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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.

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**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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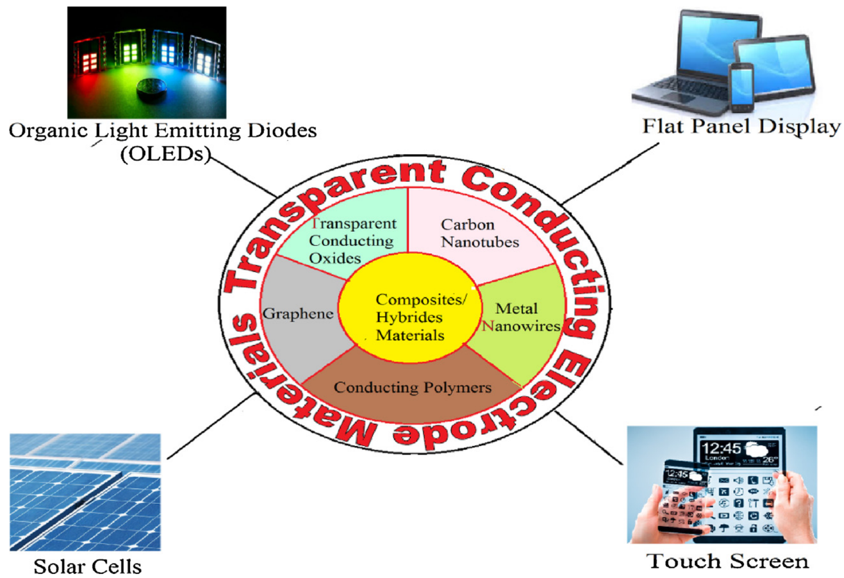


**Fig. 1.** Transparent Conducting Electrode Materials and their applications.

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,

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