

Review

# Recent Progress in Transparent Conductive Materials for Photovoltaics

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**Abstract:** Transparent conducting materials (TCMs) are essential components for a variety of optoelectronic devices, such as photovoltaics, displays and touch screens. In recent years, extensive efforts have been made to develop TCMs with both high electrical conductivity and optical transmittance. Based on material types, they can be mainly categorized into the following classes: metal oxides, metal nanowire networks, carbon-material-based TCMs (graphene and carbon nanotube networks) and conjugated conductive polymers (PEDOT:PSS). This review will discuss the fundamental electrical and optical properties, typical fabrication methods and the applications in solar cells for each class of TCMs and highlight the current challenges and potential future research directions.

**Keywords:** transparent conducting materials; solar cells; optoelectronics; semiconductors; photovoltaics; graphene; carbon nanotube; metal nanowire network



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## 1. Introduction

Transparent conducting materials (TCMs) are a unique class of materials that exhibit excellent optical transmission in the visible light spectrum coupled with very high conductivity at room temperature. Materials such as metals that show high conductivity at room temperature remain opaque in the visible range. On the other hand, materials such as glass and intrinsic metal oxides/sulfides (ZnO, ZnS, CdO, SnO<sub>2</sub>) show excellent transmittance in the visible light range but exhibit insulating behaviour at room temperature.

It is quite difficult to synthesize a material which retains transparency while becoming electrically conductive. In general, a wide bandgap ( $E_g > 3$  eV) insulator is selected as a host material and modification in its electronic structure is realized from the numerous lattice defects to obtain conductivity. These defects are electrons and positive holes, excitons, vacant lattice sites/interstitial atoms, impurity in interstitial or substitutional positions, dislocations and stacking faults. Modification of the electronic structure with vacancies, substitutional and interstitial site defects by aliovalent doping plays a very important role in determining the conductivity. Understanding the effect of these defects led to the realization of TCMs.

Another common concept for realizing a TCM is ultra-thin metal films (UTMFs). To synthesize UTMFs, metal films can be made transparent by thinning down to a nm scale, but their sheet resistivity increases significantly as compared to the bulk material. When the electron mean free path is of the order of the film thickness, additional scattering at the surface causes a significant increase in the sheet resistance [1]. Scattering at the grain boundary in polycrystalline films also contributes to increasing the sheet resistance of the thin metal films [2]. Due to these shortcomings, UTMFs can not be effectively utilized as transparent conductors. More recent research studies focus on enhancing the performance of a TCM by understanding the microscopic nature of conducting the process in order to

analyze the role of morphology, crystal structure, and bonding on charge transport. After the realization of these types of materials, a large amount of research and development has been going on to make them more conductive as well as transparent in order to commercialize them.

There exist a wide range of TCMs such as ITO, doped ZnO (Al:ZnO, In:ZnO, Ga:ZnO), F-doped SnO<sub>2</sub> (FTO) and amorphous InGaZnO<sub>4</sub> (IGZO) for today's optoelectronic applications [3–9]. Extensive work has been carried out to find new alternative materials for TCMs, in particular, to replace existing TCMs and to explore viable *p*-type TCM. Additionally, a significant amount of work is being conducted on metal nanowire meshes, carbon nanotubes (CNTs) and graphene for TCM applications [10–13]. A uniform and conformal thin film deposited at low temperatures is essential for TCMs. Several vacuum and non-vacuum approaches such as sputtering, pulsed laser deposition (PLD), atomic layer deposition (ALD), chemical bath deposition (CBD), spray pyrolysis, spin coating and thermal evaporation can be used to obtain high-quality thin films. Deposition parameters and film composition (doping or alloying) play a vital role in determining the optical as well as electrical properties of the TCMs. Optimization of these parameters can lead to a high-performance TCM.

While metal oxide-based TCMs are widely used in photovoltaics, owing to their high conductivity and transparency, the scarcity of the raw materials, high cost of vapour deposition techniques and mechanical brittleness drive researchers to look for alternative transparent electrodes [5,13,14]. That is, TCMs have comparable electrical and optical properties but can be potentially made from earth-abundant materials by scalable low-cost fabrication methods. Metal nanowire network (MNWN)-based transparent electrodes have thus attracted great attention due to the intrinsic high conductivity, tunable transparency and solution-based deposition methods [15–17]. Furthermore, the mechanical flexibility of MNWNs may open up new opportunities for the development of unconventional photovoltaics, including semitransparent solar cells and flexible solar cells [15].

Poly(3,4-ethylenedioxythiophene):polystyrene sulfonate (PEDOT:PSS) is a conductive polymer which consists of positively charged conductive PEDOT and negatively charged insulating PSS [18]. PSS serves the purposes of charge balancing and dispersion in different solvents [19]. PEDOT:PSS has been widely used in optoelectronic devices as a hole transporting layer or a transparent electrode owing to its high and tunable conductivity ( $10^{-2}$ – $10^3$  S cm<sup>-1</sup>), transparency (~90% with a film thickness of 100 nm), good film quality (roughness ~5 nm), scalability (solution processable), high mechanical flexibility and air stability [18,20–22].

One of the goals in the development and research of emerging thin-film solar cells is to produce the most energy consumption from low-cost materials with high efficiencies and scalable fabrication processes [23–26]. More specifically, perovskite solar cells (PSCs) have emerged as a promising candidate with the use of lead halide perovskite (LHP) as the light harvester in solar cells [27–29]. The cubic cell unit of LHPs can be broken down into the following:

ABX<sub>3</sub> (general cubic cell unit)

MABX<sub>3</sub>

(CH<sub>3</sub>NH<sub>3</sub>)<sup>+</sup>BX<sub>3</sub>

where MA = (CH<sub>3</sub>NH<sub>3</sub>)<sup>+</sup>; B = Pb or Sn; X = Cl, Br or I.

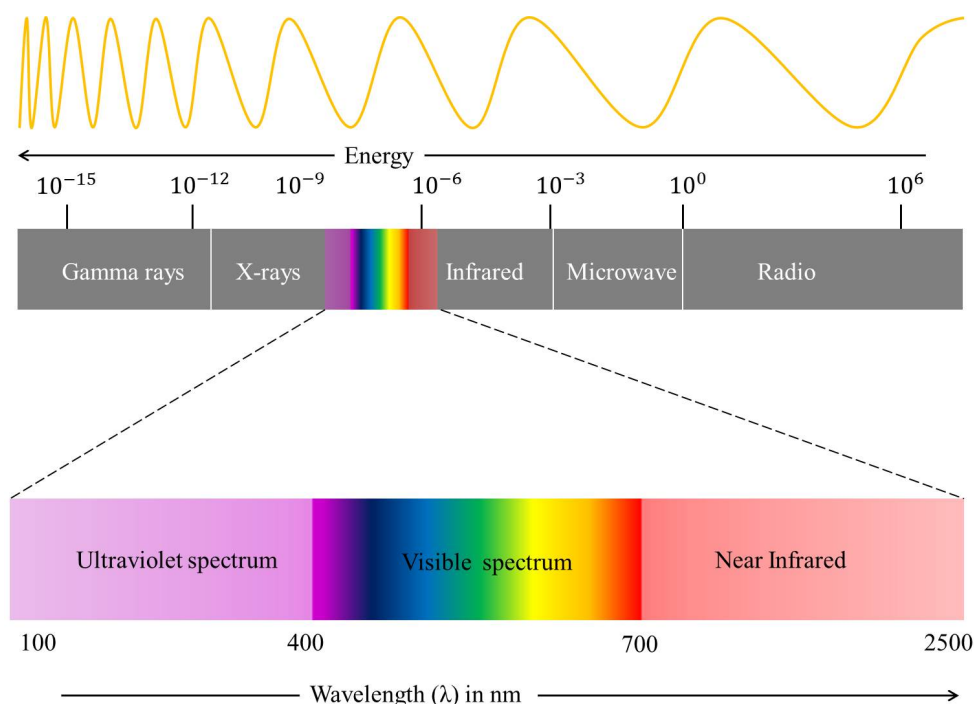
Moreover, LHPs achieve a power conversion efficiency (PCE) as high as 31% [30]. In recent years, PSCs have received more attention in the photovoltaic community because of their low cost, tunable bandgap, high carrier mobilities and rapid improvement in their power conversion efficiency [30–32]. Studies show an impressive and high-power conversion efficiency of above 14.1% and up to 25.27 within a decade [23,26]. Unfortunately, metal electrodes (e.g., Au, Ag and Al), and transparent conducting oxides (TCOs) (e.g., indium tin oxide (ITO), indium zinc oxide (IZO)) used in PSCs have high costs due to the complex processing of TCOs and are limited in their commercial applications due to the limited supply of metals (e.g., indium) [32,33]. Additionally, the perovskite material degrades in

the presence of water, which leads to the formation of hydrated phases  $\text{CH}_3\text{NH}_3\text{PbI}_3 \cdot \text{H}_2\text{O}$  and  $(\text{CH}_3\text{NH}_3)_4\text{PbI}_6 \cdot 2\text{H}_2\text{O}$  [34].

Moreover, metal-based electrodes such as Al, Ag and Au are not ideal materials in the long run due to the finite supply of them. Al is easily oxidized in air, which causes a dramatic decrease in conductivity. Ag is not stable due to the formation of an Ag-halide that degrades device performance [35,36]. Similarly, Au also reacts with halides (iodide) ions, has high reflectivity and is expensive [35,37]. Additionally, TCOs have been employed in standard PSCs as well as flexible perovskite solar cells (FPSCs) due to the low-temperature fabrication process, chemical stability, high optical transparency, low sheet resistance and established mass production process [35,38]. However, ITO is rigid and brittle. This will likely result in breakage and degradation of the devices under mechanical deformations for flexible device applications. Indium is also scarce, and the costly fabrication process (e.g., vacuum deposition techniques such as sputtering and pulsed laser deposition) makes it the most expensive component in PSCs/FPSCs; it comprises 50–60% of the total material cost [24,38,39]. An alternative to the high cost and limited supply of metallic electrodes and TCMs is to use carbon-based materials to replace standard electrodes. This is largely beneficial because carbon is a common organic material that is abundantly available on earth. It is known for its electrochemical activity and conductivity, resistance to water and flexibility when processed [39].

## 2. Physics of TCMs

TCMs have to be optically passive and favourably transparent. Figure 1 shows electromagnetic spectrum with favourable transmission window for TCMs.



**Figure 1.** Electromagnetic spectrum according to energy along with ideal transmission window for TCMs.

Knowledge of classical and quantum descriptions of solid-state materials is required to understand the carrier transport mechanism in transparent conducting films. The infrared reflectance part can be perfectly described using the classical electron sea analogy, whereas band-to-band absorbance in the UV can be explained through the quantum description. These materials are transparent due to their wide band gap ( $E_g > 3$  eV) and conducting due to excessive doping ( $\sim 10^{21} \text{ cm}^{-3}$ ). Carrier transport in these materials can be understood

easily if we treat them as metals having plasma frequency in the IR range and transparent in the visible range.

In 1924, H. Mott-Smith derived the accurate formula for the characteristic plasma frequency [40–42]

$$\omega_p = \sqrt{\frac{4\pi n e^2}{m}}, \quad (1)$$

where  $n$  is the carrier density,  $e$  is the charge of an electron and  $m$  is the mass of an electron. This frequency represents the most fundamental parameter of plasma. This frequency acts as a filter for all electromagnetic waves. Electromagnetic waves with a lower frequency than plasma frequency will not be able to pass through.

Drude theory helps in further understanding of plasma frequency which is highly useful in improving the electrical and optical properties of TCMs. The plasma dielectric function is given by the Drude formula [43]

$$\epsilon(\omega)_p = 1 - \frac{\omega_p^2}{\omega^2}, \quad (2)$$

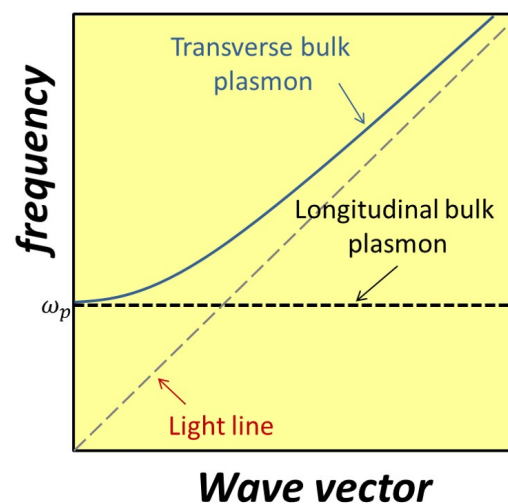
this formula can be combined with transverse wave propagating through uniform medium [41,42]

$$\omega = \frac{kc}{\sqrt{\epsilon(\omega)}}, \quad (3)$$

gives

$$\omega^2 = \omega_p^2 + c^2 k^2, \quad (4)$$

where  $c$  is the speed of light and  $k$  the wavevector. This is the dispersion relation of a transverse mode that can propagate through the transverse bulk plasmon. Figure 2 clearly shows that no other modes can propagate through the medium with frequencies less than the plasma frequency.



**Figure 2.** Photon and plasmon dispersion.

This theory suggests that films fabricated with either higher or lower permittivity can have varying optical properties. This will provide an advantage in fabricating devices such as organic light emitting diodes (LEDs) and solar cells, where the work function of TCMs plays critical role in device performance.

The second requirement of TCM is its high conductivity, as presented below

$$\sigma = ne\mu, \quad (5)$$

where  $\mu$  is the carrier mobility. Now combining Equations (1) and (5) gives following

relation

$$\sigma = \frac{\omega_p^2 \mu m}{4\pi e}, \quad (6)$$

From the equation it is quite evident that the high conductivity in a TCM can be obtained first by setting plasma frequency  $\omega_p = \omega_{pmax}$ , and increasing mobility ( $\mu$ ). Equation (6) can be re-written in terms of sheet resistance ( $R_{sh}$ ) as following

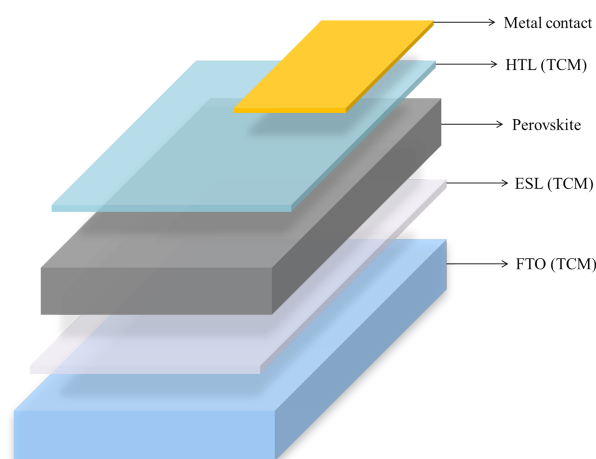
$$\omega_p = 2\sqrt{\frac{\pi e}{m\mu t}} R_{sh}^{-1/2}, \quad (7)$$

where  $t$  is the film thickness.

Increasing the conductivity from  $100 \Omega\text{-sq}^{-1}$  to  $5 \Omega\text{-sq}^{-1}$  results in a transmittance drop in the near-infrared (NIR) range. This is primarily due to free electrons changing the infrared plasma frequency. Additionally, in order to achieve n-type conductivity in wide-band gap metal oxides, the material should have a relatively high electron affinity and low CBM. For example,  $\text{SnO}_2$  with a band gap of  $\sim 3.6$  eV and electron affinity of  $\approx 4.8$  eV would be an excellent candidate for fabricating a TCM [3,44,45]. On the other hand,  $\text{HfO}_2$  with a wide band gap of 6 eV and electron affinity of only 2.5 eV will not be a suitable material for TCM. Further, in oxide semiconductors, this gives an assertion that the deficiency of oxygen in certain metal oxides can induce a shallow defect level near CBM that leads to  $n$ -type conductivity at room temperature.

### 3. Transparent Conducting Materials

TCMs are an integral part of solar cells and are used as hole transport and electron transport layers along with a window to allow light through to the PV absorber layer, as shown in Figure 3. The TCMs are generally used as an interfacial layer to improve the contact resistance between the PV absorber layer and a metallic reflector, improving the refractive index matching. The high conversion efficiency in solar cells such as micro-crystalline, Si, perovskite, CIGS, CZTS and various multi-junction requires minimal sheet resistance of  $\leq 10 \Omega \text{ sq}^{-1}$  and transparency  $> 80\%$  of TCMs in wide spectrum range (400–1300 nm). The following sections will outline all possible TCMs that can be used as a hole transport layer (HTL), electron transport layer (ETL) and back contact in solar cells.



**Figure 3.** A typical schematic of a solar cell using both n- and p-type TCMs. The cells consist of metal contacts, a hole transport layer (HTL), an absorber layer, and an electron transport layer (ETL).

#### 3.1. n-Type Transparent Conductors

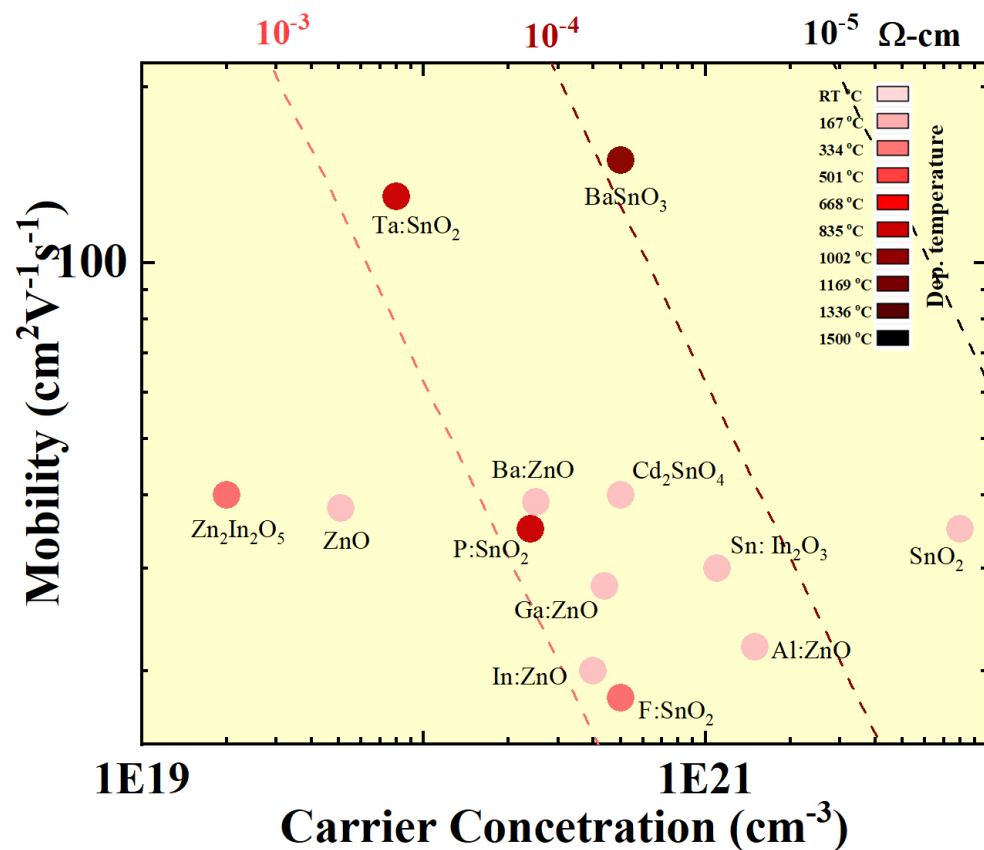
Initially,  $n$ -type TCMs were developed by oxidation of thin metal films in a reactive atmosphere ( $\text{O}_2$  and Ar). The metal thin films were deposited via evaporation and DC sputtering [46]. The resistivity of thin films, fabricated by oxidation of thin metal films,

was significantly high compared to bulk materials. This was because the electron mean free path was of the order of film thickness, also scattering at the grain boundaries in the polycrystalline film plays a major role in reducing the conductivity of the film [1]. These two major effects limited the conductivity in thin polycrystalline films [2]. Other methods to deposit TCMs were via spraying or dipping. After the early discovery of TCMs, significant improvement in the performance has been reported in a number of binary, secondary, ternary and quaternary compounds (metal oxides). These reported metal oxide compounds show appreciable conductivities and good transmission in the visible light spectrum but conductivity in these films is still quite low compared to that of metals. In order to facilitate *n*-type conductivity in metal oxides, various lattice defects such as oxygen vacancies ( $V_O^{\bullet\bullet}$ ), cation interstitials and different substitutional defects have been introduced to effectively create shallow defect sites near conduction band minima. Moreover, some metal oxides such as ZnO are naturally *n*-type in nature due to the presence of different defects. The presence of these defect levels helps electrons to directly inject into the conduction band from nearby defect donor levels. Further increase in doping leads to the delocalization of electrons from defects sites such that the electronic state at the CBM becomes filled resulting in a shift in the Fermi level above the CBM. This phenomenon is known as the Burstein–Moss shift, which effectively increases the optical band gap of the material. The band theory of solids provided an explanation of doping in semiconductors and an understanding of the properties of such oxide materials.

The first *n*-type TCM was investigated by Badeker in 1907. He fabricated cadmium oxide (CdO) via incomplete thermal oxidation (in air) of a sputtered Cd metal thin film and reported a resistivity as low as  $1.2 \times 10^{-3} \Omega \text{ cm}$ . Further, various *n*-type TCMs were fabricated, patented and commercialized in the early 1940s. An example of an SnO<sub>2</sub>-based *n*-type TCM was developed and patented in the 1930s for its use as an airplane windshield de-icer. The best-known TCM to date, *n*-type Sn-doped indium oxide (ITO), was also developed and patented by USA based company named Corning in 1951. ITO has been the most important part of the TCM industry to date. High-performing ITO films have been developed via physical vapour deposition (PVD), atomic layer deposition (ALD), chemical vapour deposition (CVD), spray pyrolysis, spin coating, sol–gel method, molecular beam epitaxy (MBE), etc. [47]. The reported minimum resistivity of impurity-doped In<sub>2</sub>O<sub>3</sub> has been unchanged for more than 40 years, which is  $1\text{--}2 \times 10^{-4} \Omega \text{ cm}$ .

In the late 1970s, CdO was further investigated for its application as a *n*-type TCM. CdO and CdO-based ternary compounds (Cd<sub>2</sub>SnO<sub>4</sub> CTO) have shown to be good TCMs with excellent transparency in the visible light range and resistivity  $<10^{-4} \Omega \text{ cm}$  at room temperature. However, CTO couldn't be used as a TCM due to the presence of toxic Cd and high processing temperature ( $>600 \text{ }^\circ\text{C}$ ). Additionally, it is quite difficult to achieve a single-phase material with a CdO-based compound [48–50]. CdO-based TCMs are very favourable to be used in II–VI compounds (CdTe and Cd(S, Te)).

Successful development of high conducting oxygen-deficient ZnO and SnO<sub>2</sub>, doped Zn/Sn oxides (M:Zn/Sn-O where M: Al, B, Ga, In, F, Sb, Ta, P) and ternary metal oxide compounds (ZnSnO<sub>3</sub>, Zn<sub>2</sub>SnO<sub>4</sub>, Zn<sub>m</sub>In<sub>2</sub>O<sub>3+m</sub>;  $m = 2\text{--}7$ , MgIn<sub>2</sub>O<sub>4</sub>, GaInO<sub>3</sub>, (GaIn)<sub>2</sub>O<sub>3</sub>, In<sub>4</sub>Sn<sub>3</sub>O<sub>12</sub>) with excellent transparency in visible light range expands the horizon of applicability of TCMs in various optoelectronic devices [51,52]. These materials also showed conductivities  $>5000 \text{ S cm}^{-1}$  with mobilities of more than  $30 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ . Further, multi-component oxides such as ZnO–SnO<sub>2</sub> and ZnO–In<sub>2</sub>O<sub>3</sub> were extensively studied for their potential application in thin film solar cell devices. SnO<sub>2</sub> and ZnO-based TCMs are frequently used in compound semiconductors of the III–V type compounds (GaAs, (Al,Ga)As, InP and (In,Ga)P). Figure 4 shows some of the best known *n*-type TCMs reported to date. Table 1 shows most of the *n*-type TCMs with deposition methods and optoelectronic properties.



**Figure 4.** Mobility is plotted against carrier concentration for the highest performing *n*-type TCMs. Datapoints present the obtained mobility for the corresponding carrier concentration. The colour of the circles signifies the deposition temperature of the film. Dotted lines represent the line of equal resistivity.

**Table 1.** List of some *n*-type TCMs with deposition methods and optoelectronic properties.

Material	Deposition Method	Deposition Temperature	Rsh ( $\Omega/\square$ )	$\sigma$ ( $S\text{ cm}^{-1}$ )	$\mu$ ( $\text{cm}^2\text{V}^{-1}\text{s}^{-1}$ )	$n$ ( $\text{cm}^{-3}$ )	Average Optical Transmisson	Thickness (nm)
Sn:In <sub>2</sub> O <sub>3</sub> [47]	EBE	~150	<20	>5000	30	$1.1 \times 10^{21}$	>85%	300
ZnO [51]	Sputter	~150	<20	>500	38	$5.1 \times 10^{19}$	>85%	550
Al:ZnO [51]	Sputter	~150	<20	>5000	22	$1.5 \times 10^{21}$	>85%	425
B:ZnO [51]	Sputter	~150	<20	>1500	39	$2.5 \times 10^{20}$	>85%	595
Ga:ZnO [51]	Sputter	~150	<20	>2000	28	$4.4 \times 10^{20}$	>85%	500
In:ZnO [51]	Sputter	~150	<20	>1200	20	$4.0 \times 10^{20}$	>85%	650
SnO <sub>2</sub> [53]	SP	450	<20	>800	>30	$8 \times 10^{21}$	>80%	720
F:SnO <sub>2</sub> [54]	SP	380	<20	>1000	18	$5 \times 10^{20}$	>80%	210
P:SnO <sub>2</sub> [55]	CVD	550	<20	>1500	35	$2.4 \times 10^{20}$	>80%	400
Sb:SnO <sub>2</sub> [56]	Sol-gel	350	<10	>10000	–	–	>60%	340
Ta:SnO <sub>2</sub> [57]	PLD	600	<20	>1500	130	$8 \times 10^{19}$	–	120
Zn <sub>2</sub> In <sub>2</sub> O <sub>5</sub> [52]	Sputter	350	<20	>2500	>30	$5 \times 10^{20}$	>80%	~400
Cd <sub>2</sub> SnO <sub>4</sub> [48]	Sputter	>150	<20	>5000	>30	$>1 \times 10^{19}$	>80%	1000
BaSnO <sub>3</sub> [58]	MBE	>1230	<20	>5000	150	$>4 \times 10^{20}$	–	31

In laser lithography and high-efficiency solar cell fabrication, new ultraviolet transparent conducting materials will be required since in the process of laser lithography, elector-conductive materials are necessary, which transmit the UV light at <250 nm. Additionally, in most of the available solar cells, the used transparent conductor has a band gap <4 eV, which leads to the elimination of photons in the UV range. The solar cell efficiency can be increased significantly if high-energy photons are utilized in power conversion. Wide band gap Ga<sub>2</sub>O<sub>3</sub> (>4 eV) had been studied for its potential application in solar cells

and lasers.  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> has shown conductivity  $\sim 30 \text{ S cm}^{-1}$  and transparency  $>80\%$  in the visible light range at room temperature [59].

Wide band gap doped/un-doped ternary oxide with perovskite structured materials such as BaSnO<sub>3</sub> and SrTiO<sub>3</sub> have been studied for potential transparent conductor application [58,60]. SrTiO<sub>3</sub> has shown a mobility below  $10 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$  and conductivity  $\approx 500 \text{ S cm}^{-1}$  at room temperature [61]. MBE-grown BaSnO<sub>3</sub> on PrScO<sub>3</sub> exhibited mobility  $\approx 150 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$  with a carrier concentration of  $7.2 \times 10^{19} \text{ cm}^{-3}$  [62]. Recently, La-doped BaSnO<sub>3</sub> grown on SrTiO<sub>3</sub> (001) substrate shows a room temperature carrier concentration of  $4 \times 10^{20} \text{ cm}^{-3}$  with mobility of  $\approx 150 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$  [58].

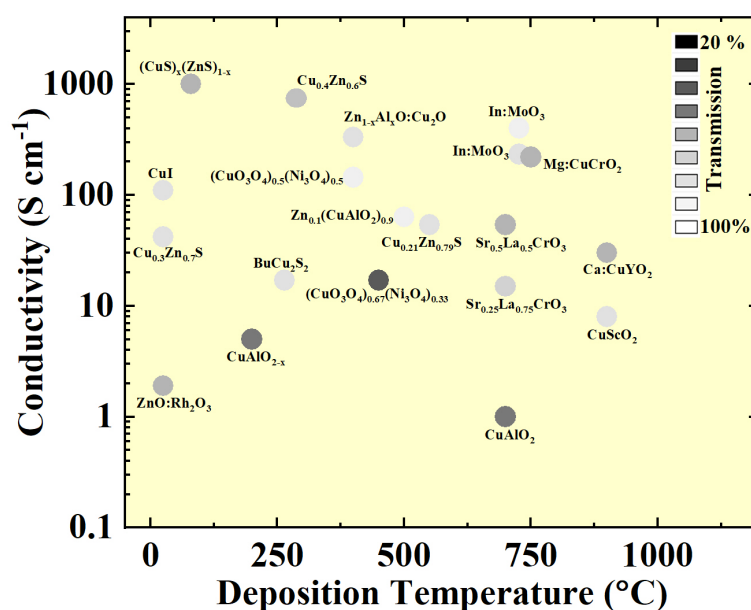
### 3.2. *p*-Type Transparent Conductors

As discussed in the last section, wide band gap oxide materials such as FTO, AZO and ITO are some of well known *n*-type TCMs and are widely used in optoelectronic devices such as LEDs, solar photovoltaics (PV), etc. However, the counterpart *p*-type TCMs are comparatively less investigated [63]. In general, *n*-type TCMs are metal oxides in nature. However, it is very challenging to fabricate oxide *p*-type TCMs as there are several fundamental complexities associated with it. The electronic structures of such metal oxides are one of the leading causes, as proposed by Kawazoe et al. [64]. The *p*-type metal oxides show strong localization behaviour of positive holes at the edge of VB. These localized state forms due to the ionicity of metallic oxide, resulting in strong localization of holes at oxygen *2p* levels, which is in the upper edge of VB. The strong localization arises due to non-stoichiometry or substitutional doping in the material. The localized oxygen *2p* levels help in the formation of deep acceptor defect levels, far from the valence orbit of metallic atoms. This gives rise to the localization of holes which results in poor conductivity and hole mobility as holes have to overcome large energy barriers in order to move in the crystal lattice. Kawazoe et al. proposed the modification of VB to overcome the shortcoming of deep localization of holes. It can be performed by mixing orbitals with suitable cations that have enough filled energy level comparable to the oxygen *2p* level [64]. To reduce the strong localization of holes in oxygen ions due to high electronegativity of oxygen, modulation of the top of the valence band was performed by chemical bonds between oxygen *2p*<sup>6</sup> and Cu *3d*<sup>10</sup> orbitals. This particular method is called “chemical modulation of the valence band (CMVB)” [63]. Additionally, higher effective mass of holes ( $m_e < m_h$ ) in metal oxide compounds also contributes in lowering overall mobility of the material [65]. Additionally, it is desirable that the materials contain highly electropositive ions to shift conduction band towards high energy levels.

The first *p*-type transparent conductor, CuAlO<sub>2</sub> crystallizing in the delafossite structure, was reported in 1997 by Kawazoe et al. and possesses a hole conductivity of  $\approx 1 \text{ S cm}^{-1}$  with carrier concentration of  $1.3 \times 10^{17} \text{ cm}^{-3}$  and hall mobility of the positive holes of  $10.4 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$  at room temperature [66]. Cation vacancy and interstitial oxygen lead to excess oxygen in the CuAlO<sub>2</sub> delafossite phase, resulting in *p*-type conductivity. These films were developed via solid-state reaction process at 700 °C in an oxygen-rich environment. Elevated processing temperature and poor optoelectric properties limited the application of CuAlO<sub>2</sub> in different optoelectronic devices. Further, several CuM<sub>x</sub>O<sub>2</sub> (where  $M = \text{Cr, Sc, Y, Fe, Co, Rh, Ga, Ca, Mg, etc.}$ ) compounds isostructural with CuAlO<sub>2</sub> were studied for a potential *p*-type transparent conductor [67–70]. Initial reports of delafossite CuScO<sub>2</sub>, CuGaO<sub>2</sub> and CuY<sub>1-x</sub>Ca<sub>x</sub>O<sub>2</sub> films showed conductivity in the range of 5 to 30  $\text{S cm}^{-1}$  with mobility  $\leq 10 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$  [64,67,71]. The mobilities in these films are quite low since the valence band consist of oxygen *2p* bands as lattice relaxation around holes strongly traps the potential carriers. To achieve higher mobility, the valence band should consist of cation *d* or *s* band instead of oxygen *2p* bands. Later studies on delafossite structure were focused on CuCrO<sub>2</sub> based compounds as *d-d* transitions possibly resulted in higher mobility. One such film, CuCr<sub>1-x</sub>Mg<sub>x</sub>O<sub>2</sub>, showed conductivity  $>200 \text{ S cm}^{-1}$  and mobility  $>10 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ . However, the overall transparency in the visible light range was still  $<50\%$  [70]. Table 2 shows most of the *p*-type TCMs with deposition methods and optoelectronic properties.



The idea of band modulation is not limited only to oxide materials. Other chalcogenides (sulphides in particular) and mixed chalcogenides (O, S, Se combined) were also reported for their excellent optoelectronic properties to be used as *p*-type TCMs. Among all, oxysulphides and sulphides are the most superior compounds due to their transparency in the visible light region. The inclusion of any alkali and rare earth ions also showed moderate dispersion at the top of the valence band resulting in better transparency. Ueda et al. reported one such layered oxysulphide (Sr-doped LaCuOS), which showed excellent transparency in visible light range but conductivity remained  $\approx 0.3 \text{ S cm}^{-1}$  at room temperature [72]. Further, Hiramatsu et al. reported elevated carrier concentration ( $10^{21} \text{ cm}^{-3}$ ) with hole mobility of  $\approx 10 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  in layered Mg-doped LaCuOSe [73]. Further, several sulphide and iodide compounds such as  $\text{CuAl}_{1-x}\text{Zn}_x\text{S}_y$ ,  $\text{BaCu}_2\text{S}_2$ ,  $\text{Cu}_x\text{Zn}_{1-x}\text{S}$ ,  $(\text{CuS})_x(\text{ZnS})_{1-x}$ , CuI and I-doped CuI were reported to have excellent conductivity and transparency [74–82]. Reported Cu-Zn-S and CuI compounds were fabricated either at room temperature or at temperature  $< 100 \text{ }^\circ\text{C}$ . Low deposition temperatures and the use of Earth-abundant elements in these compounds make them cost-effective as well as compatible to be used in many device applications. To date, Cu-based sulphide and iodide compounds are some of the best performing *p*-type TCMs. Recently, sputtered  $\text{Cu}_x\text{Zn}_{1-x}\text{S}$  have been successfully used as a hole selective back contact for poly-crystalline CdTe solar cells (Glass/FTO/Cd(S,O)/CdTe/ $\text{Cu}_x\text{Zn}_{1-x}\text{S}$ /Au) [83]. Figure 5 shows a number of commonly known *p*-type TCMs. These TCMs have been presented in the figure by plotting the conductivity of the films against deposition temperature. Secondary information has been added as a transmission for the films in the visible light range (at  $\approx 550 \text{ nm}$ ). Different films with similar conductivities show varying transmissions, suggesting the varying performance of TCMs. This interactive figure helps us to identify better TCM, which can be deposited at lower temperatures and has higher conductivity with a fair transmission in the visible light spectrum. Further, from Figure 5 we can observe that most of the *p*-type TCMs are fabricated at high temperatures ( $> 400 \text{ }^\circ\text{C}$ ). The doped and undoped Cu-based compounds constitute the typical materials that have been studied. These materials show good transmittance in the visible light range but show low hole mobility [65]. However, some non-oxide chalcogenide-based *p*-type transparent conductors such as Zn-doped  $\text{CuAlS}_2$  [84] and undoped  $\alpha\text{-BaCu}_2\text{S}_2$  [85] show transmittance  $> 80\%$  and higher hole mobility with appreciable conductivity of  $\approx 64 \text{ S cm}^{-1}$  and  $17 \text{ S cm}^{-1}$ , respectively.

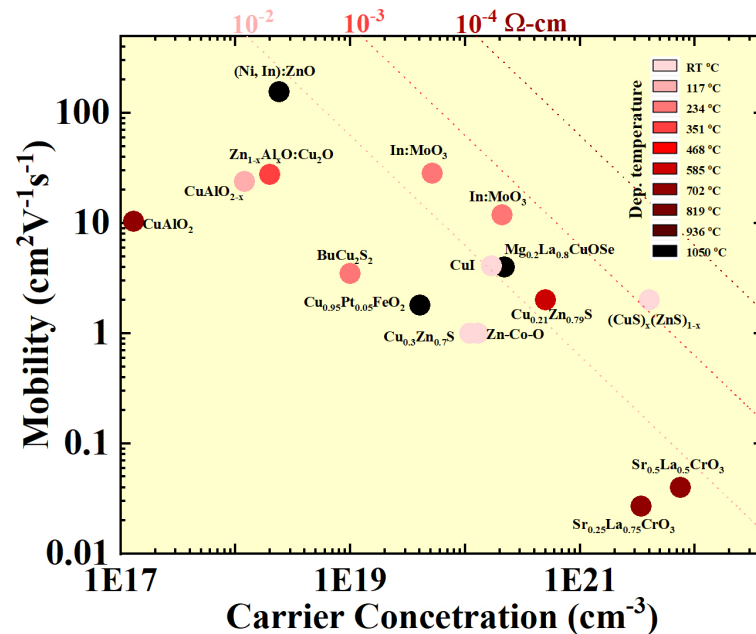


**Figure 5.** Conductivity plotted against maximum processing temperature for the highest performing *p*-type TCMs. Datapoints present in the plot give the value of maximum conductivity achieved at a given synthesis temperature. The colours of the circles represent the transmission reported at 550 nm.

**Table 2.** List of most of the *p*-type transparent conductors.

Material	Deposition Method	Deposition Temperature	Rsh ( $\Omega/\square$ )	$\sigma$ ( $S\text{ cm}^{-1}$ )	$\mu$ ( $\text{cm}^2\text{V}^{-1}\text{s}^{-1}$ )	$n$ ( $\text{cm}^{-3}$ )	Average Optical Transmisson	Thickness (nm)
BaCu <sub>2</sub> S <sub>2</sub> [85]	Sputter	264	1400	17	3.3	$1 \times 10^{19}$	~50%	430
Ba <sub>0.9</sub> K <sub>0.1</sub> CuSF [86]	SSR	650	na	82	na	na	na	na
CuAlO <sub>2</sub> [66]	Furnace	700	$2 \times 10^5$	1	10.4	$1.3 \times 10^{17}$	~35%	~500
CuAlO <sub>2-x</sub> [63]	Sputter	200	$10 \times 10^5$	5	~24	$1.2 \times 10^{18}$	~35%	500
Cu <sub>0.95</sub> Pt <sub>0.05</sub> FeO <sub>2</sub> [87]	SSR	1050	na	12.5	1.8	$4.04 \times 10^{19}$	na	na
CuScO <sub>2</sub> [67]	Sputter	900	~3030	30	na	na	~40%	110
Ca:CuYO <sub>2</sub> [88]	Sputter	900	10,500	8	na	na	55%	~120
CuAl <sub>0.9</sub> Zn <sub>0.1</sub> S <sub>2</sub> [84]	CSA	500	~1570	63.5	na	$6.9 \times 10^{19}$	~85%	~100
Cu <sub>0.21</sub> Zn <sub>0.79</sub> S [77]	PLD	550	1085	54	0.7–2.0	$1-5 \times 10^{20}$	~60%	100
Cu <sub>0.3</sub> Zn <sub>0.7</sub> S [80]	PLD	RT	1090	42	0.2–1.0	$0.3-1.1 \times 10^{20}$	~50%	218
(CuS) <sub>x</sub> (ZnS) <sub>1-x</sub> [78]	CBD	80	200	1000	0.5–2	$2-8 \times 10^{21}$	~50%	50
Cu:ZnS [81]	Sputter	350	49	752	0.5–1.6	$6 \times 10^{20}$	~75%	200
CuI [89]	RTA	RT	na	110	4.1	$1.70 \times 10^{20}$	~70	50–300
(Co <sub>3</sub> O <sub>4</sub> ) <sub>0.5</sub> (Ni <sub>3</sub> O <sub>4</sub> ) <sub>0.5</sub> [90]	Sputter	400	300	333	na	na	15%	100
(Co <sub>3</sub> O <sub>4</sub> ) <sub>0.67</sub> (Ni <sub>3</sub> O <sub>4</sub> ) <sub>0.33</sub> [91]	SC	450	6000	17	na	na	25%	100
In:MOO <sub>3</sub> [92]	Furnace	395	287	233	28.2	$5.20 \times 10^{19}$	~70%	150
In:MOO <sub>3</sub> [92]	Furnace	435	313	400	11.9	$2.10 \times 10^{20}$	84%	80
Mg:CuCrO <sub>2</sub> [69]	Sputter	750	~180	220	<1	na	35%	~250
Mg <sub>0.2</sub> La <sub>0.8</sub> CuOSe [93]	RSPE	1000	~475	140	4	$2.20 \times 10^{20}$	na	~150
Sr <sub>0.25</sub> La <sub>0.75</sub> CrO <sub>3</sub> [94]	MBE	700	8300	15	0.03	$3.40 \times 10^{21}$	54%	80
Sr <sub>0.5</sub> La <sub>0.5</sub> CrO <sub>3</sub> [94]	MBE	700	3700	54	0.04	$7.50 \times 10^{21}$	42%	50
(Ni, In):ZnO [95]	USP	450	~700	59	155	$2.40 \times 10^{18}$	na	200–300
Zn <sub>1-x</sub> Al <sub>x</sub> O:Cu <sub>2</sub> O [96]	Sol-gel	400	175	144	27.8	$2.0 \times 10^{18}$	85%	~400
ZnO:Rh <sub>2</sub> O <sub>3</sub> [97]	Sputter	RT	17,500–50,000	1.9	na	na	45%	100–300
Zn-Co-O [98]	PLD	RT	~3500	21	~1	$\sim 1.3 \times 10^{20}$	na	100–200
Cu <sub>1+x</sub> Al <sub>1-x</sub> S <sub>2</sub> [99]	SPS	800	na	250	21.2	$7.3 \times 10^{19}$	na	na

Figure 6 shows the carrier-concentration-to-mobility plot of some of the best-known *p*-type TCMs. The resistivity range shows that most of the *p*-type TCMs lie in below  $10^{-2} \Omega\text{ cm}$  resistivity range and very few lie in range  $10^{-2}$  to  $10^{-3} \Omega\text{ cm}$  range. To date, most of the *p*-type TCMs have shown very poor conductivity at room temperature.



**Figure 6.** Mobility is plotted against carrier concentration for the highest performing *p*-type TCMs. Datapoints present the obtained mobility for the corresponding carrier concentration. The colour of the circles signifies the deposition temperature of the film. Dotted lines represent the line of equal resistivity.

### 3.3. Metal Nanowire-Based TCMs

A variety of metal nanowires have been developed for potential applications in transparent electrodes, such as silver (Ag), copper (Cu) and gold (Au) NWs [17]. Metal nanowires are typically synthesized by a solution-phase chemical synthesis route, which is to start with metal nuclei reduced from metal ions, grow into nanosized crystals with capping agents an-

chored on specific facets and eventually merge into an-isotropic assembly [100,101]. Factors such as aspect ratio (length/diameter) and capping agents play an important role in the conductivity and transmittance of MNWNs [15]. In general, a higher aspect ratio is favoured for higher conductivity and transmittance due to enhanced percolation conduction, fewer junctions, more void space and reduced light scattering [17]. The dimensions of metal NWs for TCMs reported in the literature typically range from a few to tens of nanometers in diameter and a few to hundreds of microns in length, with an average aspect ratio of 100–5000 [17,102]. The long insulating capping agents (such as PVP) from synthesis would dramatically hinder the conductivity of MNWNs, and thus need to be replaced with shorter capping agents such as inorganic salts (NaCl) [103,104].

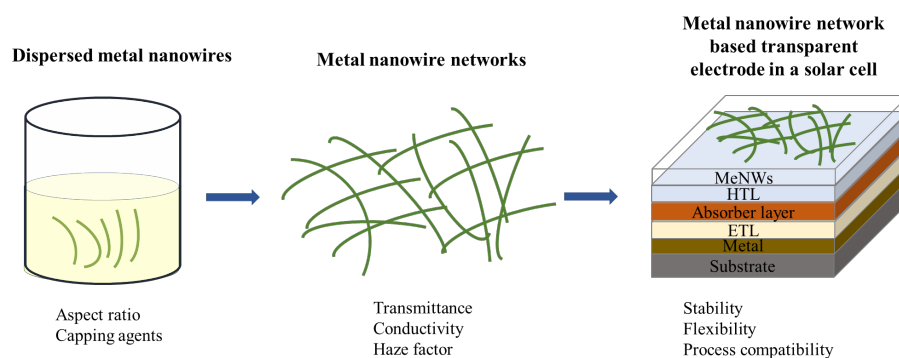
A porous interconnected network is the most common form of MNWNs applied as electrodes in optoelectronic devices. Various solution-processed methods have been exploited to assemble MNWNs from metal nanowires, including slot die coating, doctor blading, screen printing, ink-jet printing, spin coating, spray coating, dip coating, vacuum filtration, Mayer rod, electrospinning, etc. [16,17]. All these efforts are aimed to develop a low-temperature, low-cost saleable deposition method of high-performance robust MNWN electrodes.

There are a few key factors to evaluate the properties of MNWN electrodes: conductivity, transmittance and haze factor [15]. It is a trade-off between the conductivity and transmittance of an MNWN electrode [105]. Network density, the number of NWs per unit area, is a critical parameter to tune the overall electrical and optical properties of an MNWN electrode. A higher network density will typically increase its conductivity, owing to more conductive paths, however, it will reduce the transmittance as it blocks more photons from passing through [106,107]. The haze factor, defined as the amount of light scattered when light passes through a transparent sample (see equation below), is a critical parameter for light management [108].

$$\text{Haze} = \frac{\text{Forward scattered light}}{\text{Forward scattered} + \text{Forward non scattered}} \times 100\% \quad (8)$$

It is found that haze factor increases linearly with network density [109,110]. Therefore, the network density needs to be well balanced for conductivity, transmittance and haze factor. Contact resistance at the junctions is also an important factor that hinders the conductivity of the MNWNs [111]. Postprocessing treatment is typically employed to enhance its conductivity, such as annealing, mechanical pressing and nano-welding [112–115].

Figure 7 shows a schematic illustration of design principles for MNW-based synthesis for application in solar cells. The standard solar cell architecture is substrate/bottom electrode/electron transporting layer (ETL)/photoactive layer/hole transporting layer (HTL)/top electrode [116,117]. The sun passes through the transparent electrode, which can either be the top electrode or the bottom electrode. Both electrodes are required to be transparent for a semitransparent solar cell.



**Figure 7.** Schematic illustration of the design principles for metal nanowire network based transparent conductors: from the synthesis of metal nanowires, assembly of metal nanowire networks and applications in solar cells.

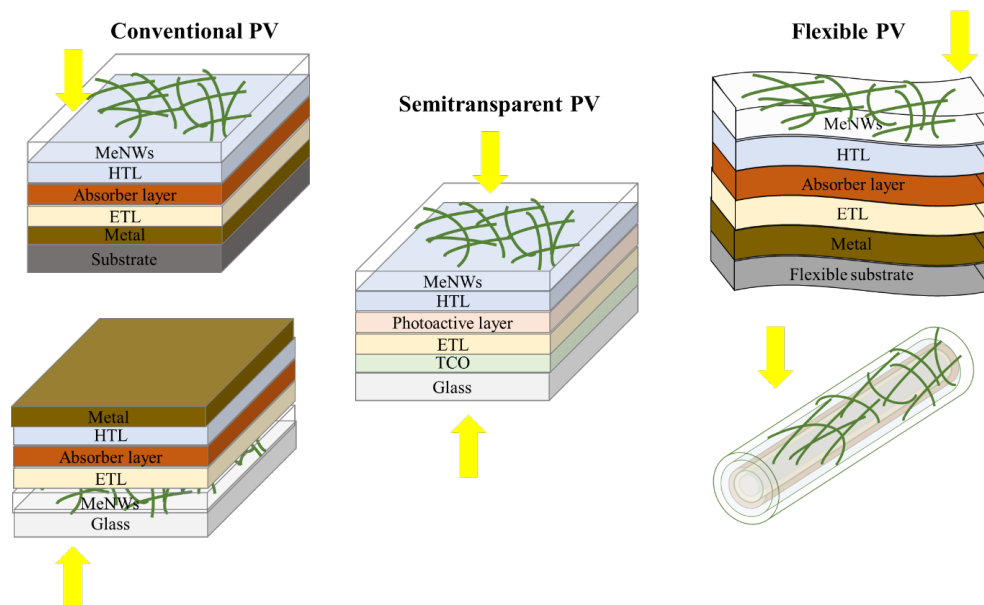
MNWNs have been considered one of the most promising transparent electrodes for organic photovoltaics (OPVs) [14]. Compared with vacuum-deposited metal oxide electrodes, MNWNs have comparable conductivity and transparency but are more compatible with the solution-processed low-cost high-throughput fabrication of OPVs [118,119]. A representative organic solar cell with metal nanowires as the transparent electrode has a device configuration of Ag NWs/PEDOT:PSS/P3HT:PCBM/Ca/Al [119]. Researchers explored different strategies to optimize the conductivity and transparency of Ag NW electrodes for a higher power conversion efficiency. With the increased  $J_{sc}$  and FF, the PCE of such devices was improved from  $\sim 1.8\%$  to  $\sim 4.2\%$  [119–123].

Semitransparent solar cells, which combine energy conversion and optical transparency in one device, have become a rising star in emerging photovoltaics [124]. As the two key features of a semitransparent solar cell, power conversion efficiency (PCE) and averaged visible transmittance (AVT) is generally used to evaluate its performance [125,126]. Owing to the same reasons as that in conventional OPVs, MNWNs are regarded as one of the most important transparent electrodes in semitransparent perovskite solar cells (PSCs). To maximize the visibility while preserving the efficiency of a cell, high conductivity and transmittance are desired for both the top and bottom electrodes. It was reported that Ag NW electrodes hardly deteriorated the transmittance of a cell in comparison with a metal thin-film transparent electrode [127]. A presentative semitransparent PSC configuration of ITO/PEDOT:PSS/perovskite/PC60BM/ZnO/Ag NWs was proposed by Guo et al., where a PCE of  $\sim 8.5\%$  and AVT of  $\sim 28.4\%$  was demonstrated [128]. Others tried to substitute the ETL and HTL with  $\text{TiO}_2$  and spiro-OMeTAD to further improve the efficiency, where a PCE of  $\sim 11.07\%$  and AVT of  $\sim 9\%$  was achieved [129]. One of the challenges for employing MNWNs in PSCs is chemical stability as the halide tends to migrate from the active layer to react with the metal nanowires and form unstable halide compounds [130,131]. Various attempts have thus been made to introduce a buffer layer in-between to reduce chemical corrosion [129,132].

As another emerging direction in photovoltaics, flexible solar cells have attracted great attention in the past two decades due to their potential in portable and wearable applications [133,134]. Other than PCE, mechanical robustness is also an important feature for flexible solar cells, such as deformability under extreme/repeated conditions [135]. Considering the Young's modulus of the photoactive layers of different photovoltaics, quantum dot solar cells and perovskite solar cells are the two most promising types to realize the feature of flexibility [136–140]. In contrast to the brittle inorganic metal oxide electrodes, the intrinsic flexibility of MNWNs make them a good candidate as a transparent electrode for flexible solar cells.

There are two representative device architectures of flexible solar cells: planar and fibre-shaped. The former is constructed on a flexible substrate, such as polyethylene terephthalate (PET) and polyethylene naphthalate (PEN) [141], while the latter is built on a thin metal wire that serves as the substrate as well as the core electrode [142]. MNWNs are typically employed as the transparent electrode in the planar flexible SCs or the shell electrode in the fibre-shaped flexible SCs.

Despite the flexibility of MNWNs, there remain several issues with using MNWNs as the transparent electrode in flexible SCs, including high surface roughness and poor adhesion [143,144]. Composite TCMs are thus widely explored to overcome these challenges. For example, Bae et al. reported that the stability of MNWNs was greatly enhanced when introducing a thin ITO coating on the networks [132]. Similarly, the PEDOT:PSS/MNWN composite electrode was also explored in flexible SCs, where increased flexibility and mechanical stability was demonstrated [145,146]. Figure 8 shows a schematic illustration of conventional PV, semitransparent PV and flexible PV.



**Figure 8.** Schematic illustration of the representative solar cell architectures employing metal nanowire networks as the transparent electrode. They are conventional solar cells with a regular stacking architecture and inverted architecture, flexible solar cells with a planar architecture and fibre-shaped architecture and semitransparent solar cells.

#### Challenges and Future Directions

Despite the great progress made in the development of solution-processable high-throughput MNW networks for photovoltaics, there remain several challenges to overcome for practical and wide applications of MNW networks.

- **Intrinsic limitations in the optical and electrical properties:** MNW transparent electrodes have a unique network geometry, where the conductive pathways come from the interconnected networks while the transparency is dependent on the voids [107]. For the electrical properties, the large contact resistance at the junctions of metal nanowires hampers its conductivity. Additionally, the voids between the junctions reduce the effective charge collection area in a solar cell. For the optical properties, the network geometry may increase the reflection loss [111]. Thermal annealing or chemical fusion will help reduce the junction resistance [112]. The “composite material strategy” will turn the non-conductive voids into effective charge pathways and minimize light loss, such as hybrid MNW networks with continuous transparent conductive thin films (ITO/MNW networks and PEDOT:PSS/ MNW networks)
- **Thermal stability:** Post-treatments are typically employed to further improve the electrical properties of MNW networks, such as thermal annealing. A fast annealing at 200 °C for 20 min can increase its conductivity by almost an order of magnitude, however, a dramatic degradation is seen at the temperature of >300 °C [147]. Optimizing the fabrication process of a solar cell may improve layer-to-layer compatibility and reduce the thermal budget of a complete device.
- **Chemical stability:** The large specific surface area of metal nanowires makes them more susceptible to oxygen and water in an ambient environment. A thin oxidation layer that gradually forms on the surface of the MNW networks will greatly increase the resistance. In addition, the reaction of metal nanowires with certain photoactive layers also poses a big issue in a device, such as perovskite solar cells. Whether MNW networks are used as the bottom electrode and in direct contact with the perovskite layer or used as the top electrode and separated from the perovskite layer, the halides will either immediately or eventually migrate to the surface of MNW networks to form unstable halide compounds [131]. Moreover, solvent compatibility needs to be considered when fabricating a solar cell. MNW networks are typically dispersed in polar solvents before

coating, such as water, ethanol and isopropyl alcohol, while perovskites are sensitive to water and ethanol [128,148,149]. To tackle the challenges discussed above, surface modification or passivation of MNWNs may help isolate metal nanowires from the reactive surroundings.

- Mechanical stability: Compared with vacuum-deposited films, MNWNs have a poorer adhesion to the substrates which inhibits their mechanical stability. Meanwhile, the junctions of the as-fabricated MNWNs are potential weak spots, especially under extreme deformations. Thermal welding or roll lamination may reinforce the adhesion of MNWNs to the substrate and metal nanowires to metal nanowires.

### 3.4. PEDOT:PSS

As the predominant candidate in conductive polymers, commercialization of PEDOT:PSS aqueous dispersion has been realized with the product name of Clevios for facile and scalable production of PEDOT:PSS films [150]. Various solution-based film depositions have been demonstrated in the fabrication of PEDOT:PSS films, such as doctor blading, screen printing, spin coating, ink-jet printing, etc. [22,151].

In photovoltaics, PEDOT:PSS has been extensively exploited either as a hole transport layer (a high work function of  $\sim 4.9$ – $5.2$  eV) [152] or a transparent bottom/top electrode in polymer and perovskite solar cells [21]. In this section, we will focus on its role as a transparent conductor. Despite its high transparency, flexibility and stability, the conductivity of PEDOT:PSS is inferior to that of ITO and Ag nanowires. There are several strategies to improve the conductivity PEDOT:PSS: solvent treatment, doping and composite materials [153].

By introducing polar solvents such as dimethyl formamide (DMF) [154], dimethyl sulfoxide (DMSO) [155,156] and ethylene glycol (EG) [157] into the aqueous dispersion of PEDOT:PSS, its conductivity can be increased by up to one order of magnitude. In addition, post-treatment of the PEDOT:PSS film with strong acid such as  $H_2SO_4$  [158,159], has also been demonstrated as an effective way to improve its conductivity to a comparable value as that of ITO. The enhanced conductivity from solvent treatment is attributed to the better-orientated PEDOT chains and increased crystallinity of the film [159].

It is shown by researchers that doping PEDOT:PSS with ammonia-based compounds (e.g., cetyltrimethylammonium bromide/CTAB [160,161]) and ammonium metatungstate hydrate/AMH [162] can increase its conductivity and adjust the valence band for a higher power conversion efficiency in perovskite solar cells. Others found that doping PEDOT:PSS with organic salts (e.g., sodium citrate [163]) and inorganic salts (e.g., sodium chloride [164] and rubidium chloride [165]) could also effectively tune the work function, conductivity and film morphology of PEDOT:PSS which favours a higher  $V_{oc}$  and FF in perovskite solar cells.

To achieve the synergetic effect of two different materials, many trials have been conducted to develop PEDOT:PSS-based composite materials for improved optical and electrical properties. Examples include PEDOT:PSS/Ag nanowires [166,167] and PEDOT:PSS/reduced graphene oxide [168]. However, extra design is typically required when a hybrid with a secondary component as PEDOT:PSS dispersion is acidic and corrosive.

In spite of the great efforts made to improve the conductivity of PEDOT:PSS, there is still room and need to further boost its conductivity for a higher  $J_{SC}$  in solar cells. The main design principle is to enhance the interactions between conductive PEDOT chains while isolating or reducing the insulating PSS segments. In addition, the acidity of PEDOT:PSS dispersion may corrode the layer underneath. Finding a co-solvent to increase the pH of PEDOT:PSS aqueous solution without compromising its performance or introducing a protection/isolation layer in-between may help resolve the issue.

### 3.5. Carbon Based Transparent Conductors

#### 3.5.1. Graphene

Of the two promising carbon-based transparent electrode materials for PSCs, graphene seems to show more promise compared to the progress made with carbon nanotubes. It

is smoother, more transparent on a broad wavelength region, and more conductive [38]. In general, graphene exhibits a plethora of promising properties including low-sheet resistance, outstanding electrical conductivity, high optical transmittance, good mechanical properties and thermal and chemical stability for PSCs [36,39,169,170]. For thin-film solar cells, graphene is typically synthesized via chemical vapour deposition (CVD) as the large area of thin-layer graphene films with fewer defects that can be produced [36,171,172]. It is then annealed at no higher than in a 10:1 Ar:H<sub>2</sub> mixture for a monolayer of graphene [173].

PSCs with high performances usually have an n-i-p architecture consisting of a scaffold metal oxide such as TiO<sub>2</sub>/perovskite material/hole transport material; however, PSCs with a reversed architecture or ones that adopt an n-i-p structure have attracted a lot of interest because of their low processing temperature, which can lead to lowering manufacturing costs and facile manufacturing processes on different substrates and for other device layers such as multi-junction solar cells [170,174]. For the electrodes, graphene is typically used to replace the top electrode or anode. There is generally a lot less effort and progress put into replacing metallic cathodes in PVs; however, it would be advantageous to do so to increase the flexibility and the overall transparency of the device specifically for FPSCs [175]. In a standard layer-by-layer design, graphene is the very top layer sealed with a layer of poly(3,4-thiophene-dioxythiophene: poly(styrenesulfonate) (PEDOT:PSS) and/or D-sorbital [170]. These are used as adhesion layers in the lamination process of layering graphene, but also between graphene and the perovskite material within the solar cell stack [36]. Below the perovskite material is the bottom electrode or cathode, usually made of fluorine-doped tin oxide (FTO), ITO, Au or Al [25]. The bottom-most layer is the substrate for rigid and flexible PSCs (i.e., glass, polyethylene naphthalate (PEN), polyethylene terephthalate (PET)).

To improve the electrical and optical properties of graphene-based transparent electrodes, multiple studies performed a combination of both stacking multiple monolayers of graphene, sealing PEDOT:PSS and D-sorbital, and doping it with molybdenum trioxide (MoO<sub>3</sub>), hydrochloric acid (HCl), nitric acid (HNO<sub>3</sub>), gold (III) and chloride (AuCl<sub>3</sub>). Multiple studies have shown a trade-off between stacking thin graphene layers to get a lower sheet resistance and the amount of transmittance. Multilayer graphene films, individually produced via CVD, are fabricated using a layer-by-layer (LBL) transfer method. A study by You et al. revealed that two-layer graphene produces the highest PCE when comparing multilayer-graphene electrodes. Another study by Li et al. that doped graphene HNO<sub>3</sub> and stacked up to four layers showed that the addition of each layer decreases the transmittance by about 2.3%. Overall, stacking more layers of graphene has resulted in less transmittance. Multilayer graphene helps accommodate an improved electrical conductivity; however, an excess of graphene nanosheets would fold together, which leads to a significant decrease of transmittance of up to 80% [36,39,176].

In addition, graphene can also be doped, specifically, *p*-doping to increase the PCE. This enhances the conductivity by increasing the carrier concentration and allows for the desired energy level to become aligned with the highest occupied molecular orbital (HOMO) level of the hole transporting layer (HTL) [35,170]. More specifically, the hydrophilicity of graphene is improved by evaporating a layer of MoO<sub>3</sub>. The presence of hydroxyl groups on MoO<sub>3</sub> allows the spreading of PEDOT:PSS to create an interfacial layer that decreases the large energy difference between graphene and HOMO of active layer poly(3-hexythiophene-2, 5-diyl):[6,6]-phenyl C61 butyric acid methyl ester (P3HT:PCBM) [170,177]. Since a monolayer of graphene suffers from low conductivity, this can be overcome by doping or mixing other elements into the graphene matrix.

### 3.5.2. Carbon Nanotubes

In addition to graphene-based electrodes, another viable carbon-based material for transparent electrodes in solar cells is the use of carbon nanotubes. Carbon nanotubes can be broken into the three following categories: single-walled (SWNTs), double-walled (DWNTs), and multi-walled (MWNTs) carbon nanotubes. CNTs are single layers of graphene bent into cylinders. SWNTs contain a single cylinder of graphene, whereas DWNTs and MWNTs

contain two or more concentric cylinders of graphene, respectively. Thus, the approaches to prepare and optimize carbon nanotubes differ drastically from that of graphene due to their distinguished geometries (e.g., 1D, 2D) and properties [38].

Like graphene, CNTs also have high optical transmittance and electrical conductivity which is what makes them attractive for transparent electrodes and hole collectors. In PSC electrodes, they are electron acceptors or hole collectors in junction with active n-type layer, specifically PEDOT:PSS layer [33]. These two properties of CNTs are critical parameters in the utility of the application of transparent electrodes. In addition, they are known for their ease of fabrication and good mechanical durability. Their stability is due to their hydrophobic nature [38]. This prevents moisture invasion in devices; however, it also poses a challenge for uniform film fabrication due to the Vander Waals interaction [38,178].

In general, most CNTs are fabricated using the floating catalyst CVD method due to the relatively low production cost, high efficiency and scalable nature [178,179]. This method is based on ferrocene vapour decomposition in a CO<sub>2</sub> atmosphere [38,180]. More specifically, Jeon et al. created a simpler solution for fabricated processible DWNTs, which were grown by a catalytic high temperature chemical vapour deposition [181]. Overall, longer, conductive and less defective and pure CNTs are desired for transparent CNT films to gain continued widespread various commercial applications [182]. The primary method to improve the conductive performance of CNTs in PSCs and FPSCs is to dope or introduce electron donors that intercalate [183]. CNTs have been doped with MoO<sub>3</sub>, HNO<sub>3</sub>, trifluoromethanesulfonic acid (TFMS), bromine (Br<sub>2</sub>), thionyl chloride (SOCl<sub>2</sub>), nafion and tetrafluorotetracyano-quinodimethane (TCNQF<sub>4</sub>) [184–188].

Moreover, doping DWNTs specifically with TFMS increased the PCE from 14.4% to 16.0% in an inverted planar PSC with specified solar architecture, as listed in Table 3. This improvement can also be noted in the fill factor (FF) and the open circuit voltage ( $V_{OC}$ ) which can be attributed to better energy alignment between DWNT and PTAA after doping. In terms of durability, TFMS doping had a lasting effect of more than 100 days, whereas the HNO<sub>3</sub> doping effect disappeared after 10 days [189]. According to Xu et al., HNO<sub>3</sub> is an effective dopant; however, there is also an energy level mismatch between HNO<sub>3</sub>-doped CNTs and the perovskite layer. This could also be due to the damage that HNO<sub>3</sub> does to the structure of SWNTs and possibly DWNTs [38].

Other studies have added a bilayer of MoO<sub>3</sub> to the PSC device with variations in thickness. Unlike doping CNTs with TFMS or HNO<sub>3</sub>, multiple studies show that PSCs with MoO<sub>3</sub>-doped CNTs electrodes have much lower PCEs [180]. This is due to the energy level mismatch between PEDOT:PSS and MoO<sub>3</sub>. Ultrathin layers of MoO<sub>3</sub> with 2 and 6 nm thicknesses were deposited via vacuum thermal evaporator which was followed by anaerobic annealing at 115 °C for 30 min. Thicker layers led to a larger energy misalignment with a lower transmission. This can be noted again in Table 3, where the thickness of the MoO<sub>3</sub> bilayer increased (2 nm to 6 nm) and the PCE decreased from 12.8% to 11.0%. Despite its PCE performance relative to other dopants, MoO<sub>3</sub> is one of the most established and chemically stable dopants for both CNTs and graphene.

The current main challenges with CNTs are balancing out the wettability and conductivity. Oftentimes, many of the chemical treatments to increase the hydrophilicity decrease the conductivity because it introduces many oxygen-containing groups [190]. Thus, there is a need for improving current wettability techniques for CNTs. Additionally, it is also difficult to optimize between transmission and conductivity. As the CNT-film thickness increases, the conductivity increases since percolation begins at around 50 nm thick. However, the thickness of a CNT-electrode layer that yields a high transmission range of 90–95% is 5–10 nm. This is because of the low percolation threshold of CNTs, which is due to the one-dimensional nature of the tubes and bundles.



**Table 3.** Literature summary of carbon-material based transparent electrodes used in solar cells.

Device Architecture	FF (%)	PCE (%)	T (%)
Glass/AgNWs-G1/SnO <sub>2</sub> /Perovskite/Carbon [39]	43.64	10.2	88
Glass/AgNWs-G2/SnO <sub>2</sub> /Perovskite/Carbon [39]	66.46	15.31	86
Glass/G-2(HNO <sub>3</sub> -doped)/PEDOT:PSS/n-Si/LiF/Al [36]	40	5.35	97
Glass/G-3(HNO <sub>3</sub> -doped)/PEDOT:PSS/n-Si/LiF/Al [36]	39	5.48	95
Glass/G-4(HNO <sub>3</sub> -doped)/PEDOT:PSS/n-Si/LiF/Al [36]	47	5.76	93
Glass/G-3(undoped)/PEDOT:PSS/n-Si/LiF/Al [36]	36	3.92	90
Glass/G-3(undoped)-(1 month)/PEDOT:PSS/n-Si/LiF/Al [36]	38	4.84	na
Glass/FTO/Compact-TiO <sub>2</sub> /MAPbI <sub>3-x</sub> Cl <sub>x</sub> /Spiro-OMeTAD/PEDOT:PSS/G-1 [169,176]	59.07	8.74	93
Glass/FTO/Compact-TiO <sub>2</sub> /MAPbI <sub>3-x</sub> Cl <sub>x</sub> /Spiro-OMeTAD/PEDOT:PSS/G-2 [169,176]	71.72	12.03	88
Glass/FTO/Compact-TiO <sub>2</sub> /MAPbI <sub>3-x</sub> Cl <sub>x</sub> /Spiro-OMeTAD/PEDOT:PSS/G-3 [169,176]	67.58	10.94	83
Glass/FTO/Compact-TiO <sub>2</sub> /MAPbI <sub>3-x</sub> Cl <sub>x</sub> /Spiro-OMeTAD/PEDOT:PSS/G-4 [176]	68.37	10.18	na
Glass/Graphene/MoO <sub>3</sub> -1 nm/PEDOT:PSS/Perovskite/C60/BCP/LiF/Al [170]	45	6.7	89
Glass/Graphene/MoO <sub>3</sub> -2 nm/PEDOT:PSS/Perovskite/C60/BCP/LiF/Al [170]	72	16.1	na
Glass/Graphene/MoO <sub>3</sub> -4 nm/PEDOT:PSS/Perovskite/C60/BCP/LiF/Al [170]	70	15.9	na
G-3/MoO <sub>3</sub> +PEDOT:PSS/P3HT:PCBM/LiF/Al (HCl interlayer and HNO <sub>3</sub> doping) [191]	45	1.9	na
Graphene/PEDOT:PSS/P3HT:PCBM/LiF/Al [191]	51	2.5	na
Glass/SnO <sub>2</sub> :F/TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /Spiro-OMeTAD/graphene [192]	56.6	8.4	64
PET/APTES/AuCl <sub>3</sub> -G/PEDOT:PSS/FAPbI <sub>3-x</sub> Br <sub>x</sub> /PCBM/Al [193]	76.2	17.4	na
PET/APTES/AuCl <sub>3</sub> -G/PEDOT:PSS/MAPbI <sub>3</sub> /PCBM/Al [193]	75.7	15.6	na
PET/AuCl <sub>3</sub> -G/PEDOT:PSS/MaPbI <sub>3</sub> [193]	75.8	15.5	na
DWNT/PTAA/MA <sub>0.6</sub> FA <sub>0.4</sub> PbI <sub>2.9</sub> Br <sub>0.1</sub> /BCP/Cu [189]	69.1	14.4	na
HNO <sub>3</sub> -doped DWNT/PTAA/MA <sub>0.6</sub> FA <sub>0.4</sub> PbI <sub>2.9</sub> Br <sub>0.1</sub> /BCP/Cu [189]	71.7	15.6	na
TFMS-doped DWNT/PTAA/MA <sub>0.6</sub> FA <sub>0.4</sub> PbI <sub>2.9</sub> Br <sub>0.1</sub> /BCP/Cu [189]	75	16	na
Glass/SWNT/IPA-PEDOT:PSS/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PC61BM/Al [194]	50	4.01	na
Glass/SWNT/surfactant-PEDOT:PSS/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PC61BM/Al [194]	38	1.85	na
Glass/70 v/v% HNO <sub>3</sub> -SWNT/PEDOT:PSS/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PC61BM/Al [194]	55	5.96	na
Glass/50 v/v% HNO <sub>3</sub> -SWNT/PEDOT:PSS/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PC61BM/Al [194]	52	5.82	na
Glass/35 v/v% HNO <sub>3</sub> -SWNT/PEDOT:PSS/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PC61BM/Al [194]	54	5.7	na
Glass/15 v/v% HNO <sub>3</sub> -SWNT/PEDOT:PSS/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PC61BM/Al [194]	39	3.80	na
Glass/SWNT/MoO <sub>3</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PC61BM/Al [194]	28	0.05	na
Glass/SWNT/MoO <sub>x</sub> /PEDOT:PSS/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /PC61BM/Al [194]	42	2.09	na
FTO/TiO <sub>2</sub> /(FAPbI <sub>3</sub> ) <sub>1-x</sub> (MAPbBr <sub>3</sub> ) <sub>x</sub> /SWNT:Spiro-OMeTAD/Au [195]	61	15.5	na
FTO/TiO <sub>2</sub> /(FAPbI <sub>3</sub> ) <sub>1-x</sub> (MAPbBr <sub>3</sub> ) <sub>x</sub> /SWNT/Au [195]	46	11	na
Ti-125(μm)Foil/TiO <sub>2</sub> NTs + CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /CNT+spiro-OMeTAD [178]	62	3.90	na
Ti-25(μm)Foil/TiO <sub>2</sub> NTs + CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /CNT+spiro-OMeTAD [178]	63	4.83	na
Glass/SWNT/PEDOT:PSS/P3HT:PCBM/Al [33]	29	1.5	na
Glass/SWNT/P3HT:PCBM/Al [33]	29.8	0.5	na
Glass/SWNT/MoO <sub>3</sub> -2 nm/PEDOT:PSS/MAPbI <sub>3</sub> /C60/BCP/LiF/Al [180]	76	12.8	na
Glass/SWNT/MoO <sub>3</sub> -6 nm/PEDOT:PSS/MAPbI <sub>3</sub> /C60/BCP/LiF/Al [180]	71	11	na
Glass/SWNT-HNO <sub>3</sub> /PEDOT:PSS/MAPbI <sub>3</sub> /C60/BCP/LiF/Al [180]	78	15.3	na
PEN/SWNT/MoO <sub>3</sub> -2 nm/PEDOT:PSS/MAPbI <sub>3</sub> /C60/BCP/LiF/Al [180]	65	11	na

Graphene (G), One Layer of Graphene (G-1); Two Layers of Graphene (G-2); Three Layers of Graphene (G-3); Four of Layers of Graphene (G-4); Silver Nanowires (AgNWs); Poly(3-hexylthiophene):phenyl-C60-butyric acid methyl ester (P3HT:PCBM); Poly(3,4 thylene-dioxythiophene: poly(styrenesulfonate) (PEDOT:PSS); Nitric Acid (HNO<sub>3</sub>); Gold (III) Chloride (AuCl<sub>3</sub>); 2,2',7,7',tetrakis(N, N-di-p-methoxyphenylamine)-9,9-spirobifluorene (Spiro-OMeTAD); Polyethylene Terephthalate (PET); Bathocuproine (BCP); Molybdenum Trioxide (MoO<sub>3</sub>); Single-Walled Carbon Nanotubes (SWNT); Double-Walled Carbon Nanotubes (DWNT); 3-aminopropyltriethoxysilane (APTES); Lithium Fluoride (LiF); TiO<sub>2</sub> NTs (Titanium Dioxide); Isopropanol (IPA); Trifluoromethanesulfonic Acid (TFMS); Poly(triaryl amine) (PTA).

#### 4. Conclusions

Most of the dominating photovoltaic technology presently uses crystalline Si and various perovskite compounds as absorbers with TCMs in the back and front sides along with metal contacts. These TCMs act as a membrane that blocks one carrier and selects the other, thereby defining the current flow direction and making the solar cell work. Various *n*- and *p*-type TCMs have been extensively used in solar cells. However, low conductivity, mobility and high contact resistance limit the performance of solar cell devices. To overcome this issue, a lot of research has been going on in developing high-performing TCMs.

Despite the great progress made in the development of solution-processable high throughput MNWNs for photovoltaics, there remain several challenges pertaining to optoelectronic properties, thermal stability, chemical stability and mechanical stability. Several pathways such as thermal annealing, chemical fusion and composite material

strategy have been proposed to improve optoelectronic properties of MNWNs. Further, the thermal, chemical and mechanical stability can be improved via optimizing the fabrication process, surface modification, surface passivation and thermal welding/roll lamination. In the case of polymer-based TCMs such as PEDOT:PSS, conductivity still remains very low. However, finding a suitable co-solvent to increase the pH of PEDOT:PSS aqueous solution may enhance overall optoelectronic performance.

PCE of PSCs resulted in increased efficiencies with the integration of transparent graphene and CNTs electrodes via optimizing between conductivity and transparency. Additionally, the mechanical transferring of monolayer graphene still remains a major challenge. Further, the current main challenges with CNTs are balancing out the wettability and conductivity. Oftentimes, many of the chemical treatments to increase hydrophilicity decrease the conductivity because it introduces many oxygen-containing groups. Thus, there is a need for improving current wettability techniques for CNTs.

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## Abbreviations

The following abbreviations are used in this manuscript:

TCM	Transparent conducting material
TC	Transparent conductor
LED	Light-emitting diode
CBM	Conduction band minima
VBM	Valence band maxima
ALD	Atomic layer deposition
CBD	Chemical bath deposition
PLD	Pulsed laser deposition
ITO	Tin-doped indium oxide
AZO	Al-doped zinc oxide
IZO	Indium zinc oxide
FTO	Fluorine-doped tin oxide
UTMF	Ultra-thin metal film
PEC	Power conversion efficiency
FPSC	Flexible perovskite solar cell
SWCNT	Single-walled carbon nanotube
MNWN	Metal nanowire network

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