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Review

Alternative transparent conducting electrode materials for flexible optoelectronic devices

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ABSTRACT

Transparent Conductive Electrode (TCE) is an essential part of the optoelectronic and display devices such as Liquid Crystal Displays (LCDs), Solar Cells, Light Emitting Diodes (LEDs), Organic Light Emitting Diodes (OLEDs) and touch screens. Indium Tin Oxide (ITO) is a commonly used TCE in these devices because of its high transparency and low sheet resistance. However, scarcity of indium and brittle nature of ITO limit its use in future flexible electronics. In order to develop flexible optoelectronic devices with improved performance, there is a requirement of replacing the ITO with a better alternate TCE. In this work, several alternative TCEs including transparent conductive oxides, carbon nanotubes, conducting polymers, metal nanowires, graphene and composites of these materials are studied with their properties such as sheet resistance, transparency and flexibility. The advantage and current challenges of these materials are also presented in this work.

Contents

1. Introduction

Transparent Conducting Electrodes materials have attracted attention of researchers and academicians due to their numerous applications in devices, such as in solar cells, photovoltaic cells, light-emitting diodes, touch-sensitive screens and flat-panel displays [1–3]. High transparency and low sheet resistance are two most desirable characteristics for a transparent conductive film. But, both transparency and conductivity are in a trade-off relationship, so it is difficult to achieve both the features simultaneously. Indium tin oxide (ITO) is a commonly used TCE having high transmittance (>80%) at visible wavelengths and a low sheet resistance (<40 Ω/sq) [4]. However, ITO has several disadvantages too, which limit its use in future flexible electronic devices. Firstly, ITO has brittle nature that restricts its use in flexible electronics [5]. Crack generates in ITO film after bending or stretching which discontinue the film and disturb the functioning of the device. Secondly, scarcity of indium material which makes ITO difficult for its use in future display and optoelectronics devices. Considering these factors, there is a requirement of alternative TCE materials for the replacement of ITO. As shown in Fig. 1, several types of materials such as transparent conducting oxides (TCOs) [6,7], carbon nanotubes (CNTs) [8,9], conducting polymers [10,11], and metal nanowires [12,13], graphene [14] and Hybrid/composites [12,13] of these materials have been explored as alternative of ITO as TCEs.

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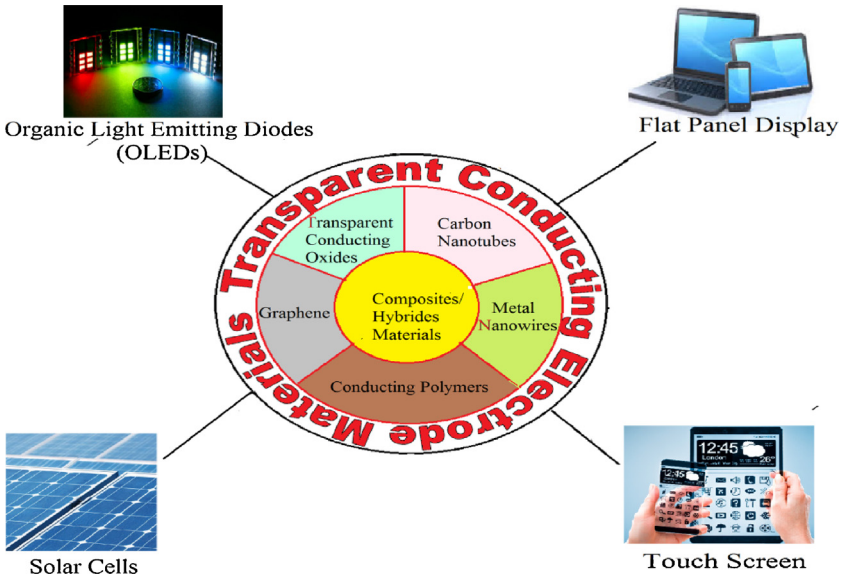

Fig. 1. Transparent Conducting Electrode Materials and their applications.

In order to discover a successful ITO replacement material, other doped metal oxide including impurity-doped ZnO and impurity doped SnO_2 were found promising candidates [15–17]. ZnO (films prepared by rf magnetron sputtering under an applied external magnetic field) showed impressive electrical and optical properties, and can be considered a good choice for using as TCE. Carbon nanotube (CNT) thin films also showed possibility of using them as an ITO replacement. Undoped CNT showed a sheet resistance up to 265 Ω /square with a transparency of 80% [18], while hybrid of a single wall carbon nanotube and silver (SWNT–Ag) showed sheet resistance up to 5.8 Ω/sq with a transparency of 83.7% [19], that proves CNTs as a suitable material for using as TCEs. Similar to ITO and CNTS, some metal nanowire such as Ag, Cu, and Ni nanowires were also tried and found impressive performance as TCEs [12,20,21]. Silver nanowire showed sheet resistance up to 10 Ω /sq with a transparency of 85% and considered as a good TCE [20], while some hybrid of Ag with other material also have been tried and proved good choice as TCEs for replacing ITO [12]. Two other materials, conducting polymers and graphene were also considered better choice for using as TCEs [22–25]. These have their own advantages in terms of flexibility with effective sheet resistance and transparency.

A number of review articles have been presented on different types of TCEs [26–28], but all the latest development in this field with a comprehensive study is not available at one place. Therefore, this review article will describe an exhaustive report on the latest developments on the TCEs for optoelectronic applications. Here, different materials including transparent conductive oxides, carbon nanotubes, conducting polymers, metal nanowires, graphene and their composites are discussed which have shown the potential of fulfilling the requirements of TCEs. Advantages and current challenges of these materials are also presented in this work which will provide a pathway to the naïve researchers working in this field.

2. Transparent conducting oxides (TCOs)

Transparent conducting oxides are the oxide materials which have made a special place in the field of TCEs in last few decades, because of their exceptional properties such as electrical conductivity and optical transparency. CdO, In_2O_3, SnO_2, Ga_2O_3 and ZnO are the well-known binary compounds of TCOs and introduced to some doping elements to improve electrical conductivity of these materials. In_2O_3: Sn, In_2O_3: F, SnO_2: F, SnO_2: Sb, and ZnO: Al are examples of some doped TCO [29]. In 1907, Bädeker introduced the first TCO material Cadmium Oxide (CdO) by using thermal oxidation of a thin film of sputter deposited cadmium (Cd) metal [30]. Later, fluorine (F), tin (Sn) and indium (In) doped CdO, and its compounds such as $CdIn_2O_4$, $CdSnO_3$ and Cd_2SnO_4 were reported in separate works and all of these showed good electrical and optical properties [29,31,32]. The lowest reported resistivity of CdO-based TCOs are of order 10^{-4} Ω cm with a transmittance of 85%–90% in visible range. However, the toxicity and narrow band gap of CdO are considerable drawbacks of Cd-based TCOs and make them less desirable for practical applications [33]. SnO_2 is another TCO developed after CdO. E. Elangovan *et al.* reported a SnO_2 layer deposited by spray pyrolysis technique having a sheet resistance of 38.2 Ω /sq with a transparency of 80% in visible range [34]. In the same work, fluorine (F) doped SnO_2 and antimony (Sb) doped SnO_2 films were also reported with a sheet resistance of 1.8 Ω/sq and 2.2 Ω/sq, respectively. However, the transparency of SnO_2: F was better than SnO_2, and transparency of SnO2: Sb was less than SnO_2.

In 1960s, a great breakthrough was achieved in the field of TCEs, when ITO a compound of indium oxide (In_2O_3) and tin oxide (SnO_2) was introduced. ITO thin films have low sheet resistances (40 Ω/sq) and a typical transmittance of 80%–95% in the visible range [4]. However, ITO's brittle nature and scarcity prevents its use in flexible practical applications such as TCEs in flexible optoelectronic devices. The mechanical properties of ITO are highly influenced by the microstructural quantities of its films, such as crystalline/ amorphous state, crystallographic orientation, crystallite size, strain and stress [35]. More specifically, the tensile strain at failure of the coating or crack onset strain (COS) is a key property that determines the functional reliability of the coated films for flexible devices. Leterrier *et al.* observed the mechanical properties of ITO film sputtered on flexible substrate and reported the effect of thickness of sputtered ITO film on the crack onset strain (COS) [36]. The deposited ITO film showed some microdefects in the form of pin-holes which caused small cracks upon a strain of 1.28%. After further increasing the strain up to 1.42%, these crack propagated from dot size to some finite size and the resistivity began to increase. At higher strain levels, the finite cracks increased further and the width covered to the whole sample. As a result, the resistivity increased significantly and indicates that the functional performance loss of the ITO coated polymers was controlled by crack propagation features,

rather than by crack initiation. They also observed that the COS decreased with an ITO thickness and for a thicker film of ITO the safe operating range could be even smaller. In some other works, the onset of cracking of ITO was measured less 2% strain using In-situ optical observations and uniaxial compression that resulted an abrupt increase in electrical resistance [37]. Since future flexible devices not so durable with ITO as TCE and are r equired to be replaced with better alternate.

After ITO, doped ZnO materials are considered as suitable TCOs because of having good transparency and low sheet resistance. ZnO: Al (AZO), ZnO: Ga (GAO) and ZnO: W has shown better results and are also easily available materials [16,17,38]. Another oxide material indium zinc oxide (IZO), which is a combination of In_2O_3 with ZnO is also widely reported [39]. But the significant indium ratio in IZO films raise the issues of indium supply which became the drawbacks of IZO films. Some TCO materials with their sheet resistance and transparency properties are given in Table 1.

Recently few multilayer structures have been reported with improved sheet resistance and transparency properties. A multilayer SnO_2/Ag//SnO_2 has been developed on glass substrates by sequentially using RF/DC magnetron sputtering at room temperature [43]. In this work, Sheet resistance of 9.51 Ω/sq with a transmittance of 85.5% was reported at the multi layers with the structure of SnO_2 (30 nm)/Ag (10 nm)/SnO_2 (30 nm). 1n 2016, low-temperature sputtered amorphous ZTO film was demonstrated as an alternative to ITO film in large-area flexible OLEDs [43]. A combinational analysis of ZnO: Al and SnO_2 compositions were used to develop the ZTO films. The developed ZTO films showed low resistivity, high transmittance, comparable brightness and luminance efficiency to ITO as shown in Fig. 2which made them suitable for using as OLED anodes. In the same work, ZTO/Ag/ZTO and ZTO/metal (Mo/Al/ Mo) grid anodes were used in OLEDs and showed approximately similar results which were shown by OLEDs having low-temperature ITO and ITO/Ag grid anodes, respectively. Besides, the composition of earth-abundant elements for developing ZTO films encourages the replacement of indium in optoelectronic applications. In other work, a tri-layer film of SnO_2 (25 nm)/Ag (5 nm)/SnO_2 (25 nm) was developed on quartz glass substrates using RF magnetron sputtering for an SnO_2 layer and DC magnetron sputtering for an Ag layer [44]. Optoelectronic properties and surface morphology were measured which reported a resistivity of $4.8 \times 10 - 5$ Ω cm, the sheet resistance of 9.61 Ω/sq and the average transmittance of > 83% in the visible light region (400–800 nm).

As discussed above CdO, SnO_2, In_2O_3 and ZnO based transparent conducting oxides have shown promising properties for replacing ITO. But, all of them pose some challenges which limit their uses in practical applications like Cd is toxic in nature while SnO_2 has the long history of work with limited success. However, work on SnO_2 in combinations with the other binaries oxides such as ZnO, InO_3 is ongoing. The combination of In_2O_3 with ZnO, indium zinc oxide (IZO) is widely reported for high conductivity and transparency, but the significant concentration of indium in IZO films again causes the issue of indium supply. Among alternative oxides, doped ZnO (i.e ZnO:Al and ZnO:W) are the most preferred material among all TCOs due to their low sheet resistance and high transparency for replacing ITO as TCEs. The crack on set of both ZnO:Al and ITO deposited on polymer substrate is approximately 2% while chemical compatibility of ITO with Poly(3,4-ethylenedioxythiophene) poly(styrenesulfonate) (which is acid-containing layers), is limited by amorphous indium and susceptible to leeching that leads the requirement of the development of alternative transparent conductor like ZnO is highly desirable [37,45]. However, ZnO:Al also has poor stability in oxidizing environments and in etchability when these are in contact with acids [43,46]. On the other hand, thus even after a lot of works in this field, the alternative TCO material to ITO for different application is not so clear and it seems difficult for a single TCO material to be a universal replacement for ITO.

3. Carbon nanotubes

Carbon nanotubes (CNTs) are cylindrical structured nanoscale diameter material, which exhibit excellent electrical and mechanical properties with exceptional work function and flexibility. Iijima *et al*. demonstrated the multi-wall CNT first time in 1991 [47] and single-wall in 1993 [48]. However, transparent conducting film of CNT was first time introduced by Wu *et al.* [49] and Saran *et al.* [50] in their work in 2004. After that, a lot of work has been done on CNTs to improve its properties for using in various applications. In 2005, Zhang *et al*. reported large area CNT sheets with 5 cm-width and as strong as the steel [51]. Miguel Contreras *et al*. explored the capacity of SWCNT film by using it solar cell in replacement of ZnO [10]. SWCNT film was developed which exhibits the sheet resistance of 50 Ω/sq and 100 Ω/sq with a transparency of 60% and 70%, respectively and found high optical transmission in the IR (>1200 nm) range which shows application possibility in PV technologies that use more of the IR portion of the solar spectrum. X. Yu and his coworkers demonstrated the use of acid (mixture of concentrated sulfuric acid and nitric acid) treated SWCNT for developing highly transparent and electrically conductive thin films on plastic substrates [9]. The fabricated SWNT thin film exhibited the thickness of 100 nm with surface resistivity of 6 kΩ/sq and transmittance of 88% in the visible range, which was three times better than untreated SWNTs films but still have high resistivity. In another work, David S Hecht achieved the sheet resistance of 60 Ω /sq and transmission of 90.9% of CNTs films in visible range [52]. In this work a unique CNT deposition technique for superacid CSA dispersions was used which resulted in uniform and superior electrical conductive CNT films.

Williams *et al.* demonstrated the boron-doped SWCNT film of sheet resistances of 7 kΩ/sq with a transparency of 89% in visible range on PET substrate [53] and developed different OLED devices with boron-doped and undoped SWCNTs as anode and compared the measurements. The comparison showed that the undoped SWCNT sample did not produce any light due to having high sheet resistance. However, B-doped SWCNT sample glowed with red light of illuminance of 0.3 lx when applied a voltage of 5 V, which showed the effect of boron doping in SWCNT. Kuan-Ru Chen and his coworkers used slot, dip and blade coating techniques separately for developing SWCNT film on polyethylene terephthalate (PET) substrate [54]. All of these techniques reported approximately same transparency above 85% and sheet resistance close to 1000 Ω/sq. In this work, the relation between the sheet resistance, transparency of the SWCNT coated film was reported with coating speed of these coating techniques. Both sheet resistance and transparency of the SWCNT coated film found decreasing by increasing the coating speed for dip and blade coatings techniques, but were independent for slot coating. That makes the slot die coating method more suitable for industrial production in terms of high coating speed and uniformity of optoelectronic properties.

Dachuan Shi and Daniel E. Resasco reported conductive and highly transparent single-wall carbon nanotube (SWCNT) films on quartz substrates [55]. In this work, Ferritin was used as the catalyst precursor, ethanol as the carbon feed. The result was analyzed for different concentrations of Catalyst and different annealing time and concluded that of both sheet resistance and transparency decreases with increasing annealing time. It was also observed that the transparency of the film decreased with increasing ferritin concentration while the sheet resistance was minimum at Conc. 33. A CNT network electrode based stretchable OLEDs was developed by Yu *et al*., in which the electroluminescent efficiency of the devices

Table 1
TCO materials with their sheet resistance and transparency properties.

S. No.	TCO Material	Deposition Technique	Sheet Resistance(Ω/sq)	Transparency(%)	References
1.	SnO_2	Spray	38.2	80	[39]
2.	SnO_2: Sb	Spray	32	85.3	[15]
3.	SnO_2: F	Sputtering	4.63	87	[39]
4.	Cd_2SnO_4	Sputtering	10	80	[6]
5.	$In_4Sn_3O_{12}$	Sputtering	400	96	[7]
6.	IZO	Sputtering	15.2	89	[40]
7.	In_2O_3	Reactive Evaporation	37	85	[41]
8.	ITO	Sputtering	5	80	[4]
9.	ZnO (deposited under an applied external magnetic field)	Sputtering	10	85	[42]
10.	ZnO:Al	Sputtering	7.65	91.2	[38]
11.	ZnO:Ga	PLD	14.5	90	[16]
12.	ZnO:W	Sputtering	6.8	91.3	[17]

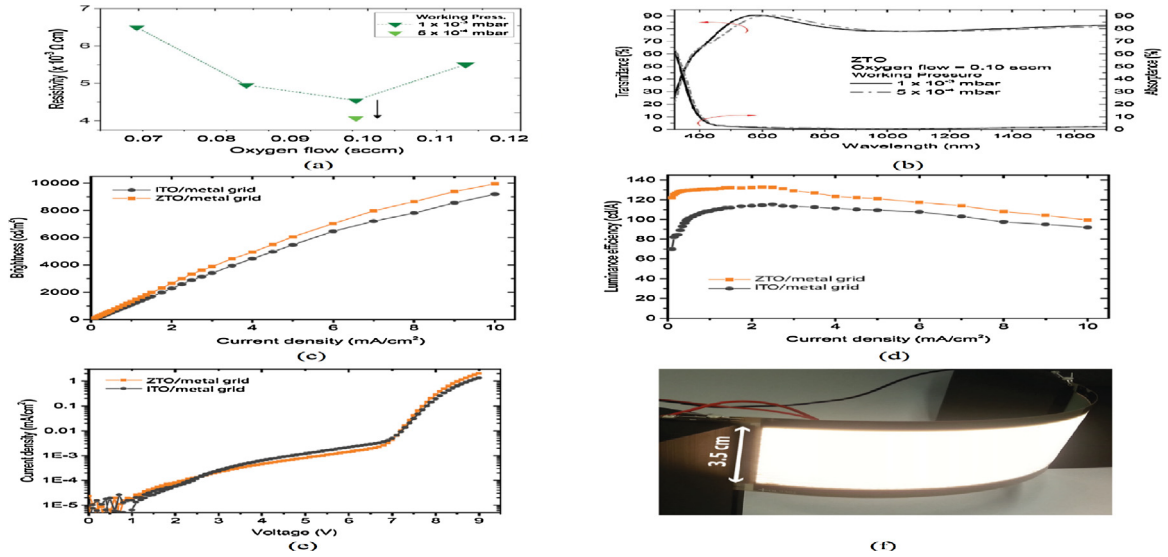

Fig. 2. (a) Resistivity of ZTO films sputtered from a single target as a function of oxygen flow, (b) Optical transmittance and absorptance spectra of two ZTO electrodes deposited at two distinct working pressures: 5×10^{-4} and 1×10^{-3} mbar, (c) Brightness *vs.* current density, (d) luminance efficiency *vs.* current density, and (e) current density *vs.* voltage characteristics for devices with ZTO/grid and ITO/grid anodes. (f) Photograph of a curved 3.5 × 11.5 cm^2 sm-OLED device fabricated on the ZTO/grid anode [44].
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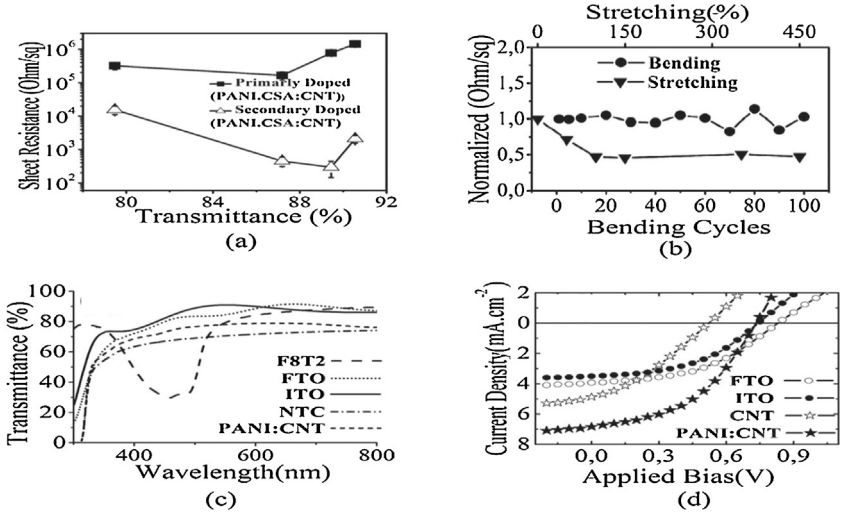

Fig. 3. (a) Sheet resistance *vs.* transmittance at 550 nm for the four PANI:CNT films before (square) and after (open triangles) the metacresol exposure, (b) the normalized sheet resistance of PANI:CNT 5 film deposited over PET after 100 bending cycles (circles) and stretching(triangles), (d) Transmittance spectra of different films (glass/ITO, glass/ FTO, PET/CNT and PET/PANI: CNT); b) current density (mA cm^{-2}) *vs.* voltage (V) for the different anodes used in the solar cells [57].
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can be sustained under a 45% strain [56]. In 2013, Salvatierra *et al.* demonstrated thin films of Polyaniline/carbon nanotube with a sheet resistance of 295 Ω/sq. and a transmittance of 89% at 550 nm shown in Fig. 3 [57]. The developed film showed good mechanical stability and can be easily deposited onto flexible substrates like PET.

The PANI/CNT films were used in Organic photovoltaic cells as transparent anodes and compared to ITO anode based equivalent devices. The results indicate that novel ITO-free optoelectronic devices can be optimized with very high performance using interfacial films of conjugated polymers and carbon nano-material species. The applications of CNT proved their importance as a TCE in optoelectronic applications. Furthermore, Jing Gao *et al.* demonstrated blue OLED devices with modified SWCNT films as electrode [58], and developed OLED devices with SOCl2-TCFs, PEDOT:PSS-coated and PEDOT:PSS-mixed SWCNT TCFs. By compare the measurements, it was found that the SWCNT TCFs mixed with PEDOT: PSS TCFs shows comparatively low sheet resistance and high transparency which leads them a suitable anode candidate for optoelectronic devices. Few more CNT TCFs with their sheet resistance and transparency in visible range are given in Table 2.

As discussed above, CNT can be a good alternative of conventional ITO in some applications. CNT have good electrical properties, mechanical properties, exceptional flexibility and proper work function. A linear increase in resistance have been reported and showed 38% at 25% nominal uniaxial tensile strain, which is 10 times higher than the strain observed in the ITO coated PET substrate. This high strain proves high flexibility of CNT in comparison to ITO. However, CNT has high sheet resistance and high roughness that affects the carrier mobility and shorten the lifetime of the device [55,57]. So still there is a requirement of suitable method of developing a high conducting and transparent CNT film with large volume production.

4. Transparent conducting polymers

Conducting polymers are the organic materials which exhibit high transparency, electrical conductivity and flexible in nature. Because of these properties the polymers found their use as a transparent conducting electrode for various optoelectronic devices. Transparent conducting polymers are more advantageous in comparison to other electrode materials, due to their light weight, mechanical flexibility and excellent compatibility with plastic substrates. In late 1960s and early 1970s, Pohl *et al.* demonstrated conjugated semiconducting polymer and introduced the conductivity in these polymers [68]. Various semiconducting phenolphthalein-type polymers were prepared by treating a no. of phenols with acid anhydrides. In another work, McNeill and Weiss prepared xanthene-type polymers related to fluorescein and reported the conductivities of order of 10^{-4} mho/cm which was very low for practical applications [68]. In 1977, Heeger *et al.* showed improvement in electrical conductivity of polyacetylene when doped with controlled amounts of the halogens chlorine, bomine, or iodine, and with arsenic pentafluoride (Asp_5) [69]. Next year they synthesized highly conducting films of derivatives of Polyacetylene, $(CH)_x$ and showed that the electrical conductivity of the synthesized films could be systematically and controllably varied over a wide range, with up to 10^{11} times of the conductivity [70]. The work was another breakthrough in the history of conducting polymers. "Nobel Prize in Chemistry 2000" was awarded for the development of electrically conductive polymers [71]. After that, conducting polymers become an area of interest for researchers and attracted attention because of their many possible applications including transparent conducting electrode.

Polyaniline (PANI), polypyrrole (PPY) and, poly(3,4-ethylenedioxythiophene) :poly(4-styrenesulfonate) (PEDOT:PSS) are the three most important conducting polymers, out of these PEDOT:PSS shows the capability to replace the conventional ITO electrode as TCEs in optoelectronic devices. These polymers are highly transparent, flexible, good conductors and ease in fabrication, which makes them suitable for optoelectronic devices. In 1992, Cao *et al.* developed transparent conducting film of polyaniline (PANI) by doping of camphor-sulfonic acid (CSA) in it [72] and reported a surface resistance less than 100 Ω/sq and a transmittance of approx. 70% between 475 and 675 nm wavelength in visible range. The work showed the ability of PANI as TCEs and needed more improvement in tradeoff between transparency and sheet resistance. In another work, Sung Soon Im and Sung Weon Byun worked on PANI and polypyrrole. Polypyrrole-nylon 6 (PPy-N) and polyaniline-nylon 6 (PA- N) composite films was developed by immersing nylon 6 films containing pyrrole or aniline into an oxidant solution, and reported a maximum conductivity of 10^{-3} S/cm and a transmittance of 75% of the PPy-N composite films and conductivity of 10^{-2} S/cm and a transmittance of 75% of the PA-N composite films [10]. The work showed small improvement in the conductivity which was still unsuitable for using the material as TCEs. L. Swathiha Priyadharshni and M. Selvaraj introduced electrically conductive films of polypyrrole sulfonate by spin coating of copolymerized pyrrole with methyl sulfonic acid [11]. The developed layer achieved a conductivity of 8×10^{-3} S/cm^2 with transparency 60% in visible range that shows poor performance in comparison to ITO. In another work, Yusuke Hoshina and Takaomi Kobayashi developed electrically conductive films by copolymerization of pyrrole (Py) and 2-formyl pyrrole (FPy) in the presence of catalyst trifluoroacetic acid (TFA) in chloroform [73]. The resulted films had a metallic greenish black colour and showed electrical conductivity of 10^{-4} to 10^{-1} S/cm after doping of I_2 in the copolymer films. However, the conductivity was not so satisfactory to use the film as TCE in replacement of ITO. Instead of these, there are some more polymer electrodes have been developed which are given in the Table 3 with their sheet resistance and transparency.

Nishii *et al.* developed a thin film of PEDOT/PSS through coating of its aqueous solution onto a PET film with a sheet-resistance of 260 Ω/sq and a transmittance of around 85% [78]. A flexible electronic-paper display was fabricated by using the PEDOT: PSS film as top electrode in it. Laser adaption method was used for high resolution electrode patterning of the polymer materials in a line shape and an 8.4-in.diagonal and 82 dpi flexible electronic paper was developed using quick response liquid-powder technology. In another work, Zhang *et al.* demonstrated conductive and transparent PEDOT: PSS films by using post-spin-rinsing method (PSRM) with polar organic solvent dimethyl sulfoxide (DMSO) [79]. The film showed a conductivity of 1335S cm^{-1}, which is much higher than the conductivity (776S cm^{-1}) of the PEDOT: PSS film. They used the fabricated PSRM PEDOT: PSS film in polymer solar cells and found a power conversion efficiency of 4.82%. In another work, an elastomeric polymer light-emitting devices was fabricated using AgNW–PUA (poly (urethane acrylate)) composite electrodes in it [80]. The resulting PLED exhibited a highly elasticity at room temperature and could emit light at strains as large as 120%. It was also showing significant improvement in efficiency in the stretched state as shown in Fig. 4

Recently, an inkjet printing embedded Ag-PEDOT: PSS electrode has been developed and used in blue polymer light emitting diodes as anode [81]. The device with 8 mm embedded grid/PEDOT:PSS showed the efficiency of 250% compared to the ITO reference device due to embedding, enhanced light out coupling, and optimized grid geometry. There is requirement of more improvement for upscale fabrication process and use it industrial use. In another work, a highly stretchable and conductive PEDOT polymer is reported by Wang *et al.* by incorporating ionic additive assisted stretch-ability

Table 2
CNT TCFs with fabrication techniques and their properties in visible range.

S. No.	Material	Deposition Technique	Sheet Resistance(Ω /sq)	Transparency(%)	References
1.	MWCNT	Roll to roll-dry	24	83.4	[59]
2.	SWCNT	Slot die coating	1000	92	[54]
3.	SWCNT	Dip Coating	100	90	[60]
4.	SWCNT	Electrophoretic deposition	220	81	[61]
5.	CNT	CVD	265	80	[62]
6.	CNT-Graphene hybrid	Spin Coating	240	86	[63]
7.	RGO/CNT hybrid	Langmuir – Blodgett	183.5	77	[64]
8.	RGO/CNT hybrid	Spin coating	631	81.3	[65]
9.	SWNT–Ag hybrid	Spray	5	83.7	[66]
10.	CNT-PEDOT hybrid	Vacuum filtration	80	75	[67]

Table 3
Polymer materials with their sheet resistance and transparency properties.

S. No.	Material	Deposition Technique	Sheet Resistance(Ω/sq)	Transparency(%)	References
1.	PANI	Spin Coating	166	80	[62]
2.	PEI/Ag/PEDOT:PSS	Spin Coating thermal evaporation	10	95	[74]
3.	PEDOT:PSS (H2SO4 -treated)	Spin coating, dip method	39	80	[23]
4.	PEDOT:PSS (methanol treated)	Spin coating, dip – drop method	90	85	[75]
5.	PEDOT:PSS– 5% DMSO	Spin coating	160	80	[76]
6.	PEDOT:PSS/AgNW	Drop Cast, Spin coating	12	82	[76]
7.	AgNW:PUA	–	15	83	[77]
8.	CNT-PEDOT hybrid	Vacuum filtration	80	75	[67]
9.	PEDOT:PSS/AgNW/graphene	Spin Coating	181.67	71.21	[12]
10.	AgNW/IZO/ PEDOT:PSS	Spin Coating, Sputtering	5.9	86	[13]

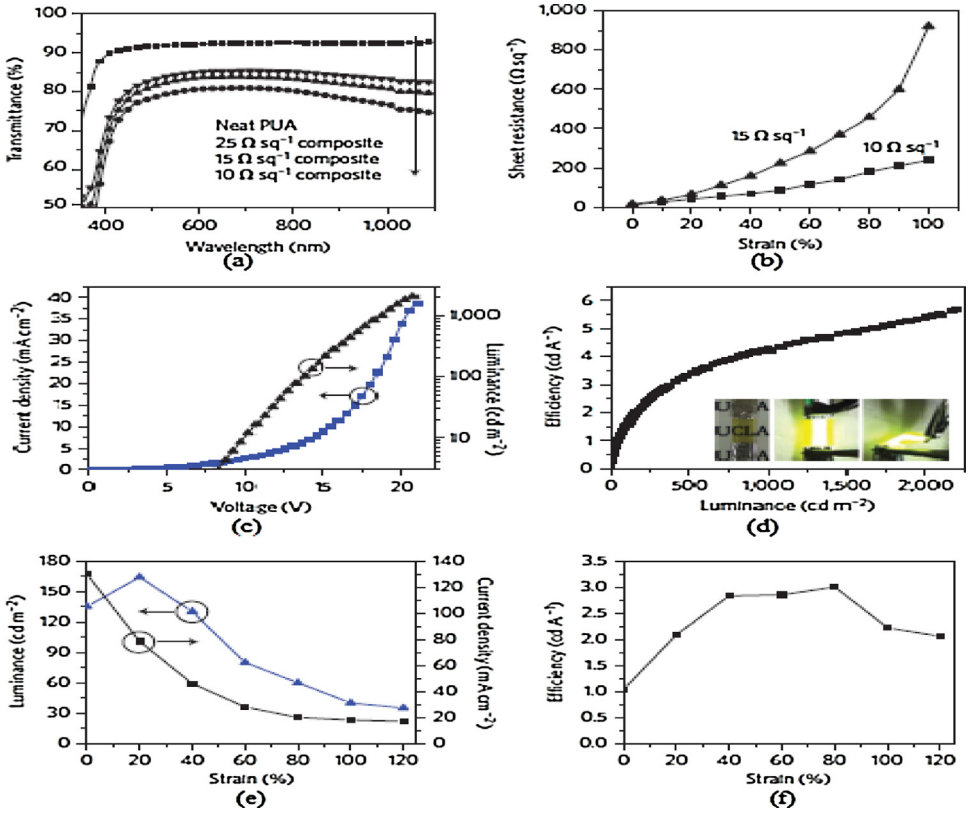

Fig. 4. (a) Transmittance spectra of a neat PUA film and AgNW– PUA composite films with specified sheet resistance (thickness, 150 mm), (b) Sheet resistance for AgNW–PUA composite electrodes with increasing strain, (c) Current density–luminance–driving voltage characteristics of an elastomeric PLEC device, (d) Current efficiency–luminance characteristics of the device. Insets: photographs of the PLEC unbiased, biased at12 V, and deformed to show light emission from both surfaces, (e) Current density and luminance characteristics of a PLEC device at 12 V with increasing strain, (f) Current efficiency characteristics of the device with strain [80].
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and electrical conductivity enhancers [82]. The film exhibited a sheet resistance of <20 Ω/sq and a transmittance of >80% with high stretch-ability of the PEDOT/STEC films. Transparent conducting polymers might be better alternative of ITO as TCE. These are easily available and provide highly flexible films in comparison to ITO. PEDOT: PSS have shown conductive up to strain 20 times higher than ITO [83]. However, these polymer are still showing poor stability at high temperature and unbalanced pairing of sheet resistance and transparency individually (without forming composite) [12,23,84], but showing better result with composites. The conductive polymers require further improvement to achieve a high performance device compared to ITO and other TCE materials.

5. Metal nanowires

Metal nanowires are also considered a very competitive candidate for using as transparent conducting electrodes (TCEs), because of their high electrical and optical properties. Metal such as Cu, Ag and Au have been reported highly conductive materials and low sheet resistance material [20,85,86]. In 2007, Lee *et al.* demonstrated the potential of silver nanowire as TCEs and reported the solution - processed silver nanowire mesh electrodes exhibiting an optical transparency(>80%) similar to or higher than that of metal-oxide thin films with approximately same sheet resistance(<20 Ω/sq) [87]. The electrodes were used in organic solar cells and showed a performance similar to that of devices based on a conventional metal-oxide transparent electrode. In another works Lu *et al.* reported a Cu NW electrode layer with a sheet resistance 35 Ω /sq with a transparency of 85% [88]. Guo *et al.* reported Cu nanowire electrode layer with a sheet resistance 51.5 Ω/sq and a transparency of 93.1% [89] and Ye *et al.* reported a CuNW TCE with 100 Ω/sq with a transparency of 95% [90]. All of these researches show tradeoff between sheet resistance and transparency, but suitable for using CuNW as TCE in different optoelectronic devices. Chahwan Hwang *et al.* demonstrated spray-coating for deposition of CuNWs on glass substrate and reported a low sheet resistance of 23.1 Ω/ sq with an optical transmittance of 84.1% in visible range [91]. In the same work, spray-coated CuNW on stretchable PDMS was also reported, which showed improvement in the sheet resistance up to 4.1 Ω/sq and a decrease in optical transmittance of 70% in visible range. For improving the performance of a metal nanowire TCE, some hybrids of metal nanowire were also investigated, i.e. Preston *et al.* introduces a solution based on silver nanowire (Ag NW) paper hybrid and achieved a low cost, flexible TCE with an excellent sheet resistance of 13 Ω/sq with an optoelectronic transmittance of 91% [92]. In another work, Kang *et al.* demonstrated a new technique named capillary printing technique for depositing Ag NW solution to generate uniformly aligned NWs by controlling the nanochannels [93]. The fabricated layer of the aligned AgNW reported a transparency of 95% at a sheet resistance 20 Ω/ sq. In the same work aligned AgNW electrodes was used in Polymer light-emitting diodes (PLEDs) and polymer solar cells (PSCs). The PLEDs showed a great enhancement of 30% in maximum luminance as compared to the maximum luminance of PLEDs with random AgNWs. However, PSCs showed a power conversion efficiency (PCE) of 8.57%, the highest value as compared to the PSCs using random AgNW electrodes. In addition to these works some more have been shown in the Table 4.

Recently, a solution-processed flexible TCE of MoO3/AgNW/MoO3/TiO2/Epoxy composite were demonstrated by Yu. *et al.* [98]. In the work, the resulted layer showed a sheet resistance of 12–15 Ω/sq with a transmittance of 82% in visible range. In the fabricated composite, the AgNW/MoO3 composite network broadened the lateral conduction range due to the built of an efficient charge transport network with long-sized nanowire and could enhance the collection efficiency and effective charge transport. However, TiO_2 layer was used to prevent epoxy penetration.

In another work, a dual-scale silver nanowire (AgNW) based transparent electrode was introduced [99]. The achieved TCE exhibit a significant enhancement in the effective electrical area by filling the large percolate voids present in a long/thick AgNW network with short/thin AgNWs. By using the TCE, a flexible OLED was developed and characterized shown in the Fig. 5.The results compared to the previous mono-scale AgNWs which showed that the efficiencies of a flexible OLED based on the dual-scale AgNW electrode have been significantly increased compared to OLEDs with single sized AgNWs.

As discussed above, AgNWs are commonly used metal nanowires and have shown better results, i.e. low sheet resistance with high transparency and are considerd as a strong alternative to ITO. AgNW flexible transparent conducting electrode has shown better mechanical robustness with only 3.9 times increase of resistance under 15% tensile strain in comparison to ITO [100]. However, AGNW films also have several drawbacks, such as high surface roughness, large junction resistance, small contact area and low mechanical/chemical durability [11,101–103]. These need to be improved to use AGNW as a better replacement of ITO.

6. Graphene

Graphene is a two-dimensional monolayer of sp2 bonded hybrid carbon atoms and exhibits excellent physical and chemical properties including high elasticity, high mechanical strength, high optical transparency and good electrical conductivity [104,105]. In 2004, Novoselov *et al.* introduced graphene first time using micromechanical exfoliation or scotch-tape technique [106], the work earned 2010 Nobel Prize in Physics for them [107]. After their invention a lot of work has been done on graphene to improve or modify its properties for using in various electronic applications including transistors [108,109], photonics and optoelectronic applications. As discussed above, TCE is an essential part of various optoelectronic devices and graphene showed enough capability to use it as a TCE. In 2007, Watcharotone *et al.* demonstrated transparent and electrically conductive GO-silica composite films and discussed the effect of temperature and GO concentration on conductivity and transmittance that reported a conductivity of 0.45 S/cm of 11 wt% graphene-silica composite films [110]. After that few graphene TCEs based devices were developed by the researchers, i.e. Peter Blake *et al.* developed a graphene TCE based liquid crystal device and showed a sheet resistance of down to 400 Ω with an optical transmission of about 98% for this particular sample [111]. In another work a graphene TCE based OLED was introduced, in which the graphene film showed the sheet resistance of 800 Ω/sq with 82% of a transmittance at 550 nm [112]. The device started with a turn-on voltage of 4.5 V and showed a luminance of 300 cd/m^2 on applying a voltage of 11.7 V. However, the sheet resistance of a graphene electrode was still high and needed more improvement for achieving high performance of the devices. 1n 2009, Cai *et al.* reported a crackles transfer technique of CVD fabricated graphene film and achieved a sheet resistance of 219Ω/sq and a transmittance of 96.5% [113]. In this work a single-layer graphene was transferred onto polymethylmethacrylate (PMMA)/AB-glue/PET up to 14-inch and achieved 5 times higher conductivity of the graphene film as compared to graphene film obtained by conventional method. In another work, Geng *et al.* reported a very simple method of developing transparent conductive graphene films of chemically converted graphene (CCG) suspension and fabricated polymer solar cell devices having the prepared CCG films as TCE [114]. However, the sheet resistance was again high of an order of 10^3 Ω/sq with a transparency of 80% at

Table 4
Metal nanowires and their hybrid materials with their sheet resistance and transparency properties.

S. No.	Material	Deposition Technique	Sheet Resistance(Ω/sq)	Transparency(%)	References
1.	AgNW	Vacuum filtration, Transfer technique	10	85	[20]
2.	AgNW	Spin coating	91	94.1	[94]
3.	Ni NW	–	19	90	[23]
4.	CuNi NW	Solution processing	36	80	[81]
5.	Graphene /AgNW	CVD, Spin coating	18	91.3	[94]
6.	RGO/ AgNW	Dip coating	27	72	[95]
7.	AgNW/IZO/ PEDOT:PSS	Spin coating, Sputtering	5.9	86	[13]
8.	AgNWs/AZO	Drop casting, Atomic layer deposition (ALD)	23	83	[96]
9.	SWNT–AgNW hybrid	Spray	30	85	[97]
10.	Graphene /AgNW/Graphene	CVD, Spin coating	20	88.6	[94]

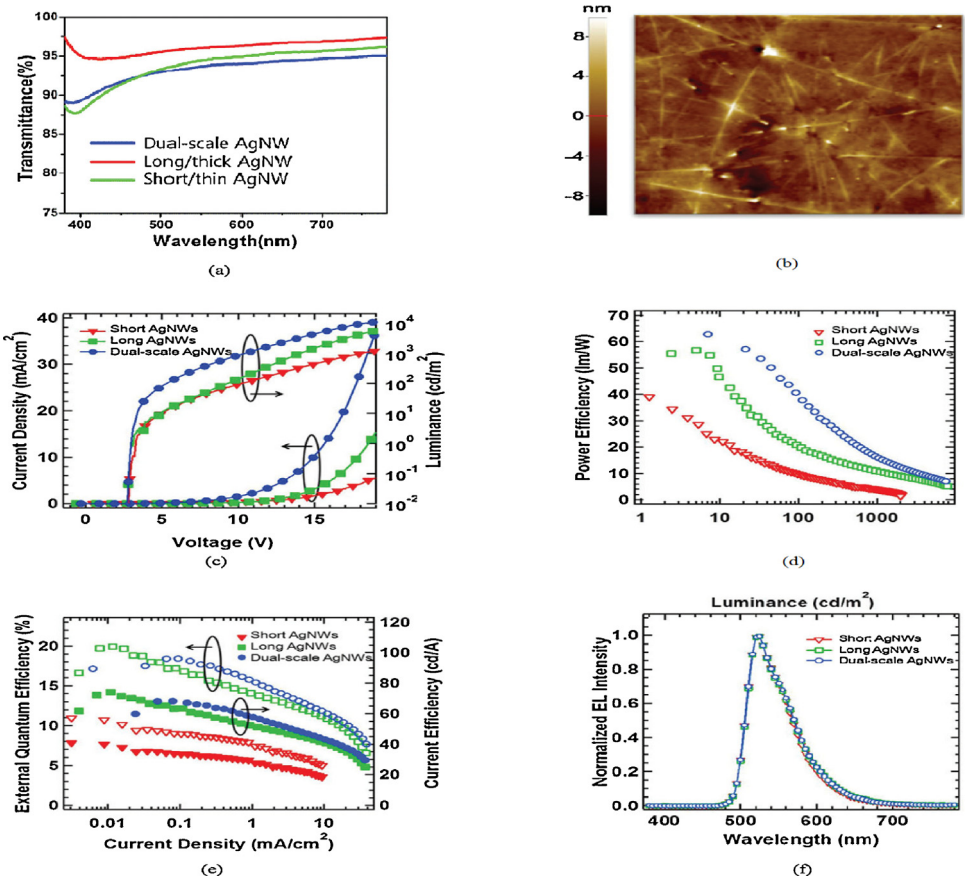

Fig. 5. (a) Spectral transmittance of dual-scale AgNWs (blue), long/thick AgNWs (red) and short/thin AgNWs (green) (b) AFM topology information of dual-scale AgNWs (c) current density and luminance (e) power efficiency, (f) external quantum efficiency (hollow symbols) and current efficiency (solid symbols) and (g) normalized electroluminescence spectra (at the current density of 25 mA cm^{-2}) of OLEDs using three different types of AgNW network transparent conductors (short AgNWs, long AgNWs, dual-scale AgNWs) [99].

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550 nm and resulted ~ 50% (half) power conversion efficiencies in comparison to the reference devices having ITO as TCE. That shows the requirement of more improvement in the material and techniques of fabrication. After that, G.X. Ni *et al.* introduces a new approach, in which they prepared a heavily doped graphene by non-volatile ferroelectric dipoles and achieved a sheet resistance of 120 Ω/sq with a 95% transmittance from the visible to the near-infrared range. In another work, a novel roll to roll production and wet-chemical doping of CVD developed graphene layer was reported by Bae *et al.* The developed film showed the sheet resistances of ~125 Ω/sq and a transmittance of 97.4% [115]. In the same work they further fabricated a doped four-layer film using layer-by-layer stacking and achieved a sheet resistance of ~30 Ω/sq and a transmittance of ~90, which showed the capability of graphene as a replacing material of ITO.

Few chemically treatment methods and hybrids of graphene were also tried for improving sheet resistance of graphene and showed better results, i.e. Khrapach *et al.* reported intercalation with $FeCl_3$ of a mechanically exfoliated few-layer graphene by a heavy charge doping up to 9×10^{14} cm^{-2} and showed an electrical resistivity of < 8 Ω/sq with a transparency of 84% in visible range [116]. In another work, Voronin *et al.* reported high conductive and transparent film of hybrid reduced graphene oxide (RGO)/Ag quasi-periodic mesh and formed AG qp-mesh by magnetron sputtering of

Table 5
Graphene and its derivatives with their sheet resistance and transparency properties.

S. No.	Material	Deposition Technique	Sheet Resistance(Ω /sq)	Transparency(%)	References
1.	Graphene	CVD	280	80	[14]
2.	Graphene	Inkjet printing	260	86	[25]
3.	Graphene	Transfer layer by layer	40	89.5	[104]
4.	RGO	LB	1100	91	[118]
5.	Graphene-CNT Hybrid	Spin coating	240	86	[63]
6.	Graphene-PEDOT:PSS Hybrid	Spin coating	180	90.9	[84]
7.	Graphene coated Cu NT	CVD	23.2	83.4	[119]
8.	Graphene silver Hybrid	Hot-pressing process	14	90	[120]
9.	Graphene – Metal Nanowire	Spin coating, CVD	33	94	[23]
10.	RGO-Au Grid	Spin coating	18	80	[121]

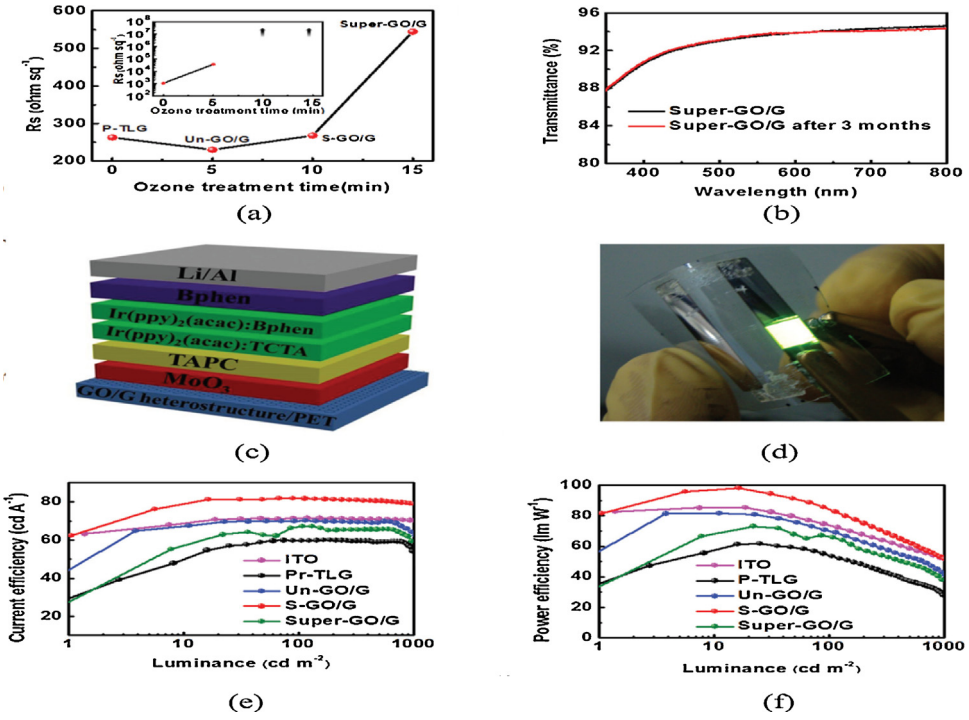

Fig. 6. (a) Sheet resistance of a three-layer and monolayer graphene film (inset) with different ozone treatment times. After 10 min ozone treatment, the resistance of the monolayer graphene is out of our measurement range (106 OΩ/sq), and the material becomes insulating. (b) Optical transmittance change of GO/G heterostructure TCEs after 3 months, (c) OLED structure, (d) Optical image of operated flexible OLEDs with GO/G heterostructure anodes, (e) current efficiency *vs.* luminance characteristics (f) power efficiency vs luminance characteristics of different electrode OLEDs [123].
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Table 6
Comparative study of Transparent Conducting Electrodes.

S. No.	TCEs	Deposition Technique	Sheet Resistance(Ω/sq)	Transparency(%)	Flexibility
1	ITO	Sputtering	5	>80	low
2	Alternate TCOs (ZnO:Al)	Sputtering	7.65	91.2	low
3	Carbon Nanotubes (SWCNT)	Dip Coating	100	90	very high
4	Metal Nanowires (AgNW)	Vacuum filtration, Transfer technique	10	85	medium
5	Conducting Polymer (PEDOT:PSS)	Spin coating, dip – drop method	39	80	very high
6	Graphene	Transfer layer by layer	40	89.5	very high

Ag and coated the RGO film by spray technique in NaBH4 solution. A low sheet resistance of 12.3 Ω/sq and a high transparency of 82.2% was achieved [117]. Low processing cost and technology scalability are main advantages showed by the proposed method. In addition of these few more Graphene TCFs with their sheet resistance and transparency in visible range are given in Table 5.

In 2016, Zhang *et al.* introduced a new hybrid approach and developed highly stable transparent electrodes having biocompatible polylactic acid (PLA) sandwiched between graphene and silver nanowires (AgNWs) [122]. Introduction of a PLA layer enhanced the surface morphology and mechanical flexibility of a hybrid film without affecting the optical and electrical properties. The sheet resistance of 13.6 Ω/sq and a transmittance of 84% were reported, and demonstrated strong adhesive and highly stable transparent conducting electrodes in flexible optoelectronic devices. In another work, a graphene oxide/graphene (GO/G) vertical heterostructure

was developed and used as TCE in OLEDs [123]. The GO/G heterostructure electrodes showed high optical transmittance, large work function, high stability, and good compatibility with MoO_3 layer, which was used as HIL layer in the fabricated OLED. Green OLEDs with GO/G heterostructure anodes show the high current efficiency and power efficiency as compare to those based on pristine graphene (ITO) shown in Fig. 6.

As discussed above, Graphene can be preferable replacement of ITO as TCE as it has a good combi nation of sheet resistance and transparency. By Yanwu Zhu *et al*. reported graphene film with a tensile strain of 6.5% having small increase in resistance and showed that the original resistance could be restored after a tensile strain of 18.7% which is very high in comparison to ITO [124]. Since it is also preferable TCE for flexuble devices. However, there are several factors that could limit the use of graphene. One of them is a limited production volume of graphene. Even there are a number of methods for synthesis of various forms of graphene, but none of them is suitable for volume production. Cost, is another important factor which hinder its use. Although cost has come down significantly since the first commercial appearance of graphene in the market. Still, the initial material cost is high that also is a drawbacks of graphene.

7. Conclusion

In this work, a comprehensive overview on recent developments and achievements in transparent conducting electrodes is presented. The conventional TCE (ITO) plays a significant role in many optoelectronic applications, but brittle nature of ITO and scarcity of indium leads to the need of alternate TCEs to improve the performance such as stretch- ability, flexibility and to solve the problem due to scarcity of indium. A brief comparison of the studies material in Table 6.

As discussed above, ZnO is easily available and doped ZnO (i.e., ZnO:Al) is the most preferred material among all TCOs for replacing ITO. However, ZnO:Al is not so flexible independently and has poor stability in oxidizing environments and also in etchability when these are in contact with acids. Carbon nanotube is another promising candidate as TCE due to its good electrical properties, mechanical properties, exceptional flexibility and proper work function. However, it shows high sheet resistance and also have high roughness that affects the carrier mobility and shorten the lifetime of the device. CNT needs some more improvement to make place as a better TCE. In matal nanowires, AgNW is the most preferred material that can be used as TCEs. Depending on the sheet resistance, transparency and flexibility, AgNWs can be a better TCE. Despite of this AGNW films have several drawbacks, such as high surface roughness, large junction resistance, small contact area, and low mechanical/chemical durability. These need to be improved to use AGNW as a better replacement of ITO. In the case of conducting polymer PEDOT:PSS attracts the attention for using as a TCE, But its poor stability at high temperature and unbalanced pairing of sheet resistance and transparency makes less desirable for TCE applications. By comparing all the above mentioned TCEs, graphene can be considered as the best alternate of ITO for TCE, because of its high flexibility, high conductivity, high transparency and easy process. In some devices graphene have been used as TCE, but some more efforts like bulk production and more easy synthesis process are required that can make it first choice of light and display industries for using as TCEs in their devices.

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