Recent Developments in Flexible Transparent Electrode

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Abstract: With the rapid development of flexible electronic devices (especially flexible LCD/OLED), flexible transparent electrodes (FTEs) with high light transmittance, high electrical conductivity, and excellent stretchability have attracted extensive attention from researchers and businesses. FTEs serve as an important part of display devices (touch screen and display), energy storage devices (solar cells and super capacitors), and wearable medical devices (electronic skin). In this paper, we review the recent progress in the field of FTEs, with special emphasis on metal materials, carbon-based materials, conductive polymers (CPs), and composite materials, which are good alternatives to the traditional commercial transparent electrode (i.e., indium tin oxide, ITO). With respect to production methods, this article provides a detailed discussion on the performance differences and practical applications of different materials. Furthermore, major challenges and future developments of FTEs are also discussed.

Keywords: flexible transparent electrodes; AgNWs; CNTs; graphene; flexible electronic devices

1. Introduction

With the development of science and technology, people have increasing requirements with respect to flexible electronic devices. Flexible transparent electrodes (FTEs) are essential components for flexible electronic equipment, such as LCD, OLED, and TFT. Hence, FTEs have attracted more and more attention, which has led to extensive research in recent years [1–4]. At present, the ITO electrode, acting as the representative transparent electrode, has been commercialized and extensively used due to its high transparency (≈85%) and low sheet resistance (100–500 Ω·sq−1). Unfortunately, the production of ITO is hindered by the severe shortage of indium, high production cost, and the brittleness of the material itself [5]. At the same time, people have increasing requirements for the flexibility, stretchability, and foldability of flexible electronic devices. However, the preparation of ITO electrodes requires subsequent processes such as high-temperature annealing, which further limits its application and development. As it is difficult for ITO electrodes to be manufactured on flexible substrates which are not resistant to high temperature, finding alternative materials to ITO and meeting commercial requirements has become a research hotspot. These FTEs have excellent conductivity, optical transmittance, and mechanical properties, and they can be mass-produced. The FTEs reviewed in this article are made of different materials, including metal materials (metal nanowire, ultra-thin metal films, and metal mesh) [6,7].
carbon-based materials (carbon nanotubes (CNTs) and graphene) [3,8], conducting polymers (CPs) [9], and composite materials [10,11]. These various electrode materials all have their own advantages, which make them good options for new-generation FTEs. Previous studies have demonstrated that these materials not only have excellent flexibility, but also have lower electrical resistivity and higher light transmittance than ITO electrode. This review comprehensively summarizes the preparation methods and performance of these electrode materials. High-performance FTEs have a wide range of applications in optoelectronic devices, such as touch screens [12], solar cells [13], super capacitors [14], flexible electroluminescent devices, and wearable devices [15]. Moreover, problems in the industry and future development directions are also outlined in this article, hoping to provide a reference for the design and manufacture of FTEs.

2. Electrode Materials

2.1. Metal

2.1.1. Metal Nanowires

Because of their excellent photoelectric properties, chemical properties, mechanical properties, and flexibility, metal nanowires have become the most likely substitute for ITO [16–19]. Noble metal nanowires such as gold and silver (Figure 1a,b) were rapidly developed and entered the market first [20]. The preparation methods include the liquid polyol method [21,22], template method (soft template method [23,24] and hard template method [25]), self-assembly method [26,27], ultrasonic reduction method [28], wet chemical method [29,30], and ultraviolet (UV) laser pattern method [31]. Among them, liquid polyols are currently the easiest method to achieve high-efficiency production owing to their high effect, simple production, and low cost, and a simple method for Ag nanowire (AgNW) synthesis is shown in Figure 1e. The template method can control the morphology and properties of the final product, which is easy to synthesize [16]. As for UV laser patterning, the light transmittance, haze, and conductivity of AgNWs can be changed by adjusting the laser power. At the same time, nanowires can be purified using the precipitation method [32,33] and heat treatment [34], which can improve the density and conductivity of the nanowires. Atkinson et al. found that the high transparency of AgNWs (94%) in the infrared region (wavelength is 2500 nm) is much higher than that of traditional ITO electrodes (35%). In addition, thinner AgNWs result in a larger diameter and higher transparency. This characteristic of AgNWs has a significant impact on the development of solar cells and window coatings [35]. Liu et al. successfully prepared AgNWs in a water environment by adjusting the appropriate pH value, and the aqueous phase environment enabled the samples to have better biological applications [36]. Previous studies improved the performance of AgNWs by coating them with transparent conducting oxide (TCO) layers (such as ITO and ZnO), but this process is complex with a high cost, and it results in brittleness of the FTEs. Yu et al. proposed a hot lamination process to solve the problems of poor electrical conductivity and adhesion of AgNWs, which greatly reduced the square resistance (from 100 to 12.5 Ω·sq⁻¹) under the condition of constant optical transmittance (94.5%) [37]. AgNWs electrodes are easily oxidized at high temperature, which increases their resistance. Photoresist encapsulation proved to be a superior method to improve the reliability of AgNWs electrodes under harsh conditions [38]. In addition, Pantoja et al. studied the performance (including reflectance, transmittance, emissivity, and haze) of AgNWs in the infrared spectrum and prepared low-emissivity AgNWs films, reducing the thermal emissivity of the substrates [39]. Copper is second only to silver in metal conductivity, with low cost and high yield. Therefore, Cu nanowires (CuNWs) were also rapidly developed in recent years; Figure 1f shows a schematic diagram of a healable nanocomposite conductor based on CuNWs, with a sheet resistance of 22.3 Ω·sq⁻¹ and 66.5% transmittance at a wavelength of 550 nm [40]. Huang et al. synthesized CuNWs (Figure 1c,d) with high purity, large aspect ratio, good dispersion, and controllable morphology using the hydrothermal method [41]. Moreover, Duong et al. innovated a simple, low-cost, and environmentally friendly method for preparing CuNW-based flexible
electrodes using polyethylene-based inks. It was also verified that copper salt, capping agent, reducing agent, reaction temperature, and other conditions play an important role in controlling the morphology of CuNWs [42]. In order to further improve the conductivity of metal nanowires, the influence of various factors on the nanowires has been studied through mathematical modeling methods and verified through experiments [43–45]. It was found that orientation restriction can improve the unidirectional conductivity of metal nanowires, while randomness is beneficial to the conductivity of the metal network.

Figure 1. (a) SEM and (b) TEM images of AgNWs [20]; (c,d) SEM images of CuNWs at different magnifications [41]; (e) schematic diagram of AgNW synthesis process [20]; (f) schematic diagram of a healable nanocomposite conductor based on CuNWs and DA-PU [40].

2.1.2. Ultra-Thin Metal Films

An ultra-thin metal film has the advantages of high stability, simple manufacturing process, and low cost [46,47]. Compared with metal nanowires, it has lower roughness and does not reduce the display resolution because of no scattering [48], which is conducive to display applications. However, because it must satisfy both high conductivity and optical transmittance, the film should be thin enough (<12 nm) [49]. Previous studies successfully prepared Au [11], Ag [50], Cu [51], Al [49], Cr [52], Ni [46], and other metal thin films. The preparation methods were mainly room-temperature sputtering deposition [48] and high-temperature thermal deposition [53]. The growth of a metal film on a substrate is generally divided into three stages (nucleation, coalescence, and thickness growth), and the performance of the film is affected by the various sputtering conditions. Results showed that silver clusters will form metal islands during the deposition process, making the film nonconductive [54]. This defect can be suppressed by multilayer deposition passivation, the process of which is shown in Figure 2a [55]. In addition, the introduction of a wetting layer can limit the migration and agglomeration of metal atoms under heating conditions to smoothen the film surface, as well as form defects, to increase the adhesion strength with the substrate. However, due to its high optical reflectivity rather than high absorptivity, the metal film has limited transmittance. It is well known that the optical transmittance of a metal film is extremely low. To deal with these problems, adding a dielectric antireflection layer to both sides of the metal layer is significant, resulting in an electrode structure known as the dielectric/metal/dielectric (DMD) structure [56,57]. However, in the DMD structure, Ag will migrate from the undisturbed region to the disturbed region with the passage of time, resulting in discontinuity of the Ag layer, thus, insulation of the electrode. One way
to slow the loss of electrode life is to keep the substrate clean and pay attention to storage conditions [58]. Figure 2b shows a schematic structure of a ZnO/Ag/ZnO transparent heater, where the addition of 5% Ni could inhibit the diffusion of Ag atoms and effectively improve the thermal stability of Ag films [50]. Xie et al. investigated a low-resistance (7 Ω·sq⁻¹) and high-transparency (73%) MoO₃/Ag/WO₃ top transparent electrode and successfully applied it to solar cells. Tuo et al. suggested replacing Ag in DMD structures with Cu, whereas adding a small amount of Ni (5%) to Cu could suppress the diffusion of Cu atoms. They also proved that WO₃₋ₓ/Cu:Ni/WO₃₋ₓ structures are fairly stable three-dimensional structures [59]. However, obviously, the highest conductivity and the best transmittance cannot be obtained at the same time. Surprisingly, it was confirmed that the fatigue strength of ultra-thin metal films is greater than that of thick films on account of the increase in yield stress and the formation of nano twins [60]. Furthermore, Cr and Ni films can maintain high optical transparency in the full wavelength range [61], while metal films such as Ag maintain high light transmittance only at specific wavelengths. In the ultra-thin gold–silver–gold multilayer film thermally deposited on flexible substrates by Bauch et al., the transmission spectrum was extended [53]. In addition, the stability of Ag thin films is poor. Studies have shown that metal doping is a good way to significantly improve the stability of Ag. In order to further improve the performance, Shi et al. deposited a micron silver grid on an Ni film, which reduced the resistivity from 390 Ω·sq⁻¹ to 20 Ω·sq⁻¹ while keeping the transparency (78%) largely unchanged [46]. Afterward, Dong et al. fabricated high-performance Au film FTEs and used them to make a foldable perovskite light-emitting diode (PELED) and a biaxially stretchable alternating current electroluminescence (ACEL) display (Figure 2c,d) [11]. Ji et al. proposed a flexible DMD-based electrode, with a relative optical transmittance higher than 100%. A thin metal film was sandwiched between two antireflective media to achieve high optical transparency and low fog, and it showed good compatibility with other substrates [48].

![Figure 2](image-url)  
**Figure 2.** (a) Schematic sketch of metal film deposition (conventional deposition and new multilayer deposition strategy) [55]; (b) schematic structure of the ZnO/Ag/ZnO transparent heater fabricated on top of a transparent PI substrate [50]; (c) schematic architecture of a stretchable ACEL device before and after stretching; (d) photographs and OM images of patterned ACUF/anisotropic conductive ultrathin film (ACUF)/Au electrodes and their interconnection [11].

2.1.3. Metal Mesh

A metal mesh has good flexibility and mechanical properties; moreover, because of its three-dimensional (3D) shape, there is no strict limit on the thickness. Different combinations of transmittance and conductivity can be customized by adjusting line widths and spacing. The current preparation processes mainly include the heat treatment...
method [62], template method [63], lithography [64], imprinting [65], and laser pulse etching [66]. In order for them to be commercially available, it is necessary to find a high-throughput and low-cost preparation process. Moon et al. produced FTEs by flash sintering with a reverse-offset Ag screen pattern, and they experimentally proved that multi-pulse flash sintering could effectively reduce the sample temperature, increase the stability of the films, eliminate the damage or microstructure defects of the substrate, and achieve better performance (R_s = 27 Ω·sq⁻¹ and transmittance = 84.7%) [67]. Kim et al. prepared silver grid films on flexible substrates through self-assembly and coupling processes (Figure 3), and they found that chemical coupling could effectively improve the conductivity (8.2 Ω·sq⁻¹) and light transmittance (>88%) with a figure of merit (FoM) value of 350 [68]. Li et al. proposed combining electroless plating with a recrystallized ice crystal template to prepare a flexible transparent copper mesh with controllable line width and mesh size (Figure 4) [69]. This method can also be applied to other metal meshes. In addition, Muzzillo et al. explored the principle of metal mesh fabrication using crack film lithography (CFL) and obtained controllable performance. This method has low cost, high speed, and high quality, and it can be widely applied to monolithically integrated thin-film photovoltaic (PV) modules [70]. However, a metal mesh cracks easily during use; thus, it was proven that it can be repaired by electrodeposition to further improve service life [71].

**Figure 3.** (a) Schematic illustration of the preparation of transparent and conductive Ag mesh films; (b,d) SEM images of Ag mesh films fabricated with Ag nanoparticle ink after a sintering process; (c,e) SEM images of Ag mesh films fabricated with Ag nanoparticle ink after a coupling process [68].

**Figure 4.** (a) Schematic illustration of the preparation of copper meshes on a flexible substrate; (b) SEM image of copper nanoparticles; (c) evolution of sheet resistance and transparency at 550 nm wavelength with mesh size; (d) evolution of sheet resistance and transparency at 550 nm wavelength with line width; (e) sheet resistance variation (R/R_0) versus the number of cycles of repeated bending to a radius of 3 mm [69].
2.2. Carbon-Based Materials

2.2.1. Carbon Nanotubes

Carbon Nanotubes (CNTs) have attracted the attention of researchers because of their excellent mechanical properties, photoelectric properties, thermal stability, and large specific surface area, which has been studied for a long period; good reviews on CNTs have previously been published [72–75]. These excellent properties are related to the sp² hybridization of carbon atoms [76]. CNTs can be categorized into multiwalled carbon nanotubes (MWCNTs) and single-walled carbon nanotubes (SWCNTs) [77], as well as armchair type, serrated type, and chiral type [68]. Figure 5a shows a TEM image of CNTs [73]. Currently, graphite is used as a raw material, whereby electrophoretic coating [78], ultrasonic spraying [79], dipping [80], brush coating [81], rod coating [82], and transfer printing [83] methods have been used to produce CNTs, and CNTs containing carbon impurities can be purified by annealing [84], chemical oxidation [85], and polymer introduction [86]. However, the abovementioned synthesis methods have the disadvantages of complicated postprocesses and large film roughness. Multilayer transfer printing (MTP) technology is a good alternative to the above synthesis methods [87]. Oytun et al. coated CNTs with positive and negative charges on the glass substrates by multilayer deposition; then, chemical and thermal post-treatment was carried out, before the electrode was transferred to the polymer film (Figure 5b). For 12-layer MWCNTs and SWCNTs, the maximum FoM is 2.52 × 10⁻⁶ Ω⁻¹ at 68% light transmittance and 1.14 × 10⁻³ Ω⁻¹ at 81% light transmittance [88]. In addition, atoms, molecules, polymers, metal nanoparticles, and cellulose can be used to modify the surface and ends of CNTs [72,89]; alternatively, they can be inserted into the hollow space of the nanotube [89]. Modification can further improve the conductivity and optical transmittance of CNTs. Sergey et al. thoroughly studied the mechanism of nitrogen-modified CNTs to increase electrical conductivity, and they determined that nitrogen has the highest binding energy in the graphite-like state [90]. Moreover, Awadallah et al. discovered a method of modifying CNTs with carbon nanoparticles, and they prepared hybrid carbon nano products with different properties and morphologies from the original CNTs [72]. It was also proven that the interaction between CNTs can increase the tensile strength of the electrodes [91]. Furthermore, the size of CNTs also has great influence on their properties. NWCNTs with a thinner diameter have higher transparency, whereas longer NWCNTs have higher conductivity [92]. In practical applications, thin-film transistors (TFT) were successfully prepared using CNTs [93].

![Figure 5. (a) TEM image of CNTs [73]; (b) representation of the layer-by-layer schematic process and multilayer transfer process [88].](image)

2.2.2. Graphene

Among carbon-based materials, CNTs are one-dimensional (1D), while graphene is a two-dimensional (2D) sheet. It not only has excellent electrical, optical, thermal, and mechanical properties, but also maintains the characteristics of a large surface area and low cost. Graphene and its derivatives (graphene oxide (GO) and reduced graphene oxide (rGO)) have been commonly used in flexible transparent electrodes [94–97]. However, like CNTs, they have a high initial irreversible capacity, which can be reduced by pre-lithification, allowing them to be used in flexible batteries [91]. Graphene can be prepared by chemical vapor deposition (CVD), chemical reduction [98], and the solution liquid
droplet method [99]; furthermore, they can be transferred to flexible substrates by screen printing, chemical etching, spraying, spin-coating, dip-coating, drop-casting, vacuum filtration transfer, Langmuir–Blodgett (LB), and roll-to-roll processes. Figure 6 shows the complete preparation process of graphene electrodes [96]. The industry requires that the transferred graphene be pure and uniform, with a smooth surface. CVD is a good method to grow large areas of high-quality graphene films; however, it requires expensive equipment and a complex etching transfer process, which may produce toxic byproducts. Meanwhile, graphene produced via CVD easily forms wrinkles due to the coupling effect with the substrate. To solve this problem, Yuan et al. invented a novel method called proton-assisted CVD to produce a super-flat graphene film [100]. Compared with CVD, the solution method involves less chemistry and a relatively low cost of production; thus, it is a simpler and cheaper way to produce and transfer graphene on a large scale than CVD. Studies have shown that with the increase of graphene layers, the electrical conductivity increased, while the optical transmittance decreased, similarly to metal thin films [101]. Therefore, the thickness of graphene should be adjusted according to the actual application for different devices. For battery applications, three layers of graphene can achieve a balance between electrical conductivity, optical transmittance, and surface roughness [102]. In addition, a graphene mesh (3D form) electrode can further improve transmittance due to its mesh structure. The main fabrication processes of a graphene mesh include lithography combined with oxygen ions and vacuum filtration; however, neither of these processes lead to both low resistance and high light transmittance. Thus, Wu et al. combined molecular carding and transfer techniques, allowing the successful fabrication of a transparent reduction oxidation graphene electrode with low sheet resistance (0.8 Ω·sq⁻¹) and high optical transmittance (>95%), with a stable performance after 10K bending [94]. Song et al. proposed a fast and energy-saving method (flame-induced foaming in a confined space) to fabricate high-performance 2D porous graphene films, and they proved that cellular graphene films had better conductivity and flexibility than disordered and lamellar structures [103]. However, carbon-based materials still have the disadvantage of higher resistance than metals, which limits their application to some extent.

![Figure 6](image_url)

**Figure 6.** The preparation processes of a Polydimethylsiloxane (PDMS) dielectric layer with micro-pyramid structures and pressure sensor-based graphene electrodes [96].

### 2.3. Conductive Polymers

Conducting polymers (CPs) have been developed rapidly since their discovery at the end of the 20th century, inaugurating a new era of organic optoelectronics. CPs are simple to synthesize and easy to process, and they can achieve the diversity of materials through molecular design and clipping to implement different functions such as light/color adjustability and electromagnetic protection. Furthermore, they have good optical and electronic properties, which are promising for the application of optoelectronics [104,105].
However, as organic matter, they have low strength and poor stability. CPs conduct electricity through a system of conjugated bonds on the polymer chain [106], described as molecular conductivity, and the range for the conductivity of polymers is $10^{-16} - 10^8$ S/cm. In the undoped state, CPs have insulating properties, but the band split can be reduced to improve electrical conductivity through doping ($n$ doping and $p$ doping) [107]. These doping techniques include electrochemical doping, acid–base doping, photo-doping, and charge injection at the metal/polymer interface [108,109]. Common CPs include polyaniline (PANI), poly(3,4-ethylenedioxy thiophene) (PEDOT), poly(m-phenylene diamine) (PMPD), polyphenylene diamine (PPD), polynaphthylamine (PNA), poly(p-phenylene sulfide) (PPS), poly(p-phenylene vinylene) (PPV), polyacetylene (PAC), polyfluorene (PF), polyethylene terephthalate (PET), and polynaphthalene (PN). CPs can be prepared by chemical polymerization [110], electrochemical polymerization [111], light-induced polymerization [106], and vapor phase polymerization [112]. Their photoelectric properties can be adjusted and improved to achieve device performance control by chemical modification, such as via the introduction of functional groups. By controlling the nucleation and growth process of the polymerization, nanostructured CPs with controllable size and shape can be obtained. Previous reviews have comprehensively summarized the regulation of the energy gap and functionalization (via doping and copolymerization) of CPs [113].

Yuk et al. investigated the high-performance 3D printing of polymers based on PEDOT:PSS, using a rapid, simple, and significantly streamlined production method [114]. Due to the unique electrical properties of CPs, they are generally not used as electrodes alone. High-performance FTEs can be prepared by combining CPs with other conductive materials, such as metals or carbon-based materials (graphene and CNTs). Lim et al. found that silver electrodes prepared with polymer-added silver paste have high conductivity [115]. Rahman et al. obtained free-forming gel polymer electrolyte (GPE) samples with variable shapes and sizes by 3D printing (Figure 7), and the addition of $N,N$-dimethylacrylamide (DMAAm) into the polyvinylidene fluoride (PVDF) network significantly improved the tensile elongation (up to 200%) and mechanical properties of GPE [116]. Zhu et al. used a combination of printing, evaporation, and wet etching techniques to produce transparent micromesh electrodes with high conductivity ($19.7 \text{ } \Omega\cdot\text{sq}^{-1}$) and high optical transmittance (80%), and the electrical conductivity remained stable over 3K bending [117]. In addition, Mahmut et al. found that unidirectional conductive transparent films can be prepared by embedding nanomaterials into polymer membranes (Figure 8) [9].

![Figure 7. Schematic diagram of PVDF/DMAAm gel synthesis process using 3D gel printer [116].](image)

### 2.4. Composite Materials

From the above discussion, it can be seen that different electrode materials have different advantages and disadvantages, whereas defects limit the application of FTEs to some extent. For example, the high surface roughness of AgNWs affects electrode stability, metal thin films usually have low light transmittance and poor tensile properties, the junction resistance between CNTs inhibits conductivity, graphene sheets have high resistance and poor hydrophobicity, and CPs mostly have poor thermal stability and are...
difficult to use as electrodes alone [118]. Figure 9 presents a radar map of different material properties. Figure 9 clearly shows the performance and production level differences of some popular materials. This has some implications for the selection of electrode materials and the commercialization of FTEs.

**Figure 8.** Schematic representation of nanocomposite thin film production: (a) glass substrate; (b) spin-coated glass with conductive material; (c) polymeric material-coated conductive glass; (d) removal of conductive polymeric film from glass; (e) self-standing conductive polymeric layer [9].

In recent years, many researchers have combined the advantages of various materials to prepare composite materials to further improve the performance of FTEs. Previous studies have shown that high-performance CP/CNT nanocomposites can be prepared by direct mixing, chemical oxidation, and electrochemical polymerization [108]. Lee et al. found that a graphene/In$_2$O$_3$ bilayer (called GI-bilayer) as a transparent conductive electrode can significantly improve the uniformity of sheet resistance. The In$_2$O$_3$ substrate reduces the sheet resistance of graphene from 150 $\Omega\cdot$sq$^{-1}$ to 7.5 $\Omega\cdot$sq$^{-1}$ [119]. By hybridiz-
ing graphene with CNTs, Kumar et al. improved the electrical conductivity of the thin film, and they decorated it with gold nanoparticles to achieve a resistance of 100 $\Omega \cdot \text{sq}^{-1}$ and a high optical transmittance of 96% [120]. Yang et al. fabricated CNT/AgNW multilayer FTEs via a spray-coating process, which showed good photoelectric performance ($Rs = 50 \Omega \cdot \text{sq}^{-1}$ and transmittance = 94%) at 500–550 nm wavelength [120]. In order to further understand the interface behavior of composites, Cui et al. simulated the contact behavior of CNTs and AgNWs at the atomic scale using molecular dynamics simulations, which showed that the CNTs and AgNWs move toward each other and then form a complete interfacial arrangement [121]. Ma et al. prepared graphene/ionic gel @PEDOT:PSS using a solution treatment, which was applied as a transparent electrode to obtain a low thin layer resistance of 30 $\Omega \cdot \text{sq}^{-1}$ and a high transmittance of 88% [122]. Kim et al. reported a way to improve the electrical conductivity of graphene by doping it with metal selective atomic layer deposition (ALD) (Figure 10). The electrical resistance of graphene was significantly reduced from 471 $\Omega \cdot \text{sq}^{-1}$ to 86.8 $\Omega \cdot \text{sq}^{-1}$ when the light transmittance was greater than 90% [123]. Furthermore, Kang et al. proposed that graphene/metal hybrid materials have better electrical properties, light scattering, and low surface roughness. The sheet resistance of a 7 nm Cu/graphene hybrid film is about 75 $\Omega \cdot \text{sq}^{-1}$, and its light transmittance is 92% [10]. Additionally, the metal CNT mixed electrode prepared by Shan et al. using a layer-by-layer (LBL) method realized a good chip resistance of 9.44 $\Omega \cdot \text{sq}^{-1}$ and an elongation at break of 104% [124].

![Figure 10. Concept of metal atomic layer deposition (ALD) on graphene. (a) Doping graphene by charge transfer between graphene and metal. (b) Selective deposition of Pt by ALD on graphene [123].](image)

3. Manufacturing Process

3.1. Coating Methods

FTEs have a wide range of applications in the fields of display, energy, and biology domains. Generally speaking, the preparation methods of thin films can be divided into dry methods (DMs) and wet methods (WMs), which correspond to small molecules and high-polymer materials, respectively. DMs are mainly vacuum evaporation methods, including chemical vapor deposition (CVD) and physical vapor deposition (PVD). These methods are seldom affected by impurities due to the vacuum environment, but they are costly and take a long time to synthesize. The films performance can be improved by adjusting the gas pressure, gas flow rate, power supply, and other parameters during the commercial process. WMs include coating (e.g., spin-coating, spraying, and cast), printing (e.g., inkjet [125], screen printing [126], and nanoimprint), and dipping (e.g., dipping, LB, and self-assembly) processes. WMs have the advantages of being cheap and easy to manufacture in a large area, but they often introduce impurities and are difficult to pattern. The advantages and disadvantages of each method are summarized in Table 1. In recent years, some new preparation methods have emerged, such as pulsed laser deposition (PLD) [127], LBL [128], digital fabrication, and drop-on-demand techniques [129]. Stanojev et al. prepared controllable MWCNTs films using the LBL method, and the relationship between the number of deposition layers and the electrode morphology, thickness, transparency, and resistivity of the films was studied [130]. As a noncontact direct molding and rapid manufacturing printing technology, drop-on-demand printing has considerable potential in future thin
In practical production, different film preparation methods can be combined to obtain the best performance.

### Table 1. The advantages and disadvantages of traditional methods.

| Method       | Advantage                                         | Disadvantage                                                   |
|--------------|---------------------------------------------------|---------------------------------------------------------------|
| CVD          | Film is very pure and dense                       | Reaction at high temperature; difficulty in mass production; toxic byproducts |
| PVD          | Good film uniformity; high process automaticity   | Difficulty in mass production                                  |
| Spin-coating | Film thickness uniformity                         | Loss of large quantities of solution                           |
| Spraying     | No loss of solution                               | Difficulties in process optimization of solvent mixing and solution adjustment |
| Cast         | High crystallinity                                | Poor homogeneity of film                                       |
| Inkjet       | Computer control; material loss is small; fast production of multi-layer structures | Ink requirements high conditions; droplet coalescence           |
| Screen printing | Simple manufacturing process                     | Thick film                                                    |
| Nanoimprint  | The scale of tens of nanometers patterning can be obtained at normal temperature and pressure | The process is not mature; the equipment is expensive          |
| Dipping      | Large area film formation                         | Thick film                                                    |
| LB           | Controlled production of monolayer and layer by layer accumulation | The corresponding materials are limited                        |
| Self-assembly | Monomolecular membrane with high orientation       | The corresponding materials are limited                        |

The prepared film usually undergoes subsequent optimization processes, including annealing and patterning [131]. Electrode patterning can be achieved through photolithography, etching, parylene lift-off, and laser-induced ablation, whereby the latter is a low-cost process that does not generate toxic chemicals. However, the rough surface leads to its optical properties being easily affected. Kim et al. found that the absorption of visible light by nanoparticles is the main cause of discoloration, and they presented a method for completely removing nanoparticles using an excimer laser source [131]. As the electrical performance of electronic devices such as TFT tends to deteriorate after repeated bending, a micropore array was proposed to release stress and prevent fatal breakdown of the conductive layer. A triangular array can withstand bending better than a square array, and a micropore array can improve the stability and durability of devices [132].

### 3.2. Performance Characterization

The photoelectric properties of thin films have received much attention by researchers, and they can be examined and evaluated by different methods. Square resistance and contact resistance can be measured using four probes and the transfer line method, respectively. Carrier mobility measurement methods usually include time of flight (TOF), extraction in a linearly increasing voltage (ELIV), and impedance spectroscopy (IS), in which the thickness of the material measured by TOF should be more than 1 µm. The surface morphology and the microstructure of the film can be observed by AFM, TEM, and SEM, from which surface roughness, the changes in grain size, and the reaction between layers can be easily obtained. In addition, the film transmittance and haze can be detected using an UV visible spectrophotometer and fog meter. The film stress can be measured using methods such as digital image correlation (DIC) or devices such as a laser beam probe detector (LBPD), fiber optical sensor (FOS), and multi optical sensor (MOS), whereas adhesion and flexibility are generally measured separately by tape testing and a flexometer. Chen et al. invented an in situ experimental system based on the colorimetric method and MOS technique which can simultaneously observe the morphology and measure the stress [133]. Carrier lifetime can generally be measured by microwave photoconductivity decay (µ-PCD), surface photoelectric voltage (SPV), and quasi-steady-state photoconduction (QSSPC). For the
Raman spectra, a blue shift or red shift of the spectral line is an expression of the energy band change, which can be inferred from the molecular microstructure. By analyzing the peak and half peak width of the XRD pattern, the phase analysis of the film, the grain size, and the crystal orientation can be easily obtained. The XPS spectrum is a good tool for analyzing surface element content, element valence states, and binding patterns. The above methods can be used to analyze the macroscopic properties and characterize the microcosmic properties of the films, which can be combined to optimize the properties of the films and thoroughly characterize the mechanism.

4. Electrode Applications

Transparent electrodes have important applications in the fields of display, energy, and biology domains. However, although the current leading market ITO transparent electrode materials have good conductivity and high optical transmittance, they face the challenge of depletion of indium resources. In addition, the rapid development of flexible electronic devices has resulted in new requirements for electrodes, which should not only feature a high photoelectric performance, but also present flexible elasticity and even foldability. It is well known that ITO has poor mechanical flexibility and is prone to cracks under strain or bending. This requires finding substitution materials for ITO as soon as possible. As mentioned above, metal materials (metal nanowires, metal ultra-thin films, metal grids), carbon-based materials (CNTs and graphene), and CPs have proven their potential as good alternative materials to ITO. According to their different advantages, they can be used in flexible touch screens, bendable displays, super capacitors, solar cells, wearable devices, biomedical equipment, and other electronic devices. Figure 11 depicts the different applications of FTEs. Furthermore, different devices have different requirements for conductivity, optical transmittance, and flexibility, as discussed below.

![Figure 11: The applications of FTEs.](image-url)
4.1. Touch Screens and Display Panels

In touch screen applications, the $R_s$ of FTEs should be between $100 \, \Omega \cdot \text{sq}^{-1}$ and $1000 \, \Omega \cdot \text{sq}^{-1}$, and the transmittance should be between 85% and 92%, and the haze should be low. In order to meet market demands, touch screens are widely used in various electronic products with LCD and E-paper screens such as smart phones, tablet computers, and public displays. Meng et al. integrated a friction electric field effect touch sensor with SWCNTs, which greatly improved the touch sensitivity [134]. Moreover, Yang et al. fabricated a flexible capacitive touch sensor using the double-sided nanoimprint lithography metal transfer method. The diamond aluminum mesh achieved electrode alignment and overlap, obtaining a transmittance of 94% and anisotropic surface resistivity, with a maximum capacitance variation of the touch sensor unit of up to 41.8%. It has great application potential in flexible touch screens [135]. Kim et al. proposed a new method to fabricate capacitive flexible touch screens by depositing two transparent electrodes in a PET film, which is a low-cost approach that can meet the requirements of flexible electronic devices [136]. Moreover, Choi et al. designed a new type of device structure of transparent pressure sensor, which exhibits high linearity over a wide pressure range [137]. Compared with traditional capacitive sensors, piezoelectric sensors have higher precision and sensitivity. Sergio et al. created an eco-friendly ink, and its use in screen printing technology resulted in dense piezoelectric films with a piezoelectric coefficient $|d_{33}| = 19 \, \text{PC/N}$. Furthermore, it can be integrated into a variety of substrates including flexible substrates [126].

Displays are often combined with touch screens to enable interaction between people and devices. As an important component of a display panel, OLED has become a research hotspot in recent years due to its excellent performance such as fast response time, low operating voltage, high contrast, strong flexibility, and large lateral size [138]. In an OLED, the $R_s$ range is 10–50 $\Omega \cdot \text{sq}^{-1}$ and the transmittance is greater than 88%. Metal materials can be used for OLED because of their low resistance and good flexibility, but metal nanowires and mesh structures have high fog due to scattering, which can be mitigated by designing appropriate structures [139]. Sarjidan et al. used spin-coated AgNWs as intermediate electrodes to manufacture vertical organic light emitting transistor (VOLET) devices with excellent performance, including Schottky barrier (SB) VOLET and static induction transistor (SIT) VOLET devices, and the manufacturing method was cost-effective and efficient [140]. In addition, quantum dot luminescence has also become a research focus in recent years. Jiang et al. fabricated a completely transparent inverse quantum dot light-emitting diode (QLED) by incorporating an AgNWs anode. The QLED had a light transmittance of 75.66% at the main peak wavelength (530 nm) [141]. Carbon-based materials are also widely used in display devices due to their high light transmittance. Gu et al. successfully prepared a highly conductive sandwich structure (a CNT/PEDOT:PSS/CNT transparent conductive film) for OLED electrodes. The device had a high brightness of 1598 cd/cm$^2$ at 14 V and a maximum current efficiency of 1.5 cd/A at 13 V [142].

4.2. Flexible Electroluminescent Devices

Flexible electroluminescent devices (FELDs) play an important role in display and communication fields. Their luminous mechanism involves emitting light under the action of electric excitation, which can be divided into DC and AC devices according to the excitation type. AC electroluminescent (ACEL) devices have the advantages of adjustable light color (from UV to infrared), proper alignment of material energy levels, and good uniformity; thus, they have been widely studied in recent years. A typical ACEL device is composed of two conductive layers and the middle EL layer. The working process is to apply an AC voltage on the conductive layer, whereby the electric field excites the electrons in the EL material, thus exciting the emission center in the EL layer and then the electrons in the excited state transition to emit light [143]. Furthermore, the luminescence spectrum can be regulated by changing the material and AC frequency. In order to make the color adjustable, Zuo et al. reported a color-tunable EL device with simple double-
stacked emissive layers (ZnS phosphors/dielectric polymer composite), and its color could be adjusted (from orange to white and to blue) by changing the electric field in a single electronic device [144]. The development of FELDs is hindered by the low dielectric constant of materials. To deal with this problem, a dielectric nanocomposite was prepared by means of filling the ceramic nanoparticles into polar elastomers, which maintained good mechanical stress and could be processed in solution [145]. To address the brightness of FELDs, Shim et al. proposed a light-transmitting layer and a retroactive radio pole device structure, improving the brightness of FELDs by 442% compared to commercial EL devices [146]. Furthermore, a screen-printed EL transducer based on the reversed ACEL structure was proposed for immunoassays, highlighting the possibility of FELD application in the biomedical field [147].

4.3. Wearable Electronic Products

In wearable electronic products, CPs can replace traditional glass substrates due to their outstanding flexibility and mechanical durability, as well as their good transparency and light weight. However, they have low thermal/dimensional stability and poor mechanical strength, which can be improved by using fiberglass-reinforced polymers [148]. Moreover, CPs can be used in conjunction with metals and other materials to improve performance. To realize wearable pressure sensors, Li et al. embedded a Cu network transparent electrode into a PDMS substrate and found that oxygen plasma treatment could inhibit template cracking and increase the photoelectric performance 20-fold [149]. Zhao et al. combined high-conductivity poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) with stretchable SWCNTs to develop a transparent capacitive sensor array. The good optical transparency and stretchable performance of the electronic skin could realize the visual perception of the wearable electronic system [150]. In addition, electronic textiles have shown excellent performance in wearable devices. Afroj et al. produced repeatable machine-washable, low-resistance (R ≈ 11.9 Ω·sq⁻¹), ultra-flexible, and durable graphene-based electronic textiles using the pad–dry–cure method [151]. The wearable smart textiles were knitted with high-performance nanostructured hybrid piezo fiber, and a knee sleeve prototype was successfully prepared to monitor real-time precise healthcare in 2019 [152]. Wu et al. prepared a wearable ultra-lightweight polymer solar textile, whose anodes and transparent substrates were transparent electronic fabrics with polyester/AgNWs/graphene structure. The solar textiles had a high-power conversion efficiency (2.27%) and were compatible with clothing; thus, they have broad application prospects [153].

4.4. Flexible Batteries and Supercapacitors

Due to the booming development of flexible touch screens, displays, wearable devices, and implantable medical devices, there is an urgent need to provide corresponding flexible energy storage devices for power, including flexible batteries (metal-ion batteries and solar cells) and supercapacitors. In solar cells, the R of FTEs should be around 10 Ω·sq⁻¹, and the transmittance should be above 90%. Graphene is a 2D honeycomb lattice with a high surface area, high carrier mobility, good ductility, and light weight [154]. Many studies have shown that surface graphene and its derivatives (GO and rGO) are suitable for the fabrication of flexible batteries, which can be used as anode materials or as flexible substrates to support electrochemically active materials to improve electrical conductivity. Dai et al. reviewed the research progress of flexible graphene-based battery electrodes, including lithium–sulfur batteries, metal-ion batteries (ions of Li, Na, Zn, Al, etc.), and metal–air batteries (Li–air and Zn–air batteries) [8]. Tiwari et al. discussed various applications of graphene films prepared using different stripping, deposition, and production methods in solar cells, and they showed that the photoelectric conversion efficiency (PCE) and device life can be improved due to the good thermal stability of graphene electrodes [101]. Moreover, Yao et al. prepared a highly dispersed graphene composite transparent electrode that meets the electrical conductivity and optical transmittance requirements of perovskite solar cells.
(PSCs) for flexible distributed generation. In addition, the graphene electrode had excellent mechanical flexibility and diffusion transmittance, which greatly improved the efficiency of flexible PSCs, with an optimal efficiency of 15.71% [155].

As an energy storage device, supercapacitors have been developed rapidly for decades. Supercapacitors have the advantages of fast speed, large power capacity, long service life, simple charging circuit, no memory effect, and low maintenance cost, which can be used in a wide temperature range [156]. The specific capacitance range of graphene-based supercapacitors is 99–550 F g\(^{-1}\), which is suitable for the electrode material of supercapacitors. However, graphene flakes tend to aggregate and stack easily, which reduces the active area, electrical conductivity, and capacitance [154]. Qin et al. designed an anthraquinone-functionalized graphene framework. The microporous structure prevented the re-stacking of graphene sheets in the manufacturing process, which could significantly improve the performance of supercapacitors [157]. In addition, graphene-based composites have been studied to improve their electrical conductivity and capacitance, and a detailed review can be found in previous article [158]. Metal-based materials have inherent advantages such as high electrical conductivity, adjustable photoelectric properties, and good thermal stability [159]. The 3D morphology of the ultra-thin metal network can increase the specific surface area of the active material, increase the number of active sites, and shorten the electron/ion transmission path, thereby improving the electrochemical performance of the supercapacitor [160]. Liu et al. developed a method of forming Ag/porous carbon and Ag/\(\text{Ni}_x\text{Fe}_x\text{O}_z\) microsystems by intruding rGO into grid-like patterns on polyethylene terephthalate foil to produce transparent thin-film ultracapacitors and hybrid ultracapacitors. The supercapacitor could provide 226.8 \(\mu\text{F}\cdot\text{cm}^{-2}\) high surface capacitance at a current density of 3 \(\mu\text{A}\cdot\text{cm}^{-2}\) with a transparency of 70.6% [161].

4.5. Electromagnetic Interference Devices

As mentioned above, high-performance FTEs are currently widely used in display, light-emitting devices, wearable devices, and other fields, but the electromagnetic waves (EMWs) emitted by these optoelectronic devices can cause product failure and harm human health. Therefore, while maintaining high photoelectric and mechanical properties, FTEs should realize an electromagnetic shielding function. It is well known that the key to high-performance electromagnetic interference (EMI) shielding films is to combine high electrical conductivity with effective optical transparency. Many studies have been carried out in this respect, and the MXene membrane was found to be an ideal candidate material. Zhou et al. proposed designing a flexible transparent conductive film based on the hybrid structure of 2D MXene nanosheet and 1D AgNWs, which presented low resistance (18.3 \(\Omega\cdot\text{sq}^{-1}\)), high optical transmission (52.3%), and high shielding efficiency of EMI (32 dB). The high shielding efficiency was due to the fact that the nano-packing not only acted as a bridge connecting the conductive path of MXene, but also formed an additional charge-polarized interface. In addition, the multifunctional membrane presented a good photothermal response, and it had good cycling stability under harsh environments such as strong light irradiation [162]. Chen et al. found that \(\text{T}_{13}\text{C}_2\text{T}_{1}\) MXene welded AgNW films had a higher electromagnetic shielding performance (34 dB) and better environmental and mechanical stability than pure AgNW films of the same density (21 dB). Furthermore, the shielding efficiency of the macroscale layered EMI could reach 49.2 dB when the transmittance was 83% [163].

5. Conclusions and Prospects

In summary, this article reviewed the latest research progress of FTEs. Metal materials, carbon-based materials, and CPs are all good alternatives to the transparent ITO electrodes, which are widely used in commercial applications, and these materials can exhibit excellent flexibility. In practical applications, different products have different requirements in terms of the conductivity, flexibility, and optical transmittance of FTEs. This requires the flexible selection of materials and manufacturing methods to achieve a balance between
performance and economic cost. However, most FTEs are still in the laboratory research stage and have not yet achieved large-scale commercial applications. There are still many difficulties and challenges in applying FTEs to various products (such as wearable devices and solar cells). At the same time, there is no uniform standard for the testing and evaluation of flexibility across the industry. Although research on FTEs is booming, it is obviously not yet mature. In the future, we still need to study additional properties of FTEs (including stability, degradability, and economic cost). Furthermore, the whole production line from material synthesis to device manufacturing needs to be further improved.

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