Solution process Flexible Light extraction	Highly flexible Ultra-thin Light extraction
Challenges to Replace ITO		Conductivity Roughness Cost	Stability Cost	Conductivity Stability	Conductivity Cost
Reference	[56]	[23,24,57,58]	[29,30,59,60]	[45–48]	[12,49,61]

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Conflicts of Interest

The authors declare no conflict of interest.

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