

Review

Synergistic Design of Flexible Substrates and Transparent Electrodes for Application in Organic Photovoltaics: A Review

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Abstract

Flexible organic solar cells (FOSCs) are a promising green energy technology due to their mechanical flexibility, light weight, low cost, and compatibility with large-area solution processing. Although the power conversion efficiency (PCE) of rigid organic solar cells has exceeded 20%, a significant performance gap remains for flexible devices, primarily constrained by the limitations of two key components: the flexible substrate and the transparent electrode. This review systematically summarizes recent research progress on flexible substrates, including ultrathin glass, polymer substrates, stretchable substrates, and bio-based substrates, and flexible transparent electrodes, including ITO, conductive polymers, carbon-based nanomaterials, ultrathin metal films, metal grids, and metal nanowire networks. Building on this, the review explores strategies for the synergistic design of substrates and electrodes, analyzing critical pathways for their co-optimization across four dimensions: interface engineering, mechanical compatibility, optical coupling, and process integration. Examining representative case studies from the literature, optimal substrate–electrode pairings for different application scenarios are summarized. Finally, the review outlines a future perspective on the evolution from compatibility toward functional integration, offering a systematic framework for the development of next-generation flexible photovoltaic devices that are efficient, stable, and adaptable to diverse application requirements.

Keywords: flexible organic solar cells; flexible substrates; flexible transparent electrodes; synergistic design; functional integration



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1. Introduction

Organic solar cells (OSCs) are considered a promising technology for green energy generation, primarily due to their beneficial characteristics such as mechanical flexibility, light weight, low cost, and compatibility with solution-based, large-area printing techniques [1–4]. This technological development has increased the need for flexible power generation in applications ranging from smart buildings to portable and wearable devices [5–7]. In this context, flexible organic solar cells (FOSCs) emerge as an ideal candidate, combining the general advantages of OSCs with specific benefits like ease of processing and satisfactory power conversion efficiencies (PCEs) [8,9]. Significant progress has been made

in OSC research. The development of novel photoactive materials has driven the certified PCE of rigid devices beyond 20% [10,11]. However, despite continuous improvements in materials, device architecture, and operational understanding, the highest performance benchmarks are still held by cells fabricated on rigid substrates. In contrast, the PCE record for FOSCs currently stands around 19.40%, indicating a performance gap that needs to be addressed [12]. This gap highlights the unique challenges associated with transferring high efficiency to flexible platforms.

As illustrated in Figure 1, a standard FOSC device comprises several layers: a flexible substrate, a flexible transparent electrode (FTE), interfacial layers, the photoactive layer, and a back electrode. The fundamental difference from rigid counterparts lies in the flexible substrate and the FTE. The substrate primarily provides mechanical support, serves as an optical window, and offers thermal stability and solvent resistance, whereas the transparent electrode is responsible for achieving high conductivity, high optical transmittance, low surface roughness, and good mechanical durability. Therefore, these two components are critical as they directly determine not only the mechanical integrity of the device but also its ultimate photovoltaic performance, flexibility, and operational reliability [13].

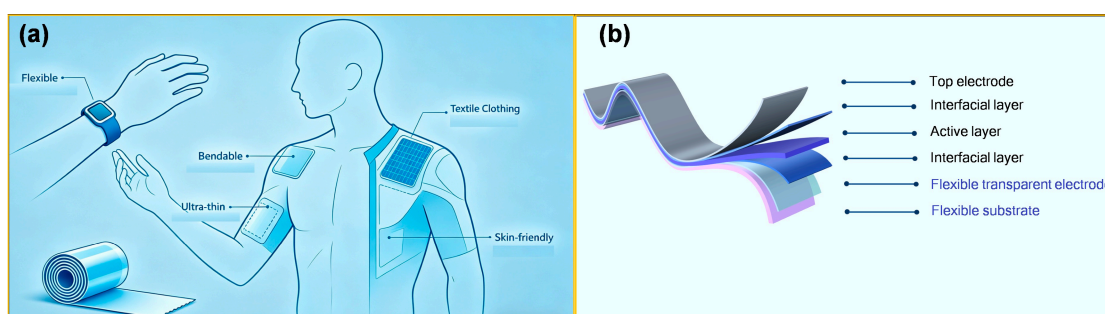


Figure 1. (a) Schematic of wearable flexible organic solar cells with key performance features and application scenarios. (b) Typical device architecture of flexible organic solar cells.

To achieve flexible devices with both high PCE and robust mechanical properties, stringent requirements must be imposed on these core components. The flexible substrate must typically possess the following: (1) high optical transmittance to maximize light absorption by the active layer; (2) low surface roughness to enable the formation of uniform, pinhole-free thin films and prevent shunting; (3) excellent mechanical flexibility to withstand bending or folding; (4) good solvent resistance to endure processing steps; (5) adequate thermal stability to tolerate annealing temperatures; (6) high surface energy for good wettability by processing solutions; (7) a low coefficient of thermal expansion (CTE) to minimize stress and crack formation during fabrication and operation; and (8) effective barrier properties against oxygen and moisture ingress, which are key factors in device degradation [14,15].

Similarly, the flexible transparent electrode must fulfill a demanding set of criteria: it should exhibit (1) high electrical conductivity (low sheet resistance) for efficient charge collection with minimal resistive losses; (2) high optical transparency, particularly across the absorption spectrum of the active layer, to reduce parasitic absorption; (3) low surface roughness and good morphology to ensure optimal interfacial contact with adjacent layers without damaging the ultrathin active film; (4) exceptional mechanical robustness, maintaining electrical and structural integrity under repeated deformation; (5) compatibility with both the underlying substrate (e.g., matched CTE) and the subsequent device processing steps; and (6) scalable, cost-effective, and environmentally benign fabrication processes [16,17].

To ensure the overall device performance, the two components must be matched and co-optimized across multiple dimensions. Optically, the transmission spectra of the substrate and electrode should be complementary, particularly in the active layer absorption range (e.g., 300–900 nm), to synergistically enhance light harvesting. Mechanically, they should possess similar elastic modulus and elongation at break to mitigate interfacial stress concentration and improve the device's resistance to bending and stretching. Thermally, their coefficients of thermal expansion should be closely matched to avoid interlayer delamination or cracking caused by thermal stress during processing or under varying environmental temperatures. Moreover, the electrode material must exhibit good interfacial compatibility and strong adhesion to the substrate surface to prevent peeling or the formation of interfacial defects, thereby jointly ensuring the efficient and stable operation of the device.

This review systematically summarizes recent research progress on flexible substrates and flexible transparent electrodes for FOSCs. A particular focus is placed on exploring strategies for their co-design and performance optimization. The aim is to provide a coherent perspective and guidance for the development of next-generation flexible photovoltaic devices that are efficient, stable, and tailored for diverse applications.

2. Flexible Substrates

2.1. Ultrathin Glass Substrate

Flexible ultrathin glass (UTG) substrates have emerged as a promising candidate for high-efficiency solar cells, meeting stringent demands with a combination of outstanding properties. They offer over 90% transmittance in the visible range, providing ample photon flux to the photoactive layer, along with excellent chemical stability and resistance to most solvents. With a low CTE (3–3.5 ppm·K^{−1}) and thermal tolerance exceeding 600 °C, they can also be engineered for high transparency in specific spectral bands [18]. Modern fabrication techniques enable thicknesses below 100 µm, granting these substrates notable flexibility.

A representative example is Corning® Willow® Glass, a flexible glass that enhances bendability through its high transparency and sub-100 µm profile. Its high surface energy allows compatibility with various advanced techniques including roll-to-roll (R2R) processing, photolithography, printing, coating, and vacuum deposition [19]. In 2019, Yu et al. demonstrated efficient ITO-free FOSCs on 100 µm Willow Glass using modified PEDOT:PSS as the flexible transparent electrode [20]. Leveraging the glass's solvent resistance, they achieved a PCE of 8.0%. When scaled to an active area of 1.6 cm², the device retained a PCE of 5.2% with good stability. The high-temperature tolerance of glass also enables unique processing routes. For instance, Han et al. prepared a ZnO/AgNW hybrid electrode at 180 °C [21], this hybrid layer functions as both a transparent electrode and an electron transport layer. Furthermore, light-scattering from nanopores in the combustion-derived ZnO enhances transmittance between 450 and 550 nm. With this ZnO/AgNW electrode, PTB7-Th-based polymer solar cells reached a PCE of 8.48%. Table 1 summarizes the efficiencies of flexible organic photovoltaic devices based on different substrates.

Despite these advantages, flexible UTG substrates face limitations, primarily due to their inherent bending stiffness. Even at reduced thickness, glass remains considerably more rigid than polymer films of comparable dimension. This has motivated exploration of alternative flexible substrates.

2.2. Bendable Polymer Substrate

2.2.1. Polyethylene Terephthalate (PET) and Polyethylene Naphthalate (PEN)

In the field of FOSCs, polyethylene terephthalate (PET) and polyethylene naphthalate (PEN) (Figure 2a) have been widely adopted as common petroleum-based polymer

flexible substrates, owing to their mature processing technology, good manufacturability, and excellent mechanical flexibility. Both are commercially available semi-crystalline thermoplastic polyester resins with a long history of industrial use, similar to glass and metals, and are extensively employed as flexible substrate materials in various electronic and photovoltaic devices. In terms of optical and mechanical properties, PET and PEN exhibit high transparency in the visible range with transmittance exceeding 85%, along with considerable ductility featuring elongation at break up to approximately 70%. Their low Young modulus further ensures reliable mechanical flexibility under bending or folding conditions. Additionally, these hydrophobic polymers demonstrate strong resistance to many organic solvents, excellent dielectric properties, and relatively low moisture and oxygen permeability. For instance, the water vapor transmission rate for 100 μm thick PET is about $9 \text{ g}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$, while that for PEN is around $2 \text{ g}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$, which is notably lower than that of most common polymer substrates [22,23].

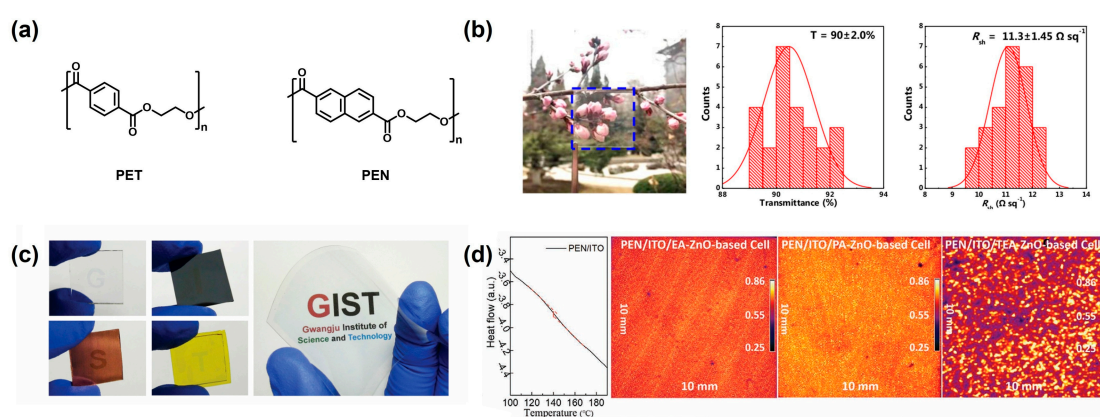


Figure 2. (a) The chemical structures of PET and PEN. (b) FlexAgNE on large-area (12.25 cm^2) flexible PET substrates and the transmittance and sheet resistance values of each pixel in a 5×5 array. The histograms of the transmittance counts for 25 pixels in a 5×5 array and the histograms of the R_{sh} counts for 25 pixels in a 5×5 array (Reproduced with permission [24], Copyright 2019, Springer Nature). (c) Photographic images of H_2SO_4 -treated PEDOT:PSS films transferred onto various substrates, including a glass slide, a silicon wafer, Cu foil, Kapton tape, and large-area PET foil (Reproduced with permission [25], Copyright 2015, John Wiley and Sons). (d) The T_g of PEN/ITO is calculated from the result of temperature-dependent heat flow. The respective lowest TA of ETLs gives rise to the folds of varying degrees, which are demonstrated in the EQE mappings (Reproduced with permission [26], Copyright 2021, John Wiley and Sons).

In 2018, Ge et al. reported an all-solution-processed FOSC utilizing PET/PEDOT:PSS electrodes, which achieved a record-high PCE of 10.12% at that time [27]. The performance improvement originates from a room-temperature gentle acid treatment process, which enables the formation of a high-performance PEDOT:PSS interface on PET substrates with a matched work function ($\approx 4.91 \text{ eV}$), along with interface engineering that optimizes contact and enhances hole transport. Furthermore, the flexible device demonstrates excellent mechanical robustness, retaining approximately 94% of its initial efficiency after 1000 bending cycles. In 2019, Chen et al. developed high-performance flexible transparent electrodes by fabricating well-dispersed AgNW networks with poly(sodium 4-styrenesulfonate) (PSSNa) on PET substrates (Figure 2b) [24]. The resulting electrodes exhibited excellent electro-optical properties (sheet resistance $\approx 10 \Omega \text{ sq}^{-1}$, transmittance $\sim 92\%$) and were directly integrated into flexible organic photovoltaics. Notably, single-junction and tandem devices achieved remarkable PCEs of 13.1% and 16.5%, respectively, demonstrating performance comparable to devices on rigid ITO electrodes.

However, PET suffers from a relatively low glass transition temperature (T_g) of 78–120 °C, which limits its application in device fabrication processes that often require high-temperature treatments [28]. In contrast, PEN possesses a similar chemical structure to PET, except that the benzene ring in the molecular chain is replaced by a more rigid naphthalene ring. This structural modification endows PEN with superior properties, including higher physical and mechanical strength, enhanced gas barrier performance, improved chemical stability, as well as greater resistance to heat, UV light, and radiation. Consequently, PEN substrates can serve as a viable alternative to PET under more demanding processing conditions [29].

For instance, in 2015, Lee et al. demonstrated a transfer-printing method to fabricate FTEs under harsh conditions (e.g., concentrated H_2SO_4) and subsequently transfer them onto PEN substrates for use in FOSCs (Figure 2c) [25]. In another study, Hou et al. developed a sol-gel ZnO layer for FOSCs by substituting ethanol with propylamine, which reduced the required annealing temperature from 200 °C to 130 °C while still forming a high-quality ZnO film (Figure 2d). Although the temperature was lowered, PET substrates remained unsuitable for even 130 °C processing. Therefore, they employed PEN/ITO as the FTE and PM6:BTP-eC9 as the active layer to construct FOSCs, achieving a notable PCE of 16.71% with an active area of 1.00 cm². Nevertheless, the relatively high manufacturing cost of PEN compared to PET remains a limiting factor for its widespread adoption [26].

2.2.2. Polyimide (PI)

Polyimide (PI) is a polymer containing imide functional groups and is widely regarded as an ideal substrate for flexible electronics. Its structure can be tailored via functional group design, offering a combination of desirable properties. Most notably, PI exhibits superior thermal stability and strong solvent resistance against chemicals commonly used in semiconductor processing, outperforming many other polymer materials [15]. Furthermore, PI possesses excellent mechanical strength, maintaining both support capability and chemical stability even at ultrathin thicknesses, making it an optimal choice for ultra-thin FOSCs, which require the total device thickness to be below 10 µm.

In 2022, Someya and Zhou et al. reported an ultrathin OPV device with a total thickness of only ≈ 3 µm, fabricated on a 1.4 µm transparent polyimide (tPI) substrate in an inverted architecture (ITO/ETL/active layer/MoOx/Ag) [30]. The electron transport layer (ETL) was engineered using either conventional sol-gel ZnO or a Zn^{2+} -chelated ethoxylated polyethyleneimine (PEI-Zn) interlayer, combined with the high-performance PM6:Y6 bulk-heterojunction as the photoactive system. The resulting devices achieved a notable PCE of 15.8% and an outstanding power-per-weight ratio of 33.8 W g⁻¹. Moreover, by employing a chelation strategy to suppress Zn diffusion from the ETL, the group demonstrated excellent air stability (89.6% PCE retention after 1574 h in air) and remarkable mechanical deformability, retaining 87.3% of the initial performance after 5000 compression-stretching cycles (Figure 3a). In 2022, Li et al. developed an ultraflexible transparent composite electrode (FTCE) by semi-embedding a silver nanowire (AgNW) network into a colorless polyimide (CPI) substrate (Figure 3b), with the CPI molecular weight optimized above 81.4 kDa to ensure processing stability [31]. This ~ 9 µm thick FTCE exhibited excellent optoelectrical properties ($R_{sq} \sim 12.7 \Omega \text{ sq}^{-1}$, $T_{550} \sim 86.3\%$) and outstanding mechanical robustness. Based on this electrode, the resulting ultraflexible organic solar cell achieved a high PCE of 14.37% and retained over 96% of its initial efficiency after 1000 bending cycles at a radius of 0.5 mm. In 2025, Chen et al. reported flexible devices based on ultrathin PI substrates achieving efficiencies exceeding 19% (Figure 3c), with a large-area module also attaining a PCE of 14.61% [12].

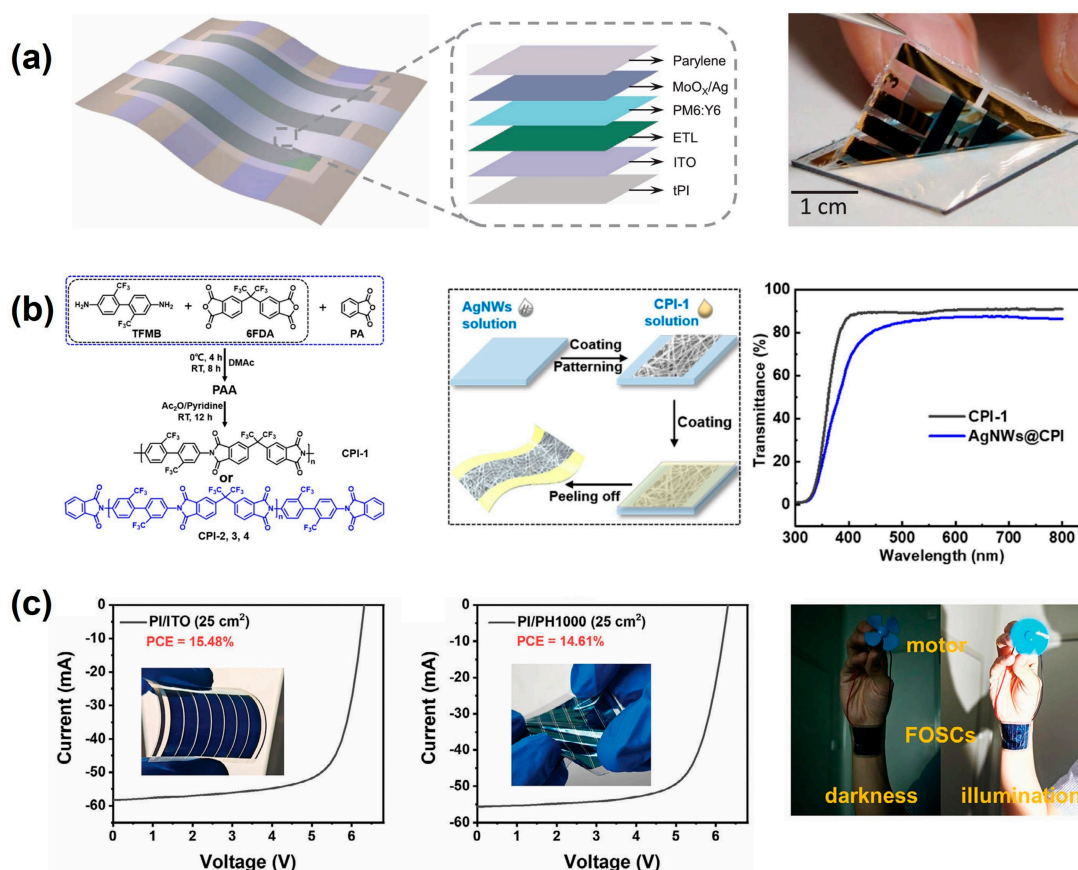


Figure 3. (a) Structure of the ultrathin organic photovoltaics device based on tPI and the photograph of the delamination process of the ultrathin OPV device from glass substrates (Reproduced with permission [30], Copyright 2022, John Wiley and Sons). (b) Schematic two-step synthetic process for the molecular weight-controlled CPIs, schematic of the preparation of the AgNWs@CPI electrode, and optical transmittance of the CPI film (CPI-1) and AgNWs@CPI electrode (Reproduced with permission [31], Copyright 2022, American Chemical Society). (c) The J - V characteristic curves based on 25 cm² substrate flexible and attachable modules, and digital photos of the module's demonstrative applications (Reproduced with permission [12], Copyright 2024, John Wiley and Sons).

However, PI tends to absorb blue light in the visible range, resulting in a yellowish tint and reduced optical transmittance, which may restrict its application in flexible displays and photovoltaic devices. Another limitation lies in its relatively low elastic strain (typically <1%), which is lower than those of most polymer substrates and poses challenges for stretchable electronics or applications requiring large deformation. Furthermore, current PI products exhibit appreciable solubility in certain polar solvents, making them susceptible to swelling or damage during solution-based layer-by-layer fabrication processes. Therefore, low transmittance and poor solvent resistance remain key issues for PI substrates that need to be addressed [32].

2.3. Stretchable Polymer Substrate

2.3.1. Polydimethylsiloxane (PDMS)

Polydimethylsiloxane (PDMS), a commonly used flexible substrate material based on a siloxane structure, belongs to the family of transparent and elastomeric silicone polymers. This class of materials typically exhibits very low permeability to oxygen and water vapor. Furthermore, PDMS demonstrates high stretchability, excellent optical transparency, and good solvent resistance, making it an ideal substrate for stretchable electronic devices [33].

In 2014, Yu et al. fabricated an inverted moth-eye nanopatterned (IMN) PDMS film which served as both a flexible substrate and an efficient antireflection layer for OSCs employing PEDOT:PSS (PH1000) as the transparent electrode [34]. This nanostructured PDMS substrate achieved a high solar-weighted transmittance of $\approx 94.2\%$ and strong hydrophobicity (water contact angle $\approx 120^\circ$). When integrated into encapsulated OSCs, the device delivered a PCE of 6.19% and a short-circuit current density (J_{sc}) of 19.74 mA cm^{-2} , significantly outperforming the reference cell on bare glass (5.16% , 17.25 mA cm^{-2}) (Figure 4a). In 2021, Li et al. developed a stretchable ITO-free organic photovoltaic device by employing a surface-textured PDMS substrate paired with a PEDOT:PSS transparent electrode (Figure 4b). This textured PDMS/PEDOT:PSS architecture enabled high light-harvesting efficiency, yielding a PCE of 15.3% under 1 sun illumination and an impressive indoor PCE of 20.5%. Remarkably, the device exhibited strong angular insensitivity, retaining 38.5% of its initial efficiency at a 10° incident angle, far outperforming rigid glass/ITO-based counterparts [35]. In 2023, Tang et al. demonstrated an effective passive cooling strategy for enhancing the operational stability of FOSCs. By fabricating devices on a PDMS substrate with AgNW electrodes, the team integrated a spectrally selective optical structure composed of a subwavelength hemisphere pattern and a distributed Bragg reflector [36]. This coupled design collectively promoted the out-coupling of infrared radiation while minimizing near-infrared absorption, thereby reducing the steady-state operating temperature by 9.6°C under solar illumination. The resulting FOSCs based on the D18:Y6:PC₇₁BM photoactive system achieved a high PCE of over 17% along with a significantly extended operational lifetime, projecting a T_{80} of more than one year under real outdoor conditions (Figure 4c).

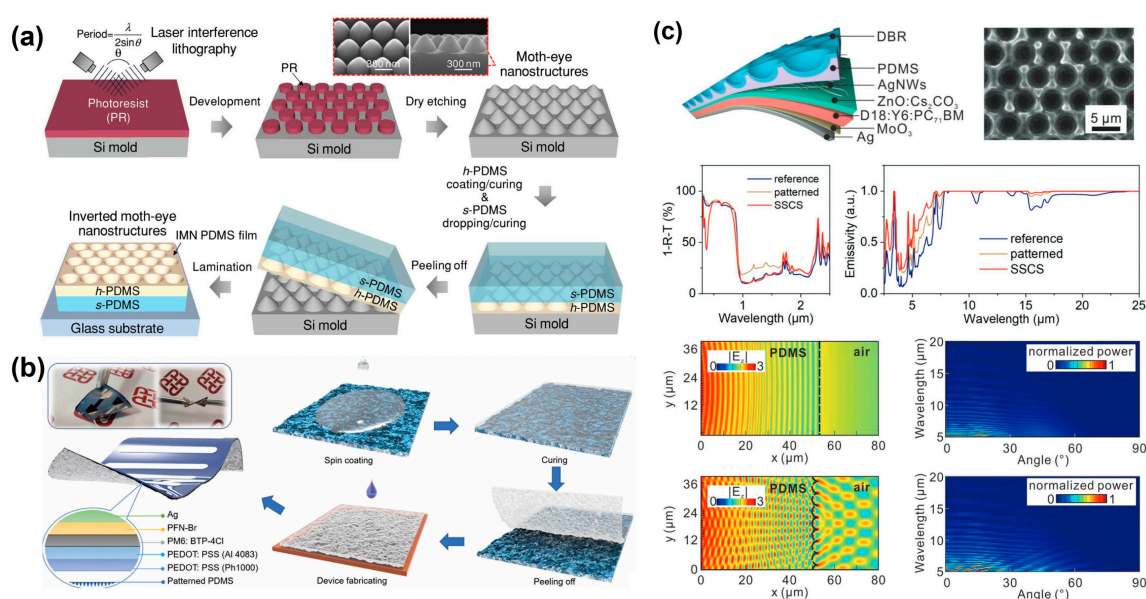


Figure 4. (a) Schematic illustration of process steps for the fabrication of inverted moth-eye nanostructures (IMN) on one side of PDMS films by the soft imprint lithography using a Si master. The IMN PDMS film serves as an AR layer when laminated on glass substrate. SEM images of the Si master with periodic conical subwavelength grating arrays (i.e., moth-eye nanostructures) for a period of 380 nm are also shown (Reproduced with permission [34], Copyright 2014, John Wiley and Sons). (b) Schematic diagram for surface-textured PDMS substrate fabrication and corresponding device structure (Reproduced with permission [35], Copyright 2021, John Wiley and Sons). (c) The influence of optical structure on optical properties of FOSCs (Reproduced with permission [36], Copyright 2023, John Wiley and Sons).

These results confirm the stretchability of PDMS as a flexible substrate; however, its thermal tolerance and mechanical flexibility still require improvement. Furthermore, the low surface energy of PDMS leads to poor wettability, resulting in unsatisfactory film formation of functional layers. Its relatively low glass transition temperature ($T_g \approx -50^\circ\text{C}$) further constrains its processing compatibility in device fabrication [37].

2.3.2. Thermoplastic Polyurethane (TPU)

Thermoplastic polyurethane (TPU) has been employed in OSCs primarily as a flexible substrate, an encapsulation material, or for device structural optimization, aiming to enhance mechanical flexibility, stability, and overall performance. TPU exhibits a smooth surface and favorable wettability, making it suitable as a flexible substrate. Moreover, its high elastic modulus and fatigue resistance further establish TPU as a promising candidate for stretchable and FOSCs [38–40].

In 2021, Lee et al. developed intrinsically stretchable organic solar cells (SOSCs) using a TPU substrate and incorporating the stretchable conductive polymer PH1000 as the transparent bottom electrode, along with a sprayed eutectic gallium–indium (EGaIn) layer as the deformable top electrode (Figure 5a). This all-stretchable architecture enabled the devices to achieve a high PCE exceeding 11% while demonstrating exceptional mechanical robustness, retaining over 74% of their initial PCE after 1000 stretching cycles at 10% strain. The combination of the TPU substrate with the compliant PH1000/EGaIn electrode system effectively reconciles high efficiency with substantial stretchability, marking a significant step toward the practical application of SOSCs in wearable electronics [41].

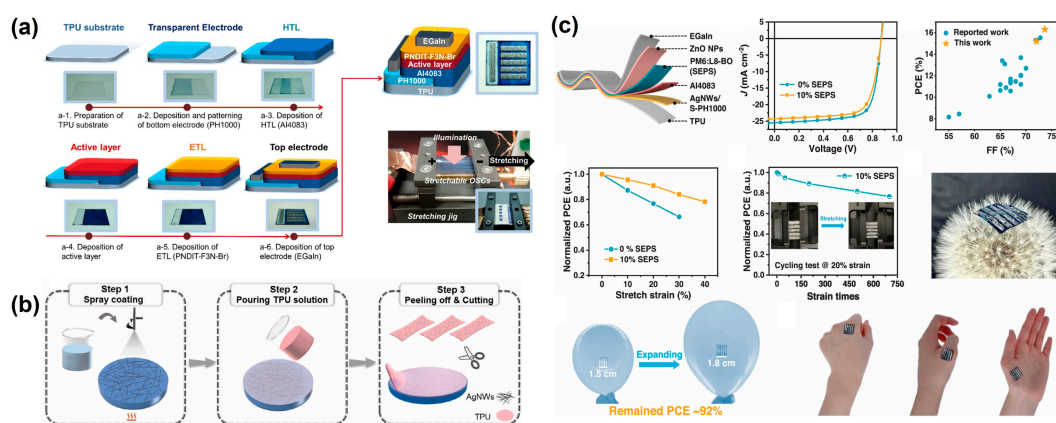


Figure 5. (a) Schematic illustration of the fabrication processes for SOSCs, device structure and an actual image of a SOSC and measurement system for stretching/releasing cycles of SOSC in conjunction with a solar simulator and stretching jig (Reproduced with permission [41], Copyright 2021, American Chemical Society). (b) Schematic diagram of the preparation method of the new stretchable electrode (i.e., Strem-AT) (Reproduced with permission [42], Copyright 2024, John Wiley and Sons). (c) The device structure of is-OPVs; J - V characteristics without and with 10% SEPS contents; comparison of PCE and FF in this work with those of is-OPVs reported in previous studies; normalized PCE change without and with 10% SEPS under different strains; the PCE evolution of is-OPVs with 10% elastomer content blend systems for cyclic stretching at 20% strain; The display of an is-OPV device on the crown of the dandelion, balloon and different parts of a human hand (Reproduced with permission [43], Copyright 2024, John Wiley and Sons).

In 2024, Ye et al. developed a stretchable AgNWs/TPU composite electrode (Strem-AT) using a straightforward spray-transfer embedding strategy. This AgNW-TPU electrode exhibits excellent tensile behavior, low surface roughness, and stable electrical properties [42]. When integrated as a bottom electrode in intrinsically stretchable organic photovoltaics (IS-OPVs), the device achieved a notable PCE of >12.5% and retained 76% of its initial PCE

after 1000 stretching cycles at 50% strain, with a PCE retention of over 50% even under 80% strain. This work demonstrates an efficient and scalable approach to fabricating high-performance stretchable electrodes for durable wearable energy-harvesting applications (Figure 5b). In the same year, Chen et al. proposed a hybrid stretchable electrode based on S-PH1000-AgNWs on TPU [43]. Their is-OPVs reached a high PCE of 16.32% while maintaining 82% of the initial efficiency after 500 stretching cycles (20% strain) and 75% after 390 h of thermal annealing at 85 °C. The ultrathin and ultralight substrate enables the OSCs to be held up by a dandelion as shown in Figure 5c.

Although TPU demonstrates notable advantages as a flexible substrate for OSCs, its practical application is constrained by several inherent limitations. These include a relatively low glass transition temperature ($T_g \approx 80\text{--}120\text{ }^\circ\text{C}$), poor resistance to organic solvents such as chloroform and chlorobenzene, and insufficient optical transmittance, which collectively restrict both processing compatibility and photon absorption in devices. Addressing these shortcomings remains a key direction for improving TPU-based flexible substrates [39,44].

2.4. Bio-Based Substrates

2.4.1. Silk Fibroin

Bioproteins represent an important class of natural polymer materials, featuring remarkable mechanical properties, good biocompatibility, and biodegradability. Silk fibroin (SF), for instance, is a fibrous protein produced by silkworms or spiders, with a polypeptide chain following a repeating sequence of (Gly-Ala-Gly-Ala-Gly-Ser) $_n$ (where Gly is glycine, Ala is alanine, and Ser is serine). The amide groups in this structure facilitate intra- and intermolecular hydrogen bonding, driving the self-assembly of SF into highly ordered nanofibrils with crystalline orientation, which endow the material with high strength and toughness. Furthermore, SF exhibits unique advantages such as light weight and high optical transmittance, enabling its broad application in flexible electronics [45].

In 2014, Sun et al. developed FOSCs using a biocompatible silk fibroin substrate integrated with an embedded AgNW network as the transparent electrode. The AgNW/SF substrate exhibited a sheet resistance of $\sim 11.0\ \Omega\ \text{sq}^{-1}$ and a visible-range transmittance of $\sim 80\%$, while the pristine SF film itself showed high transparency ($\sim 92\%$) due to its amino-acid-based structure (Figure 6a). With this configuration, the OSCs achieved a maximum PCE of 6.62%. The devices also demonstrated notable mechanical flexibility, retaining their conductivity after 200 bending cycles owing to the embedded electrode architecture and the robust interfacial adhesion provided by the SF matrix. Fabricated entirely via aqueous processing under ambient conditions, this work highlights the potential of biocompatible SF-based substrates paired with AgNW electrodes for wearable and skin-compatible photovoltaic applications (Figure 6b) [46].

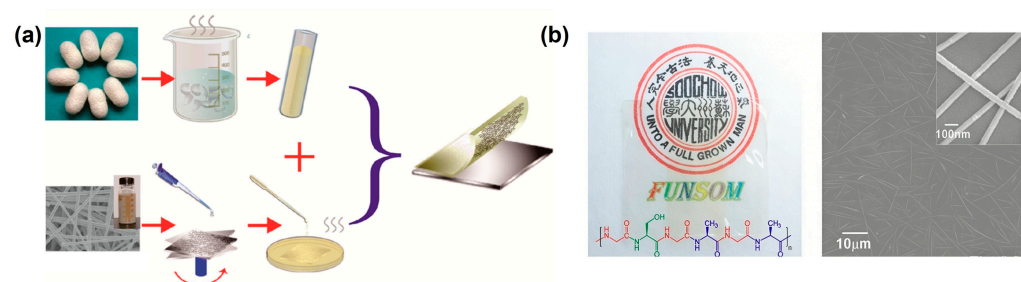


Figure 6. (a) Schematic process of the fabrication of AgNW/SF conductive thin films. (b) Image of the AgNW/SF substrate and the SEM micrograph of AgNWs embedded in SF film and (inset) high-resolution AgNW mesh (Reproduced with permission [46], Copyright 2014, American Chemical Society).

2.4.2. Cellulose

Cellulose, the most abundant natural polymer on earth, is primarily derived from conventional pulping industries. Furthermore, it is an organic polysaccharide with the chemical formula $(C_6H_{10}O_5)_n$, where its repeating unit consists of two glucose rings linked via β -1,4-glycosidic bonds. Strong intermolecular hydrogen bonding drives the self-assembly of cellulose into a hierarchical architecture, forming multi-scale aggregates from nanofibrils to microfibers. In addition to its flexibility, light weight, and biodegradability, cellulose also exhibits a relatively low CTE (28–40 ppm·K^{−1}) and high thermal stability (up to ~250 °C), which are advantageous for the fabrication and outdoor deployment of flexible electronics [47].

To meet the requirements of high transparency and smooth surface for cellulose-based substrates, researchers have developed transparent cellulose materials composed of micro-/nanoscale fibers. By applying high-shear and high-impact treatments using equipment such as grinders, high-pressure homogenizers, intense ultrasonication, high-speed blenders, and twin-screw extruders, cellulose nanofibers (CNF) can be isolated from cellulose pulp. CNF are significantly finer than fibers in conventional paper, with diameters of 4–60 nm and lengths of 0.5–2 μ m. Because their diameter is much smaller than the wavelength of visible light, CNF-based films are highly transparent, allowing light to transmit efficiently from the substrate into the device layers [48].

In 2013, Cui et al. demonstrated the use of nanocellulose paper as a flexible and highly scattering substrate for optoelectronic devices. This material was prepared by disintegrating wood-derived cellulose into nanofibrillated cellulose (NFC) and compressing it into a transparent sheet (Figure 7a). When coated with a 30 nm thick ITO electrode, the CNF/ITO flexible transparent substrate exhibited a transmittance of 80% and a sheet resistance of 12 Ω sq^{−1}. The flexible device based on the P3HT:PCBM active layer achieved a maximum PCE of 0.4% (Figure 7b), only one-tenth that of its rigid counterpart, which is likely attributed to surface inhomogeneity introduced during device fabrication on the CNF substrate [49]. In 2021, Chueh et al. developed a flexible and transparent conductive substrate by embedding AgNWs into TEMPO-oxidized cellulose nanofibers (TOCN). The oxidation process increases the width and decreases the length of the cellulose nanofibers, resulting in higher packing density and reduced AgNW aggregation compared to untreated CNF substrates, which leads to a smoother surface with a roughness of 28 nm. The resulting TOCN/AgNW composite exhibited excellent optoelectrical properties, including a low sheet resistance of 2.62 Ω sq^{−1} and a high optical transmittance of 78.5% at 550 nm, alongside remarkable mechanical flexibility and a low CTE. Flexible organic photovoltaics fabricated on this substrate with the PM6:Y6 active layer achieved a notable PCE of 7.47%, representing the current highest reported efficiency for CNF-based FOSCs (Figure 7c), highlighting the potential of cellulose-based materials as sustainable, high-performance alternatives in wearable electronics [50].

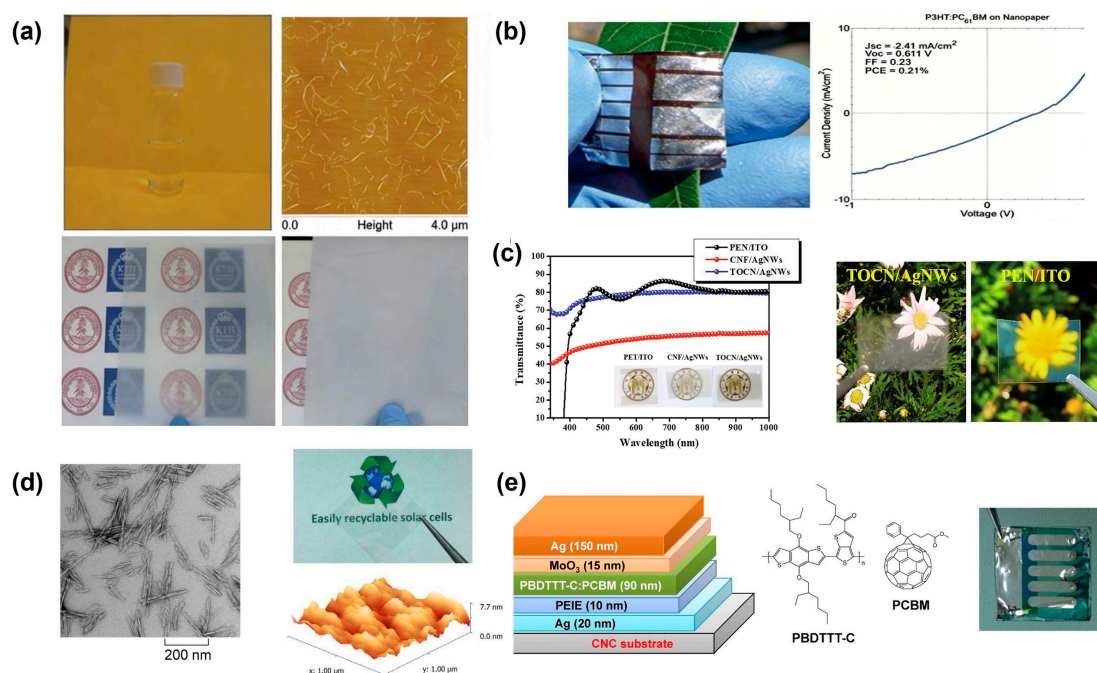


Figure 7. (a) Transparent paper prepared from nanocellulose dispersion. (b) Printed solar cells on transparent and conductive nano paper (Reproduced with permission [49], Copyright 2013, Royal Society of Chemistry). (c) The transmission spectra of the studied conductive substrates and their real photo images (inset) and the photographic images comparison of TOCN/AgNWs and ITO/PEN substrates on the parterre (Reproduced with permission [50], Copyright 2021, Elsevier). (d) TEM image of isolated CNCs which comprise the film used as the substrate for solar cells, a picture of the CNC film on top of paper to demonstrate its transparency, an AFM image of the surface of a CNC film. (e) Device structure of solar cells on CNC substrates: CNC/Ag/PEIE/PBDTTT-C:PCBM/MoO₃/Ag, chemical structure of PBDTTT-C and PCBM and a picture of a fabricated solar cell (Reproduced with permission [51], Copyright 2013, Springer Nature).

To further optimize the surface roughness of cellulose-based flexible substrates, researchers have employed strong acid hydrolysis to treat cellulose pulp. This method selectively degrades the amorphous regions of cellulose while preserving the highly crystalline segments, yielding rod-like materials known as cellulose nanocrystals (CNC). The size and morphology of CNC are closely related to the cellulose source [52,53]. In 2014, Kipelen et al. fabricated CNC substrates for manufacturing efficient and recyclable OSCs [51]. As shown in Figure 7d,e, an 18 μm thick CNC substrate demonstrated an average transmittance of nearly 80% (300–1000 nm) and a very smooth surface with a roughness of $(1.8 \pm 0.6) \text{ nm}$, comparable to commercial PET. The device employing a PBDTTT-C:PCBM active layer, which achieved an average PCE of 2.7%, is superior to FOSC on conventional paper or CNF substrate at the time. Notably, owing to the good water dispersibility of CNC, these FOSCs could be easily recycled through simple water immersion. In a follow-up study, the same group selected PH1000, which offers higher transmittance, as the transparent electrode and used P3HT:ICBA as the active layer. The resulting flexible device based on the CNC substrate exhibited a best PCE of up to 4.0%, comparable to that of control flexible devices on commercial PES substrates [54].

The substantial performance gap between CNF and CNC substrates arises from three key factors. First, surface roughness: CNF films are roughly 20–40 nm rough, while CNC substrates achieve an ultrasmooth surface of only $1.8 \pm 0.6 \text{ nm}$ (comparable to PET), enabling uniform active layer deposition and minimizing shunting losses. Second, morphology and packing density: CNF consists of randomly entangled nanofibers with void spaces that cause light scattering, whereas CNC comprises rod-like crystalline nanoparticles

that self-assemble into densely packed, highly ordered structures with superior optical uniformity. Third, crystallinity: CNC is produced by acid hydrolysis that removes amorphous regions while preserving crystalline segments, yielding higher crystallinity and smoother surfaces. However, this high crystallinity also introduces brittleness, motivating composite approaches with ductile cellulose matrices to improve mechanical properties while retaining surface smoothness. These differences collectively explain why CNC-based devices substantially outperform CNF-based counterparts.

2.4.3. Cellulose Composite Substrates

Although the CNC substrate shows improvements in morphological uniformity, surface smoothness, and crystallinity compared to the CNF substrate, the brittleness resulting from the high crystallinity of both CNF and CNC limits their application in flexible devices. Therefore, some researchers have begun to combine CNC with other celluloses to form polymer composite materials, thereby enhancing the mechanical properties of the substrate [55].

For instance, Zhang et al. developed a transparent nanocomposite paper by incorporating tunicate cellulose nanocrystals (TCNCs) into a ductile cellulose matrix, which exhibited enhanced toughness while maintaining high optical transparency. Using this reinforced cellulose substrate, ITO was directly coated as an electrode without additional adhesives (Figure 8a). The resulting flexible inverted polymer solar cells demonstrated a PCE of up to 4.98%, highlighting the potential of cellulose-based materials in high-performance, flexible electronics. This approach illustrates how optimized substrate–electrode integration can advance the development of biodegradable and wearable optoelectronic devices [56]. In 2024, Wan and Chen et al. developed a high-performance flexible transparent electrode by employing a biocompatible ethyl cellulose (EC) film as the substrate (Figure 8b), which was further optimized with an ultrathin PI coating to improve surface smoothness and mechanical robustness. The conductive component of the FTE consisted of a AgNW network, achieving an excellent optoelectronic performance with a transmittance of 88% at 550 nm and a low sheet resistance of $17.65 \Omega \text{ sq}^{-1}$. FOSCs fabricated on this EC/PI/AgNW electrode demonstrated a remarkable PCE of 18.05%, rivaling the performance of devices on conventional rigid ITO glass. Importantly, these flexible devices exhibited outstanding mechanical durability, retaining 95% of their initial PCE after 5000 bending cycles at an extreme radius of 1 mm. This work underscores the critical role of synergistically engineering both the flexible substrate and the transparent electrode to achieve high-efficiency and robust flexible optoelectronics [57].

To compare the substrate categories summarized in Table 2, each material exhibits a distinct performance balance governed by its chemical structure. Ultra-thin glass, owing to its amorphous network, offers the best overall performance in transparency, thermal stability, and surface smoothness. However, its intrinsic brittleness and high cost severely restrict bending and roll-to-roll processing. PET and PEN are the most commercially mature polymer substrates, combining low cost, good flexibility, and reasonable transparency. Their common drawback is inadequate thermal stability, especially for PET with a glass transition temperature of only about 80 °C, which prevents the use of high-temperature annealing steps. PI is the only polymer that can withstand temperatures above 400 °C while maintaining a sub-millimeter bending radius. Yet its yellowish color and limited transmittance (roughly 80–85%), arising from the conjugated aromatic structure, together with swelling in polar solvents, compromise both optical performance and compatibility with all-solution processing. PDMS and TPU, as elastomers, provide large stretchability (>20% strain) that other substrates cannot match. PDMS suffers from low surface energy and poor dimensional stability, whereas TPU is vulnerable to common organic solvents

like chloroform. Bio-based substrates such as cellulose and silk fibroin are renewable and biodegradable. Cellulose nanocrystals, after acid hydrolysis, can achieve a surface smoothness comparable to PET (RMS $\approx$ 1.8 nm), but their transmittance and moisture resistance still fall short of those of synthetic polymers. Based on these comparisons, the choice of substrate–electrode pairing should follow the application priority. For maximum efficiency, ultrathin glass with metal grids or AgNWs is recommended. For ultrathin and flexible devices, PI with AgNWs or graphene is preferable. For stretchable applications, elastomers with embedded AgNWs are the best fit. For low-cost, large-area manufacturing, PET with solution-processed electrodes offers a practical route. For environmentally friendly electronics, cellulose-based substrates combined with AgNWs represent a promising direction.

Table 1. Summary of photovoltaic parameters of FOSCs based on different flexible substrates.

Substrate	Electrode	Active layer	V_{oc} (V)	J_{sc} (mA cm ^{−2})	FF (%)	PCE (%)	Ref.
Flexible Glass	PEDOT:PSS	BQR:PC ₇₁ BM	0.860	14.20	63.60	7.70	[20]
Flexible Glass	AgNWs/combustion ZnO	PTB7-Th:PC ₇₀ BM	0.800	16.48	64.00	8.48	[21]
PET	PEDOT:PSS	PBDD-T:IT-M	0.930	15.49	70.27	10.12	[27]
PET	FlexAgNE	PBDB-T:F-M/PTB7-Th:PC ₇₁ BM:O6T-4F	0.696	26.85	69.45	13.02	[24]
PET	FlexAgNE	PTB7-Th: PC ₇₁ BM: O6T-4F	1.629	13.99	69.70	16.01	[24]
PEN	PEDOT:PSS	PTB7-Th:PC ₇₁ BM	0.770	14.22	77.00	7.70	[25]
PEN	ITO/PA–ZnO	PBDB-TF:BTP-eC9	0.838	26.41	75.53	16.71	[26]
PI (tPI)	ITO/PEI-Zn	PM6:Y6	0.826	25.70	72.00	15.80	[30]
PI (CPI)	AgNWs@CPI	PM6:BTP-4Cl	0.780	24.77	74.35	14.37	[31]
PI	ITO	PM6: PBQx-TF:PY-IT:PU	0.950	25.46	79.87	19.40	[12]
PDMS (IMN)	PEDOT:PSS	PBDTTT-C:PCBM	0.700	19.74	44.84	6.19	[34]
PDMS	PEDOT:PSS	PM6:BTP-4Cl	0.853	25.00	72.30	15.30	[35]
PDMS (DBR)	AgNWs	D18:Y6:PC ₇₁ BM	0.860	26.94	76.48	17.72	[36]
TPU	PH1000	PM6:Y7	0.820	21.10	65.00	11.20	[41]
TPU	AgNWs	PBQx-TCI:PYF-IT:N2200	0.900	20.86	66.90	12.57	[42]
TPU	AgNWs/S-PH1000	PM6:L8-BO	0.871	25.44	73.66	16.32	[43]
Silk fibroin	AgNWs	PTB7:PC ₇₁ BM	0.700	14.56	65.00	6.62	[46]
Cellulose nanofibers	ITO	P3HT:PC ₆₁ BM	0.378	2.41	23.00	0.21	[49]
TEMPO-oxidized cellulose nanofibers	AgNWs	PM6:Y6	0.811	17.00	54.20	7.47	[50]
Cellulose nanocrystals	Ultrathin Ag	PBDTTT-C:PCBM	0.650	7.50	54.00	2.70	[51]
DHPC/TCNC Nanocomposite Papers	ITO	PTB7:PC ₇₁ BM	0.710	13.65	51.37	4.98	[56]
EC/PI	AgNWs	PM6:L8-BO	0.875	26.38	78.26	18.05	[57]

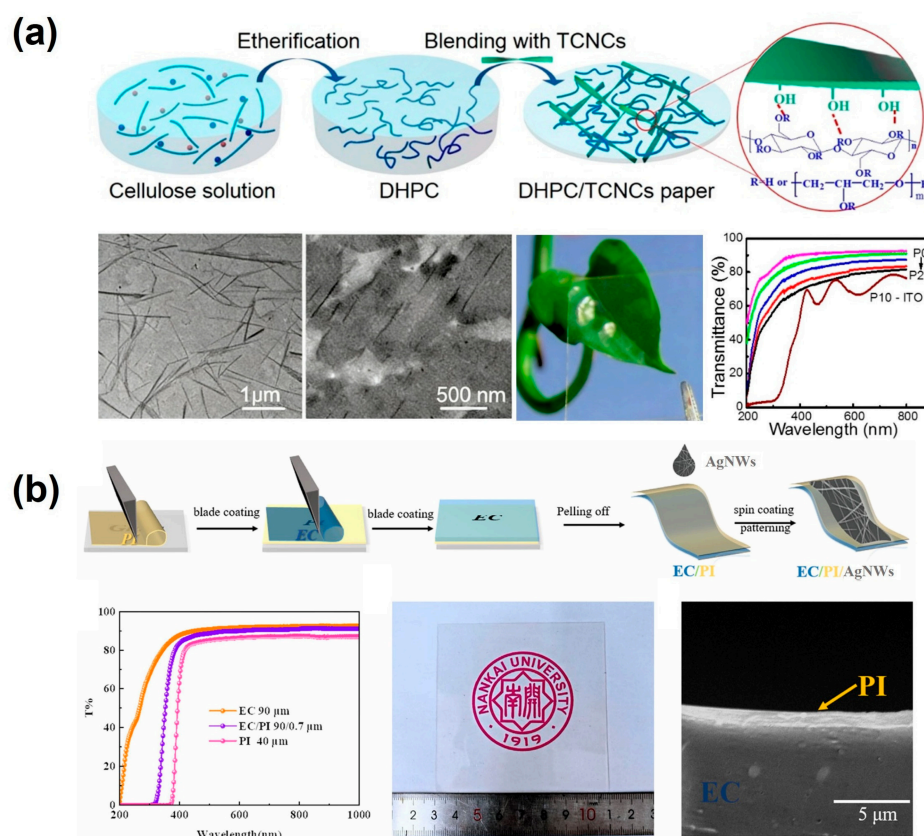


Figure 8. (a) Schematic illustration of the fabrication of cellulose-based nanocomposite papers (Reproduced with permission [56], Copyright 2018, American Chemical Society). (b) Schematic of the fabrication of EC/PI/AgNWs (Reproduced with permission [57], Copyright 2024, John Wiley and Sons).

Table 2. Key performance comparison of the flexible substrates.

Material	Transmittance (@550 nm) (%)	Thermal Stability (°C)	Bending Radius/Flexibility	CTE (ppm/K)	Solvent Resistance
UTG	>90	>600	>5 mm	3–3.5	Excellent
PET	>85	78–120	<1 mm	15–20	Good
PEN	>85	~120	<1 mm	~15	Good
PI	~80–85	>400	<0.5 mm	3–5	Moderate (swelling in some solvents)
PDMS	~92–94	~−50 (T _g)	Stretchable >20%	~300	Good
TPU	~85	80–120	Stretchable >20%	~100	Poor (organic solvents)
Cellulose (CNC)	~80	~250	<5 mm	28–40	Needs crosslinking treatment
Silk Fibroin	~92	N/A	<5 mm	N/A	Good

3. Flexible Transparent Electrodes

3.1. Indium Tin Oxide Transparent Electrode

As a transparent and conductive oxide material, Indium Tin Oxide (ITO, In₂O₃:Sn) films exhibit a bandgap greater than 3.0 eV and possess a high concentration of highly mobile free charge carriers [58]. These characteristics enable ITO to demonstrate excellent electrical conductivity and high transmittance in the visible light spectrum, making it widely adopted as a transparent electrode in optoelectronic devices. However, ITO requires high-temperature processing to enhance its crystallinity and conductivity, which is incompatible with plastic substrates used for flexible electrodes. More critically, for prac-

tical applications in flexible devices, ITO is inherently brittle and prone to cracking under bending. Despite these drawbacks, flexible ITO transparent electrodes remain extensively used in FOSC research, largely because ITO is one of the few commercially established FTEs that dominates the flat-panel display and touch-screen markets.

Although flexible ITO electrodes have been widely employed in flexible OSCs, the brittleness of the ITO layer remains a fundamental limitation. Kim et al. reported on flexible OSCs based on ITO transparent electrodes and investigated their mechanical fatigue resistance [59]. They observed that under bending with a radius of 8 mm, the device efficiency dropped sharply as the number of bending cycles increased. After just five bending cycles, efficiency decreased by 40%, and after 75 cycles, it nearly dropped to zero. The study attributed this decline mainly to the formation and propagation of cracks in the ITO layer, which led to a dramatic increase in sheet resistance and consequently a significant rise in series resistance. The authors further noted that ITO cracking could potentially induce fractures in the overlying layers, including the photoactive layer and the top metal electrode. To address this issue, studies have shown that the strain experienced by the device can be reduced by decreasing the substrate thickness and optimizing the structural design (Figure 9). By adopting this strategy, even when using brittle ITO as the ultrathin FTE in FOSCs, a PCE of 9.4% was achieved after 100 deformation cycles, retaining 88% of its initial efficiency [60]. Table 3 summarizes the efficiencies of flexible organic photovoltaic devices based on different transparent electrodes.

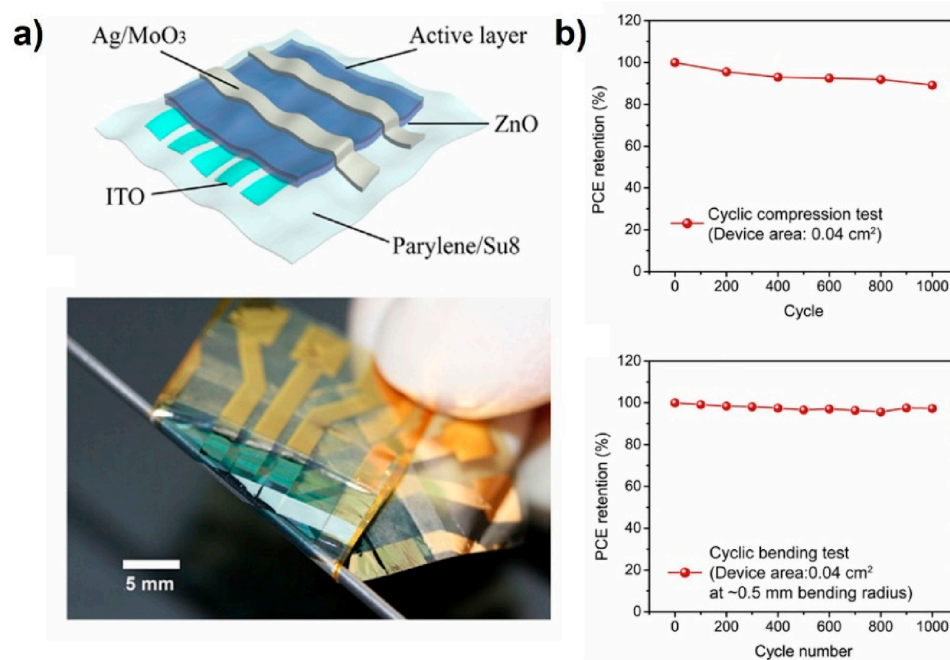


Figure 9. (a) Schematic of the ultra-flexible organic solar cell based on ITO electrode and its photograph. (b) Efficiency of the ultra-flexible organic ternary solar cell with an area of 0.04 cm² as a function of cycle number under the cyclic compression-stretching test and the cyclic bending test (Reproduced with permission [60], Copyright 2020, Elsevier).

However, ITO electrodes are typically fabricated using physical vapor deposition or magnetron sputtering, processes that can easily damage the substrate, thereby limiting efficiency improvement. To address this issue, it is still necessary to identify substrate materials compatible with the ITO fabrication process, with the aim of reducing resistance and thus enhancing device performance.

3.2. Transparent Conductive Polymers Electrode

Transparent conductive polymer materials, owing to their inherent flexibility and compatibility with low-cost roll-to-roll printing production techniques, are promising candidates for flexible transparent electrodes. Water-soluble conductive polymers, represented by poly(3,4-ethylenedioxythiophene):polystyrene sulfonate (PEDOT:PSS; the structure is shown in Figure 10a), are commonly used as electrode materials or hole transport layers in OSCs due to their facile solution processability, high optical transmittance, and excellent mechanical flexibility [61,62]. Among these, Heraeus' Clevios™ series PEDOT:PSS products (Leverkusen, Germany) vary according to conductivity and application scenarios: PH 1000, its eco-friendly aqueous dispersion version PH 1000, and the next-generation PH 750 are all high-conductivity grades (with conductivities reaching 1000–1500 S/cm or higher, which can be further enhanced through secondary doping or acid treatment), specifically designed for high-performance applications such as transparent electrodes; PH 510 exhibits slightly lower conductivity (approximately 500–800 S/cm), yet remains suitable for transparent electrode fabrication due to its good film-forming properties and stability. In contrast, medium-conductivity grades such as P AI 4083 (approximately 0.01–0.1 S/cm, specifically used for hole transport layers) are unsuitable for transparent electrode applications requiring high conductivity due to insufficient electrical performance.

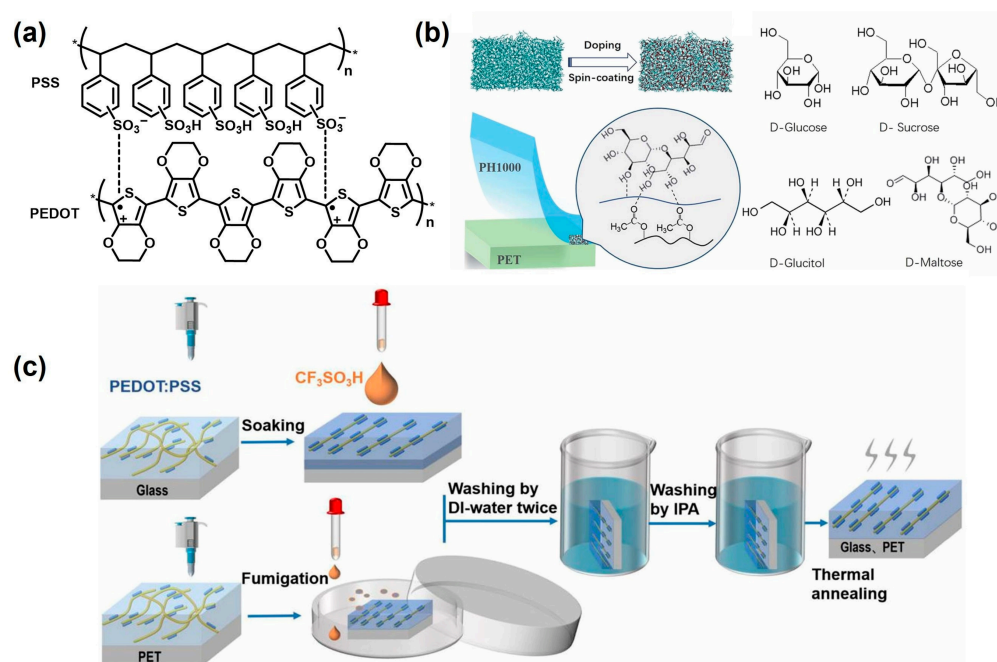


Figure 10. (a) The chemical structure of PEDOT:PSS. (b) The schematic of the preparation procedures for PET/PH1000 FTEs; hydrogen bond and van der Waals bond effect on the PH1000/PET interface is demonstrated in the inset figure and the molecular structures of natural D-glucose, D-sucrose, D-glucitol, and D-maltose, respectively (Reproduced with permission [63], Copyright 2019, Royal Society of Chemistry). (c) Diagrammatic sketch of the flexible PEDOT:PSS preparation via superacid fumigation and soaking treatments (Reproduced with permission [64], Copyright 2021, John Wiley and Sons).

FTE based on conductive polymers, exemplified by the typical PEDOT:PSS, have demonstrated their potential for application in FOSCs. However, the acidic and hygroscopic nature of PEDOT:PSS leads to degradation in device performance, which critically limits its use in OSCs. These issues can be addressed through acid treatment; therefore, it is essential to develop mild acid treatment techniques for fabricating high-performance PEDOT:PSS-based FTEs. In 2019, Ge et al. enhanced the adhesion between PH1000 electrodes and PET

flexible substrates by incorporating hydroxylated polyhydroxy compounds (e.g., D-maltose) into the PH1000 matrix via solution processing. The mixture was spin-coated onto PET, followed by methyl sulfonic acid post-treatment and thermal annealing to form uniform composite electrodes. The hydroxyl groups of D-maltose established strong hydrogen bonds and van der Waals interactions with the carbonyl groups of the PET substrate (Figure 10b), significantly improving interfacial bonding and mechanical flexibility. Based on the PBDB-T-2Cl:IT-4F photoactive system, the resulting D-maltose-doped PET/PH1000 composite electrode achieved a PCE of 12.35% with an open-circuit voltage (V_{oc}) of 0.885 V, a J_{sc} of 19.13 mA/cm², and a fill factor (FF) of 72.92%. Moreover, the flexible device retained 92% of its original PCE after 1000 bending cycles at a radius of 4 mm, demonstrating excellent mechanical stability. This work represents one of the highest efficiencies reported for ITO-free FOSCs at the time [63]. In 2021, Fan et al. developed a novel and cost-effective fumigation treatment using trifluoromethanesulfonic acid (CF₃SO₃H) vapor, yielding a highly conductive, smooth, and flexible PEDOT:PSS electrode. FOSCs fabricated on this treated electrode achieved a notable PCE of 13.62% while retaining approximately 91% of their initial efficiency during long-term storage. This work demonstrated that vapor-phase acid treatment offers a promising route to simultaneously achieve high performance and good stability in solution-processed FOSCs (Figure 10c) [64]. Nonetheless, an intrinsic trade-off between the conductivity and optical transmittance of PEDOT:PSS remains unresolved, rendering it still not an ideal FTE for high-performance flexible devices.

3.3. Carbon-Based Nanomaterials

Carbon-based nanomaterials, such as carbon nanotubes and graphene, offer advantages including abundant resources, high electrical conductivity, excellent transmittance, and outstanding flexibility, making them highly promising electrode materials in the field of organic optoelectronics.

Graphene is a two-dimensional material composed of sp²-hybridized carbon atoms arranged in a honeycomb lattice. It possesses a range of exceptional properties, including inherent high chemical and thermal stability, outstanding mechanical flexibility, and low contact resistance with organic materials, making it well-suited for use as a FTE in flexible OSCs. In 2014, Gradečak et al. achieved flexible devices with PCE of 6.1% and 7.1% for conventional and inverted structures (Figure 11a,b), respectively, using PTB7:PC₇₁BM as the active layer and based on graphene FTE [65]. Subsequently, Jang et al. fabricated tandem FOSCs and obtained a PCE of 8.48% based on FTE of Au-doped single layer graphene nanoribbons (Figure 11c) [66]. In 2020, Park et al. developed a highly functional graphene-based FTE by directly integrating PI onto CVD-grown graphene. Unlike conventional plastic substrates such as PET and PEN, PI offers outstanding thermal stability exceeding 400 °C, along with excellent flexibility and high optical transmittance (Figure 11d). In this approach, PI serves a dual role as both a carrier film for graphene transfer and a flexible substrate, effectively eliminating polymer residues to achieve an ultra-clean graphene surface while enhancing interfacial adhesion through intimate contact. The resulting three-layer stacked graphene electrode on a PI substrate exhibited a low sheet resistance of 83 Ω sq^{−1}, outstanding mechanical durability retaining performance over 10,000 bending cycles, and high optical transmittance exceeding 92%. Consequently, FOSCs based on this PI/graphene electrode achieved a PCE of 15.2%, which is among the highest values reported to date and is comparable to the 15.7% efficiency of ITO-based reference devices on rigid glass substrates [67].

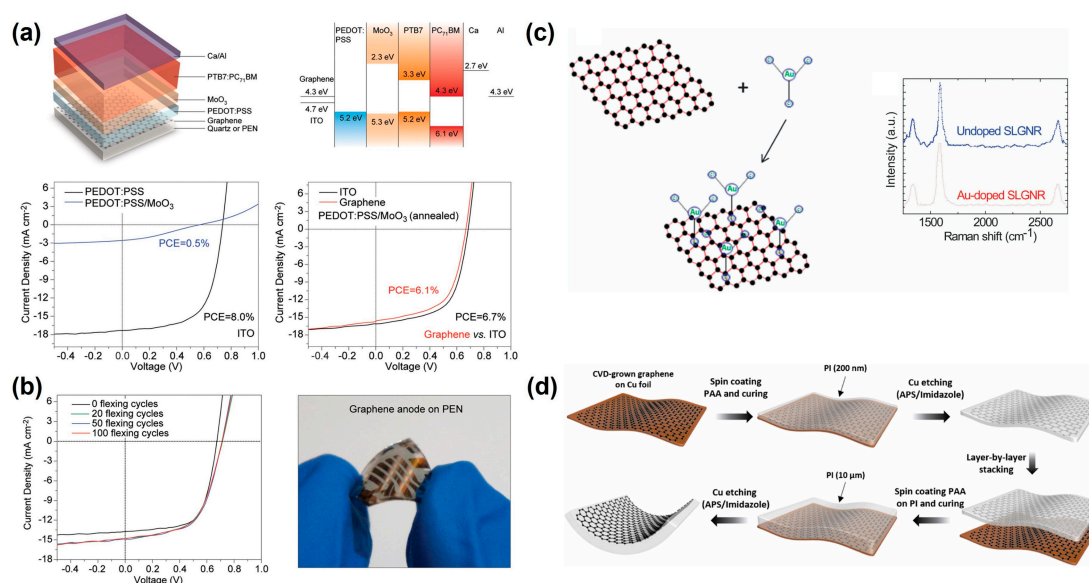


Figure 11. (a) Device structure, energy levels, and performance of graphene- and ITO-based PSCs on quartz. (b) Graphene-based flexible PSCs on PEN (Reproduced with permission [65], Copyright 2014, American Chemical Society). (c) The doping process illustration and Raman spectra of undoped SLGNRs and Au-doped SLGNRs (Reproduced with permission [66], Copyright 2015, Royal Society of Chemistry). (d) Schematic Describing the Two-Step Fabrication Process of PI@GR 200 nm thin PI and 10 mm thick PI were applied as a carrier film during graphene transfer and a substrate for the graphene electrode, respectively (Reproduced with permission [67], Copyright 2020, Elsevier).

Carbon nanotubes (CNTs) have attracted significant attention due to their uniform transmittance across the entire visible range, good electrical conductivity, flexibility, and facile solution processability at room temperature. Compared with other FTE materials, CNT-based electrodes exhibit no characteristic absorption peaks [68,69]. The key requirement for fabricating CNT-based FTEs is that the nanotube density must exceed the percolation threshold of the conductive network. However, high-density CNT conductive films inevitably face a trade-off between reduced sheet resistance and decreased transmittance. The sheet resistance of CNTs is determined by both the contact resistance between overlapping nanotubes and the intrinsic resistance of individual nanotubes, which depends on factors such as nanotube length, diameter, the ratio of metallic to semiconducting CNTs, synthesis method, and purity. Additionally, surface roughness is another critical consideration for CNT-based FTEs in FOSCs, as high roughness can lead to significant shunt resistance losses. Recently, Matsuo et al. reported MoO_x-doped CNT electrodes, where single-walled carbon nanotubes (SWCNTs) grown by floating catalyst chemical vapor deposition were directly deposited via dry process (Figure 12a). These electrodes could be transferred onto plastic substrates such as PI and PET. Using PTB7:PC₇₁BM as the active layer, flexible devices based on this FTE achieved a PCE of 3.91% [70]. In 2018, Matsuo et al. functionalized SWCNTs by incorporating three distinct dopants—tetrafluoroethylene-based fluoropolymer-copolymer sulfonic acid (TFES), trifluoromethanesulfonic acid (TFMS), and nitric acid (HNO₃)—for use as anode materials in conventional OSCs. The devices utilizing TFES-, TFMS-, and HNO₃-doped SWCNTs achieved PCEs of 8.0%, 8.3%, and 8.5%, respectively (Figure 12b). These values significantly surpassed the performance of pristine SWCNT-based devices, which exhibited a PCE of only 4.4%, and were comparable to the ITO-based reference devices with a PCE of 9.1%. The inferior efficiency of the undoped SWCNT devices was primarily attributed to their high series resistance (60 Ω·cm²) and low shunt resistance (320 Ω·cm²), which hindered hole transport and exacerbated charge carrier recombination, thereby limiting both the J_{sc} and FF. Furthermore, stability assessments

revealed that after 60 days of storage in a dark nitrogen atmosphere, devices based on pristine SWCNTs, TFES-SWCNTs, TFMS-SWCNTs, and HNO₃-SWCNTs retained over 70%, 87%, 63%, and 41% of their initial PCEs, respectively, underscoring their excellent long-term operational stability. These findings suggest that chemical doping of CNT-based electrodes represents a promising strategy for the future development of highly efficient, durable, and sustainable optoelectronic devices [71].

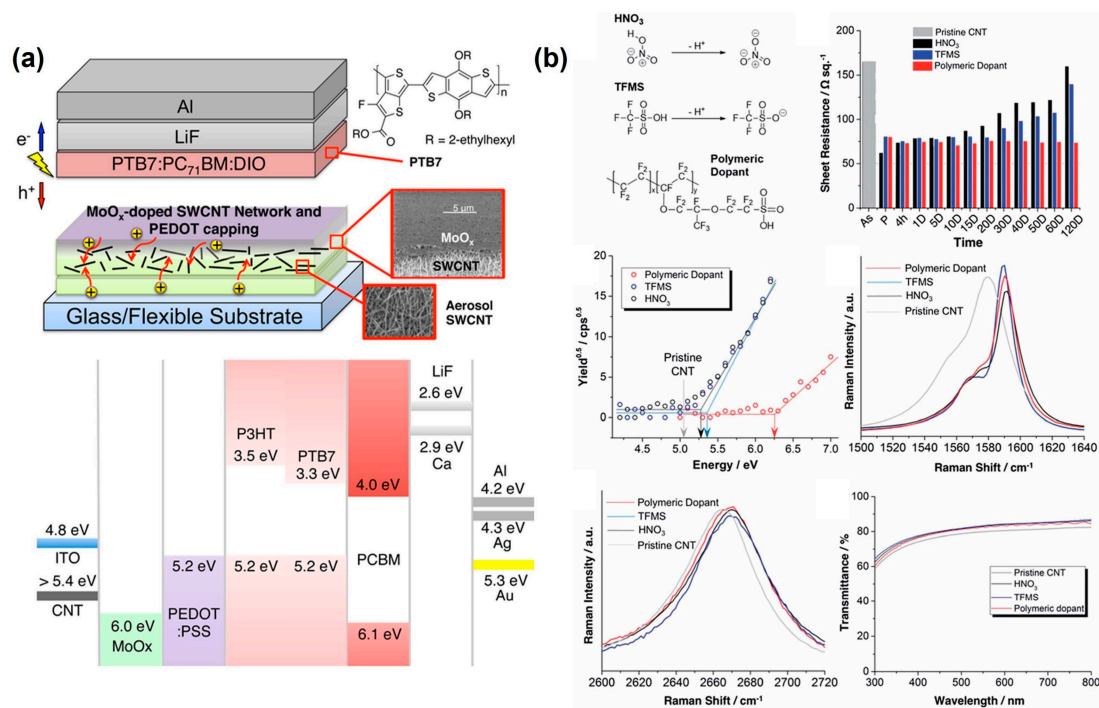


Figure 12. (a) SWCNT OSC configurations of the device (glass or flexible substrate/MoO_x/SWCNT/MoO_x/PEDOT:PSS/PTB7:PC₇₁BM/LiF/Al) and the energy band alignment diagram of SWCNT OSCs (Reproduced with permission [70], Copyright 2015, American Chemical Society). (b) The dopants and the properties of the three types of doped CNT films and the pristine CNT film (Reproduced with permission [71], Copyright 2018, Royal Society of Chemistry).

Despite the widespread application of graphene- and CNT-based FTE in FOSCs, significant challenges remain in enhancing their conductivity and stability. Moreover, the high fabrication cost of CNT- and CVD-grown graphene electrodes is another critical consideration, as low-cost production is essential for future large-scale manufacturing of FTEs.

3.4. Ultra-Thin Metal Films and Metal Grids

Ultra-thin metal electrodes offer both low sheet resistance and excellent mechanical fatigue resistance (thickness ~5–40 nm), and they are compatible with large-area coating processes for metallized plastic films.

Silver (Ag) exhibits the highest electrical conductivity among metals ($\sigma = 6.17 \times 10^7 \text{ S m}^{-1}$) and low optical loss in the visible spectrum (refractive index ~0.1), making it an ideal candidate for transparent electrodes. To function as an FTE, Ag films must be extremely thin to ensure high transmittance. However, ultra-thin metal films (UTMFs) deposited via physical vapor deposition tend to grow in an island-like mode, resulting in large grains that cause high surface roughness and increased resistivity, thereby degrading device performance [72,73]. Thus, low-temperature evaporation is necessary to obtain thin, smooth Ag films suitable for FTEs. Alternatively, transparent metal oxides are often employed as seed layers to enhance wettability and chemical interaction with Ag, enabling

high-performance ultra-thin Ag conductive films. For instance, Lee et al. fabricated a polymer-metal hybrid electrode (PAP electrode, structure shown in Figure 13a by depositing an ultra-thin Ag film (9 nm) onto polyethyleneimine (PEI)-modified PEN [74]. The researchers proposed that PEI provides ideal nucleation sites for the growth of continuous ultra-thin Ag films. Following the deposition of an anti-reflective coating, the resulting FTE exhibited excellent properties, including a bending radius of less than 1 mm, transmittance exceeding 95% in the visible region (referenced to the PEN substrate), and a sheet resistance below $10 \Omega \text{ sq}^{-1}$. Flexible OSCs based on this hybrid FTE, using PTB7-Th:PC₇₁BM as the active layer, achieved a high efficiency of 9.9%, nearly matching that of rigid devices (Glass/hybrid electrode, PCE = 10.1%) and even surpassing ITO-based counterparts (PEN/ITO, PCE = 9.5%; Glass/ITO, PCE = 9.7%).

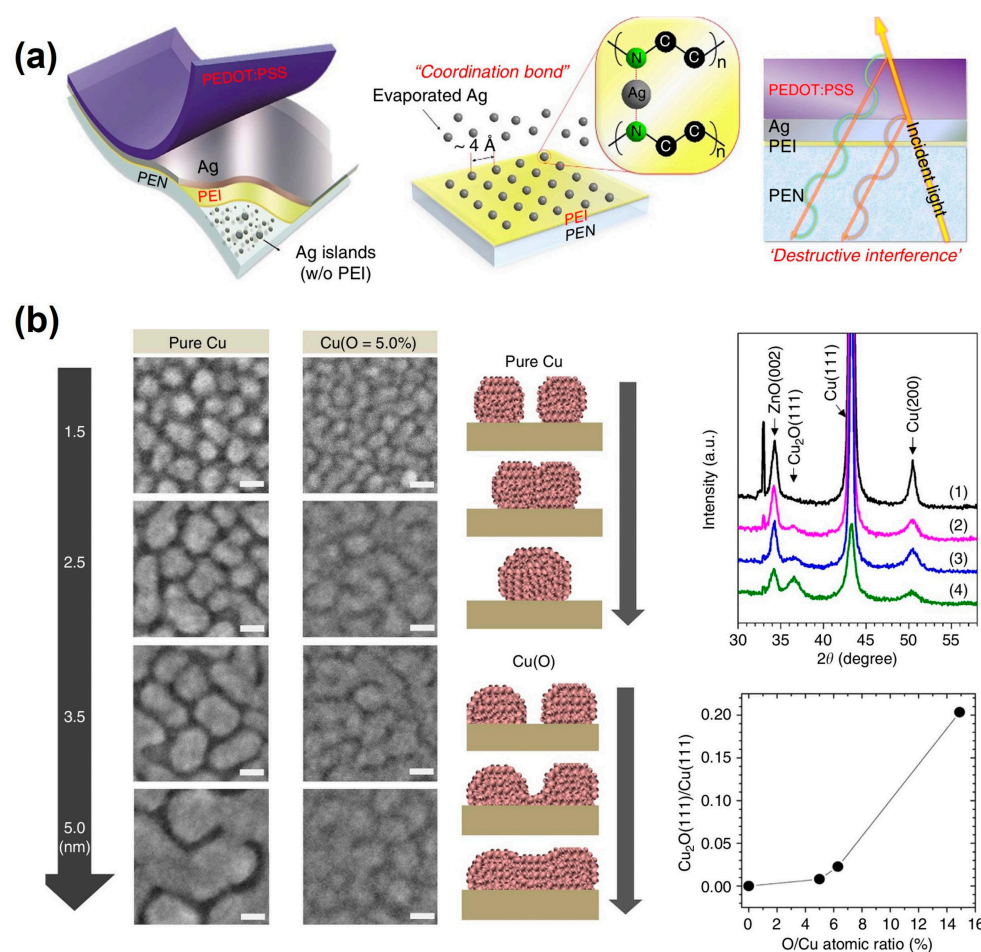


Figure 13. (a) Flexible PAP electrode consisting of the ultra-thin Ag film between the PEI and PEDOT:PSS supporting layers (Reproduced with permission [74], Copyright 2015, Springer Nature). (b) Highly magnified field-emission SEM images showing the coalescence behaviour of Cu and Cu(O = 5.0%) clusters with low thicknesses (1.5–5.0 nm) deposited on ZnO films during the very early stages of growth and the conceptual diagram showing the cluster coalescence mechanism of Cu and Cu(O = 5.0%); X-ray diffraction patterns demonstrating the change in crystallography as a function of O/Cu atomic ratio: (1) 0%, (2) 5.0%, (3) 6.3% and (4) 14.9%; corresponding oxidation levels, as determined by the peak intensity ratio of Cu₂O(111)/Cu(111). Scale bar, 40 nm (Reproduced with permission [75], Copyright 2015, Springer Nature).

In addition, copper (Cu) has garnered attention as the second most conductive metal ($\sigma = 5.96 \times 10^7 \text{ S m}^{-1}$) at only 1% of the cost of Ag. However, due to its susceptibility to oxidation, special conditions are required for the fabrication of ultra-thin Cu-based electrodes. In 2015, Yun et al. demonstrated that copper-based transparent electrodes,

previously limited by low optical transmittance and poor oxidation resistance, could be successfully realized through a controlled oxidation strategy (Figure 13b). By introducing trace amounts of oxygen during deposition, they achieved a fully continuous and smooth ultra-thin copper film. This slightly oxidized copper layer, sandwiched between two zinc oxide films, exhibited excellent optoelectronic properties, including an average transmittance of 83% across the visible spectrum (400–800 nm) and a low sheet resistance of $9 \Omega \text{ sq}^{-1}$, along with robust resistance to oxidation. Notably, FOSCs incorporating this electrode architecture achieved a record PCE of 7.65%, underscoring its potential as an effective transparent electrode solution for flexible photovoltaic devices [75].

Although UTMFs have been widely adopted in FOSCs due to their high electrical conductivity, bulk or thick metal layers are optically opaque, and achieving uniform UTMFs remains challenging, which making them less than ideal for high-performance devices. To overcome this limitation, researchers have developed transparent electrodes with patterned metal grids, which maintain highly conductive pathways through the grid structure while allowing efficient light transmission through the open regions. Typically, these patterned metal grid electrodes are fabricated into two-dimensional stripe or hexagonal mesh structures via thermal evaporation with shadow masks, photolithography patterning, or printing techniques. Metal grid electrodes are typically combined with transparent conductive materials (TCM) like ITO, AZO, or conductive polymers to achieve complementary functionality. The metal grid provides ultra-low sheet resistance for long-range charge transport, while the continuous TCM layer ensures efficient local charge collection from the active area and offers a planar surface for uniform solution processing. Additionally, the TCM helps modulate the electrode's work function for better energy level alignment and mitigates optical shading caused by the opaque grid. This hybrid structure decouples electrical and optical performance, overcoming the individual limitations of each material to enable high-efficiency, large-area optoelectronic devices.

For instances, in 2014, Chen et al. reported a hybrid transparent electrode consisting of an embedded silver grid and PH1000 was fabricated on flexible substrates, achieving a low sheet resistance of $1.2 \Omega \text{ sq}^{-1}$ (Figure 14a). Inverted organic solar cells with an area of 1.21 cm^2 based on this electrode delivered PCEs of 3.36% for P3HT:PC₆₁BM and a record 5.85% for PTB7:PC₇₁BM—the highest reported for large-area (up to 41 cm^2) flexible OSCs at that time [76]. In 2024, Meredith et al. proposed that the recessed metal grid-enhanced transparent electrode (g-TCE) be fabricated using a photolithography-based microfabrication process combined with a planarization technique [77]. Initially, a hexagonal grid pattern is used (with a line width of $40 \mu\text{m}$ and inner radius of 1 mm , Figure 14b is defined via photolithography on a fused silica substrate, followed by dry etching to create $1 \mu\text{m}$ deep trenches). A 200 nm nickel adhesion layer and an 800 nm silver layer are then deposited by sputtering to fill the trenches. After a lift-off process, mechanical polishing is employed to eliminate surface “fences” and ensure a planar surface, reducing the step height between the metal and glass to approximately 10 nm . Finally, a transparent conductive material, such as AZO deposited via molecular vapor deposition (MVD) or AgNWs applied by spin-coating, is coated over the entire substrate. This design offers several key advantages. The embedded structure maintains excellent surface planarity, which is critical for solution-processed layers, preventing shunt defects and ensuring uniform coating. Furthermore, the metallic grid effectively decouples the electrical and optical properties of the electrode, enabling even poorly performing transparent conductive materials to achieve a drastically reduced effective sheet resistance (R_{eff} as low as $0.3 \Omega \text{ sq}^{-1}$) while preserving high transparency. This allows for the use of more economical and sustainable materials. Regarding device performance, the g-TCE demonstrates superior scalability for organic solar cells. Experimental results show that the grid reduces the sheet resistance of AZO

by a factor of ≈ 1700 and AgNWs by a factor of ≈ 100 , outperforming reference ITO films. The optical loss is minimal, with a geometric fill factor (GFF) of 0.96 and a reduction in average visible transmittance (%AVT) of only 2.3%. Consequently, the g-TCE achieves the second-highest figure of merit (FOM_{TCE}) among surveyed materials and significantly outperforms standard ITO under outdoor irradiance conditions, effectively addressing the key scaling barrier for large-area organic photovoltaics. Furthermore, in 2025, a flexible transparent electrode with ultra-low sheet resistance ($<1 \Omega \text{ sq}^{-1}$) and 90% transmittance was developed for OSCs by Wei et al. Using this electrode, a 1 cm^2 single cell achieved a certified efficiency of 16.88%, and a 42 cm^2 module reached 15.60% PCE (Figure 14c). When integrated into solar-powered unmanned aerial vehicles, the devices demonstrated excellent mechanical flexibility and enabled a 24.2% extension in flight time [78].

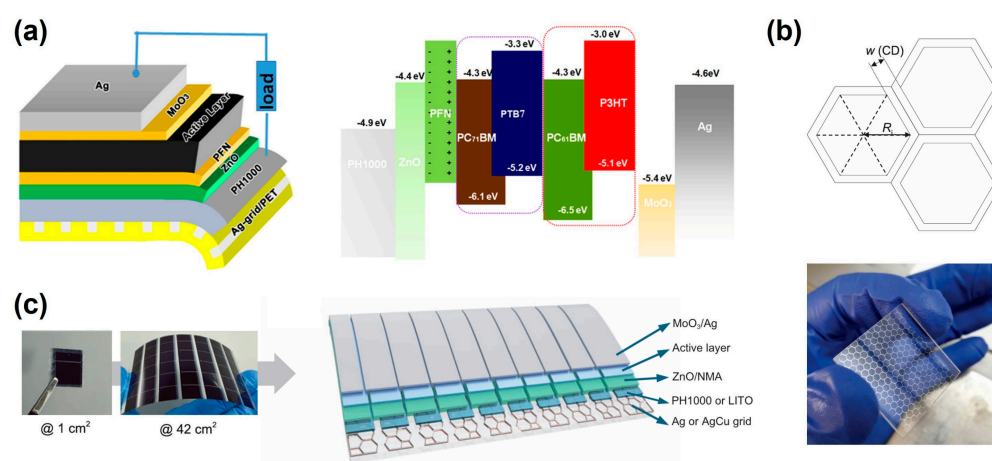


Figure 14. (a) The device structure of the flexible inverted solar cell based on Ag-grid and PH1000 and the energy level diagram of the materials used in the devices (Reproduced with permission [76], Copyright 2014, Elsevier). (b) Geometrical definition of the hexagonal grid and the photograph of $20 \times 25 \text{ mm}$ fabricated metallic grid recessed into $25 \times 25 \times 1 \text{ mm}$ fused silica, with $R_1 = 1 \text{ mm}$, $w = 40 \mu\text{m}$ and thickness = $1 \mu\text{m}$ (Reproduced with permission [77], Copyright 2024, John Wiley and Sons). (c) The images of the single cell and the module, the schematic diagram of the OSC module structure and series connection (Reproduced with permission [78], Copyright 2025, Oxford University Press).

Despite their advantages, metal grid electrodes face several inherent challenges. Additional processing steps are often required to ensure surface planarity and eliminate fabrication defects, increasing manufacturing complexity and cost. The presence of the metal can also alter the effective work function of the electrode, necessitating careful interface engineering to ensure optimal energy level alignment with subsequent layers. Optical losses are difficult to predict with high precision, as the uniformity of the accompanying transparent conductor may vary across the patterned substrate. Furthermore, grid geometry must be tailored to specific applications, as the trade-off between conductivity and transparency shifts depending on operating conditions such as light intensity. Finally, while the electrodes show promise, comprehensive data on long-term stability issues such as metal corrosion or migration, as well as device reproducibility, remain limited.

3.5. Metal Nanowire Networks

Based on metallic nanowires (NWs), particularly AgNWs, the resulting FTEs offer high optical transmittance and electrical conductivity, excellent mechanical fatigue resistance, and compatibility with large-scale solution processing. Common solution-based methods for fabricating AgNW conductive films include Mayer rod coating, roll-to-roll slot-die coating, transfer printing, drop casting, and spray coating—all of which significantly

reduce the overall cost of mass production. For instance, AgNWs present a promising low-cost alternative to ITO in terms of module cost per watt, and FTEs based on AgNWs exhibit an energy payback time nearly 17% lower than that of ITO-based modules. Due to these advantages, AgNWs have gained significant research attention as an emerging material for next-generation FTEs in recent years.

In 2008, Peumans et al. first fabricated randomly dispersed AgNW conductive films via solution processing under low-temperature annealing conditions [79]. Using a hybrid electrode composed of AgNWs and PEDOT:PSS, they constructed vacuum-deposited bilayer OSCs with an active layer of CuPc/PTCBI, achieving a 19% increase in J_{sc} compared to reference devices based on ITO electrodes. Subsequently, You et al. developed AgNWs/PET-based transparent electrodes through simple solution processing and employed them in flexible OSCs to investigate mechanical fatigue resistance [80]. FOSCs based on PBnDT-DTfBT:PC₇₁BM retained 90% of their initial PCE after 10 bending cycles at a curvature radius of 7.2 mm and a bending angle of 120°.

In 2011, Pei et al. discovered that the length of AgNWs significantly influences both sheet resistance and transmittance of FTEs [81]. They developed a composite electrode based on AgNWs embedded in polymethacrylate, where long AgNWs were embedded within the polymer matrix to enhance conductivity, while shorter AgNWs accumulated at the surface to improve surface coverage. This hybrid electrode exhibited a low sheet resistance of 10 $\Omega \text{ sq}^{-1}$ and high visible transmittance of 80%. Flexible OSCs based on P3HT:PCBM achieved a PCE of 3.28%, comparable to rigid devices fabricated on ITO-coated glass. In 2017, Seo et al. developed a simple cold isostatic pressing method to achieve robust junction formation between AgNWs, resulting in a sheet resistance as low as 20.7 $\Omega \text{ sq}^{-1}$ (Figure 15a). Flexible OSCs based on PTB7-Th:PC₇₁BM achieved a PCE of 8.75%, and after 1000 consecutive bending cycles at a radius of 4.8 mm, the devices retained 95.5% of their initial PCE [82].

In 2019, Chen et al. reported a strategy to fabricate flexible transparent electrodes by incorporating poly(sodium 4-styrenesulfonate) (PSSNa) as a polyelectrolyte into an aqueous AgNW suspension. The adsorption of PSSNa imparted a negative charge to the AgNW surfaces, and the resulting ionic electrostatic repulsion induced a grid-like self-assembly during spin-coating, significantly lowering the percolation threshold (Figure 15b). The optimized electrode exhibited a transmittance of 92% at 550 nm (excluding the substrate) and a sheet resistance of approximately 10 $\Omega \text{ sq}^{-1}$. When applied in FOSCs, single-junction devices based on PM6:Y6 achieved a PCE of 13.1%, while tandem devices utilizing PBDB-T:F-M as the front cell and PTB7-Th:O6T-4F as the rear cell reached a PCE of 16.5%, demonstrating performance comparable to rigid ITO-based devices [24].

In 2022, Li et al. developed a controllable chemical welding strategy to achieve atomic-level contact at AgNW junctions by introducing a low concentration of AgNO₃ and an ionic liquid containing Cl[−] and dihydroxyl groups into the AgNW solution. The Cl[−] ions regulated the Ag⁺ concentration through the reversible formation and dissolution of AgCl, while the dihydroxyl group slowly reduced the released Ag⁺, enabling in situ growth of metallic Ag exclusively at the junction sites via a twin-crystal growth mode. This welding process significantly reduced contact resistance, and the resulting electrode (with 30% ionic liquid) exhibited a transmittance of 95% at 550 nm (excluding the substrate) and a sheet resistance of 12 $\Omega \text{ sq}^{-1}$ (Figure 15c). FOSCs based on the PM6:BTP-eC9:PC₇₁BM active layer achieved a record PCE of 17.52% for small-area devices (0.062 cm²) and 15.82% for large-area devices (1 cm²). Moreover, the welded electrodes demonstrated outstanding mechanical stability, retaining 82.5% of the initial PCE after 6000 bending cycles at a radius of 4 mm [83].

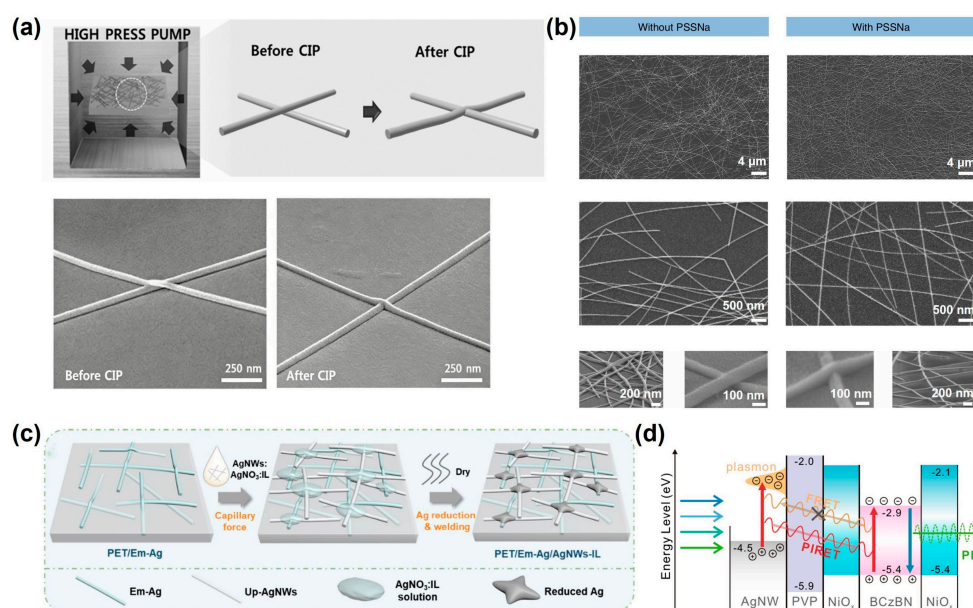


Figure 15. (a) Schematic of the fabrication of a AgNW electrode using cold isostatic pressing (CIP) for AgNW junctions on a suspended plastic substrate in water (Reproduced with permission [82], Copyright 2017, John Wiley and Sons). (b) SEM images of FlexAgNEs prepared from the pristine AgNW suspension (1 mg mL⁻¹) and the AgNW suspension (4 mg mL⁻¹) with PSSNa (10 mg mL⁻¹) (Reproduced with permission [24], Copyright 2019, Springer Nature). (c) Schematic diagram showing the Ag reduction and welding processes involved in the fabrication of AgNW FTEs (Reproduced with permission [83], Copyright 2022, American Chemical Society). (d) Schematic energy level diagram of AgNWs/PEEL and energy transfer dynamics (Reproduced with permission [84], Copyright 2025, Springer Nature).

In 2025, Tang et al. proposed a plasmon energy recycling strategy to mitigate optical losses in FOSCs by incorporating a green fluorophore (BCzBN) into the nickel oxide hole transport layer. This design enables plasmon–exciton energy exchange with AgNW electrodes (Figure 15d), converting parasitic plasmonic absorption into useful emission. The approach improves electrode transmittance by 10.74% and achieves a PCE of 19.51% (certified 18.69%), offering a practical pathway to overcome energy losses in plasmonic optoelectronic devices [84].

As mentioned above, AgNWs have attracted significant attention due to their excellent electrical conductivity and solution processability. Various strategies have also been explored to address issues such as the high roughness of AgNW electrodes. However, there are still many challenges when using AgNWs as ultrathin flexible transparent electrodes, and further optimization of their performance is required to achieve film uniformity, as well as to improve adhesion and stability.

3.6. Emerging Electrode Materials: MXenes, Conductive Hybrids, and Novel Oxides

In addition to the well-established electrode materials discussed above, several emerging candidates have recently attracted considerable attention for flexible transparent electrode applications, including MXenes, conductive hybrids, and novel oxide-based electrodes. Although still in early stages of development, these materials offer unique advantages that may address some of the limitations of conventional electrodes.

MXenes are a family of two-dimensional transition metal carbides and nitrides with the general formula $M_{n+1}X_nT_x$, where M represents a transition metal, X is carbon or nitrogen, and T_x denotes surface terminations [85]. They have garnered significant interest due to their exceptional properties, including metallic conductivity (10^4 – 10^5 S/cm), tunable

bandgap (0.7–2.0 eV), strong optical transmittance (>90%), and a tunable work function (4.0–5.5 eV). Unlike traditional materials such as ITO, MXenes exhibit superior charge carrier mobility, flexible mechanical properties, and excellent chemical stability, ensuring long-term performance and durability in solar cell applications. These characteristics make them highly versatile for use as transparent conductive electrodes, electron/hole transport layers, and even as light-absorbing components in next-generation solar cells.

In the context of flexible organic photovoltaics, several MXene-based transparent electrode schemes have been developed. For instance, Son et al. fabricated a MXene/AgNW/colorless polyimide (cPI) hybrid electrode via a reverse layer-by-layer process, achieving a transmittance of 79% at 550 nm and a sheet resistance as low as 13.08 Ω /sq. Flexible OPV devices based on this electrode achieved a PCE of 10.3%, significantly outperforming MXene/cPI electrodes without AgNWs (6.70%) [86]. Kim et al. reported a multilayer MXene/Ag/MXene structured flexible transparent conductive electrode with a transmittance of 84%, a sheet resistance of only 9.7 Ω /sq, and reliable operation after 2000 bending cycles, enabling OSCs with a PCE of 11.39%. These demonstrations highlight the promising role of MXenes in next-generation flexible optoelectronics [87].

Conductive hybrid materials combine different dimensional conductive components (0D, 1D, 2D) to overcome the performance limitations of single materials. For example, hybrid nanostructured transparent electrodes composed of silver nanoparticles, silver nanowires, and exfoliated graphene sheets exhibit high optical transparency and electrical conductivity, along with good air stability and all-solution low-temperature processability. Flexible non-fullerene OSCs based on such hybrid electrodes achieved an average PCE of 8.15%, with UV stability superior to ITO-based control devices [88]. Ge et al. utilized graphene and silver nanowire composite transparent conductive films (GR&AgNWs) as electrodes, with high-conductivity PEDOT:PSS (PH1000) modifying the rough surface to improve interfacial contact and work function matching. Non-fullerene flexible OSCs based on GR&AgNWs/PH1000 hybrid electrodes achieved a PCE of 13.44% and retained 84.6% of their initial efficiency after 1000 bending cycles at a bending radius of 2 mm [89].

While ITO remains the most widely used transparent conductive oxide, its brittleness and dependence on the rare metal indium have motivated the search for alternative oxide-based electrodes. One promising direction is the development of oxide/metal/oxide (OMO) multilayer structures on flexible plastic substrates. These structures can achieve high transparency and low sheet resistance while maintaining mechanical flexibility. A notable example is the work by Kim et al., who developed an ultra-transparent conducting metal oxide mesh electrode using a self-cracking-assisted template method. The resulting vanadium-doped indium zinc oxide (mIZVO) mesh electrode achieved an exceptional transparency of 97.39%, a low sheet resistance of 21.24 Ω /sq, an elevated work function of 5.16 eV, and good mechanical stability. The unique mesh structure provides favorable stress distribution, ensuring excellent mechanical durability under bending conditions. Flexible OSCs based on the mIZVO electrode achieved a PCE of 14.38%, outperforming conventional ITO or IZO electrode devices. Moreover, the electrode exhibited a resistance change of only 6.30% after 2000 bending cycles at a 2 mm bending radius, demonstrating outstanding mechanical robustness [90]. Other novel oxide electrodes have also been explored. Amorphous V₂O₅-graded indium zinc oxide (VGIZO) films have been investigated as promising flexible anodes for FOSCs, offering a high work function suitable for hole extraction [91].

Overall, these emerging materials each offer distinct advantages. MXenes provide metallic conductivity combined with tunable surface chemistry and mechanical flexibility. Conductive hybrids leverage synergistic effects among multi-dimensional components to surpass the performance boundaries of single materials. Novel oxide electrodes maintain high

transparency and conductivity while significantly improving mechanical flexibility and stability through innovative structural designs. Although challenges remain in terms of large-area uniformity, long-term stability, and cost control, continued progress in materials synthesis, interface engineering, and device integration is expected to position these emerging alternatives as viable replacements for conventional electrodes in future flexible optoelectronics.

Table 3. Summary of photovoltaic parameters of FOSCs based on different flexible transparent electrodes.

Substrate	Electrode	Active layer	V_{oc} (V)	J_{sc} (mA.cm ⁻²)	FF (%)	PCE (%)	Ref.
Parylene/Su8	ITO	PBDTTT-OFT:IEICO-4F:PC ₇₁ BM	0.720	26.10	69.00	13.00	[60]
PET	D-maltose + PH1000	PBDB-T-2Cl:IT-4F	0.885	19.13	72.92	12.35	[63]
PET	CF ₃ SO ₃ H-fumigated PEDOT: PSS	PM6:Y6	0.830	24.80	66.20	13.62	[64]
PEN	Graphene	PTB7:PC ₇₁ BM	0.720	14.10	69.50	7.10	[65]
Glass	Au-doped single layer graphene nanoribbons	SMPV1:PC ₇₁ BM/PTTBDT-FTT:PC ₇₁ BM/	1.560	8.45	64.32	8.48	[66]
PI	PI@Graphene	PM6:Y6	0.840	25.80	70.20	15.20	[67]
PET	SWCNT/MoO ₃ /PEDOT:PSS	PTB7:PC ₇₁ BM	0.690	12.60	45.00	3.91	[70]
Glass	HNO ₃ -CNT	PBTZT-stat-BDTT-8:PC ₇₁ BM	0.810	14.20	74.00	8.50	[71]
PEN	PEI/Ag/PEDOT:PSS	PTB7-Th:PC ₇₁ BM	0.790	16.94	74.00	9.90	[74]
PET	ZnO/Cu(O = 5.0%)/ZnO	PTB7-Th:PC ₇₁ BM	0.780	16.52	57.91	7.65	[75]
PET	Ag-grid/PH1000	PTB7:PC ₇₁ BM	0.720	13.60	60.00	6.01	[76]
PET	AgCu-grid/LITO	PM6:TrimerC8C10	0.895	24.37	78.42	17.12	[78]
PET	AgNWs	PBnDT-DTfBT:PC ₇₁ BM	0.590	9.64	48.00	2.80	[80]
PET	CIP-AgNWs	PTB7-Th:PC ₇₁ BM	0.764	17.40	64.20	8.56	[82]
PET	PSSNa-AgNWs	PBDB-T:F-M/PTB7-Th:PC ₇₁ BM:O6T-4F	1.629	13.99	69.70	16.01	[24]
PET	Em-Ag/AgNWs	PM6:BTP-eC9:PC ₇₁ BM	0.835	27.37	76.90	17.52	[83]
PET	AgNWs/BCzBN-doped NiOx	D18:L8-BO	0.908	26.78	80.17	19.51	[84]
cPI	MXene/AgNW	PM6:N3	0.780	23.70	62.70	10.70	[86]
PEN	MXene/Ag/MXene	PM6:Y6:PC ₇₁ BM	0.561	28.41	71.47	11.39	[87]
PET	AgNWs:AgNPs/graphene	PBDB-T:ITIC	0.890	13.40	67.70	8.15	[88]
PET	GR&AgNWs/PH1000/PEDOT:PSS	PM6:Y6	0.829	23.20	69.98	13.44	[89]
PEN	mIZVO	PM6:Y6:PC ₇₁ BM	0.724	28.39	70.00	14.38	[90]
PET	VGIZO	P3HT:PCBM	0.571	8.104	59.48	2.75	[91]

To facilitate a comparative assessment of the transparent electrode materials summarized in Table 4, we have compiled their key performance metrics, including sheet resistance, optical transmittance, bending radius and solution processability. ITO remains the performance benchmark with low sheet resistance ($\sim 10 \Omega/\text{sq}$) and high transparency ($>85\%$), benefiting from its high carrier concentration and wide bandgap. However, its ceramic nature causes fracture strain below 1%, leading to rapid performance degradation under bending, while the high processing temperature ($>300^\circ\text{C}$) is incompatible with most polymer substrates. PEDOT:PSS enables all-solution processing and intrinsic flexibility, with conductivity reaching 1000–1500 S/cm after acid treatment, yet its hopping conduction mechanism creates an inherent trade-off between conductivity and transparency, and its acidic hygroscopic nature accelerates device degradation. Carbon nanomaterials such as graphene and CNTs provide excellent flexibility and chemical stability, with single-layer graphene showing 97.7% transmittance, but their conductivity is constrained by different limitations: grain boundaries and polymer residues in CVD graphene scatter charge carriers, while CNT networks suffer from high junction resistance that is orders of magnitude higher than individual tube resistance, resulting in performance far below metallic electrodes. Ultra-thin metal films, particularly Ag and Cu, possess the highest intrinsic conductivity, with 9 nm Ag films on seed layers achieving $<10 \Omega/\text{sq}$ and $>95\%$ transmittance, yet their fabrication is challenged by island growth on amorphous surfaces, requiring additional seed layers or co-evaporation of oxygen to improve wettability, which increases process complexity. Metal grids are the only structure achieving sheet resistance below $1 \Omega/\text{sq}$ with over 90% transmittance by decoupling conduction and transmission, but they face three major challenges: surface unevenness causing shorting during solution coating, high manufacturing cost due to lithography steps, and the need for delicate interface engineering between the metal and the transparent conductive overcoat. AgNWs combine high conductivity, nanowire flexibility, and solution processability, attaining $\sim 10 \Omega/\text{sq}$ and $>92\%$ transmittance, with the network retaining 82% efficiency after 6000 bending cycles at 4 mm radius through stress release via sliding and rearranging. Nevertheless, AgNWs encounter issues of high surface roughness (RMS 10–30 nm) leading to shunting, large junction contact resistance requiring chemical welding, and susceptibility to oxidation and electrochemical migration, which are being addressed by embedding strategies and plasmonic energy recycling.

Table 4. Key performance comparison of the flexible electrodes.

Material	Sheet Resistance (Ω/sq)	Transmittance (@550 nm) (%)	Bending Radius (mm)	Solution Processability
ITO	~ 10	>85	>5 (brittle)	No (sputtering)
PEDOT:PSS (PH1000)	$\sim 100\text{--}300$ (treated ~ 50)	>90	<1	Yes
Graphene	$\sim 80\text{--}100$	>92	<1	No (CVD/transfer)
CNT	$\sim 50\text{--}200$	>85	<1	Yes
Ultra-thin Ag	<10	>95 (with AR)	<1	No (evaporation)
Metal Grid	<1	~ 90	<1	No (lithography)
AgNW	$\sim 10\text{--}20$	~ 92	<1	Yes
MXenes	$\sim 10\text{--}21$	$\sim 79\text{--}84\%$	<1	Yes
Conductive Hybrids	$\sim 10\text{--}20$	$\sim 85\text{--}90\%$	<1	Yes
Novel Oxides	~ 21	$>97\%$ (with mesh structure)	<1	Limited (requires templating)

In addition to these established options, emerging materials such as MXenes, conductive hybrids, and novel oxides have recently attracted interest for flexible transparent electrodes. MXenes combine metallic conductivity with solution processability, achieving sheet resistance down to $\sim 10\text{--}20\ \Omega/\text{sq}$ and transmittance around 80–85%, yet their oxidation susceptibility and synthesis complexity remain key obstacles. Conductive hybrids, which integrate multiple components such as graphene, AgNWs, or conductive polymers, exploit synergistic effects to attain balanced optoelectrical and mechanical performance, but their multi-component stability and interface engineering require further investigation. Novel oxide electrodes, particularly those with structured mesh designs, can deliver ultra-high transmittance ($>97\%$) with low sheet resistance and good bending durability, though their fabrication often relies on complex templating processes that limit scalability.

Based on these comparisons, ITO remains suitable for rigid high-efficiency devices; AgNWs offer the best overall balance for flexible high-performance applications; metal grids excel in large-area modules requiring ultra-low resistance; PEDOT:PSS fits all-solution, low-cost production; ultra-thin Ag films are suited for ultra-flexible form factors; and graphene or CNTs are promising for future exploration but require further cost and performance improvements. MXenes, hybrids, and novel oxides represent promising long-term directions, though they are not yet competitive with established technologies for immediate deployment.

4. Synergistic Design and Integration Strategies

The preceding analysis of various flexible substrates and transparent electrodes has outlined their respective strengths and limitations. However, achieving high-performance FOSCs moves beyond simply optimizing individual components. Instead, it requires a holistic, synergistic design approach where the substrate and electrode are conceived and engineered together, as an integrated system (Figure 16 presents a radar-chart comparison of the overall performance of different flexible substrates and typical flexible transparent electrodes fabricated on them, evaluated across multiple key performance indicators). Their combined properties govern not only the initial PCE but also the mechanical robustness, operational stability, and scalability of the final device. Achieving this synergy requires deliberate co-design across multiple dimensions, including interface engineering, mechanical compatibility, optical coupling, and process integration, each offering strategic opportunities to close the performance gap that still exists between devices made on rigid and flexible platforms.

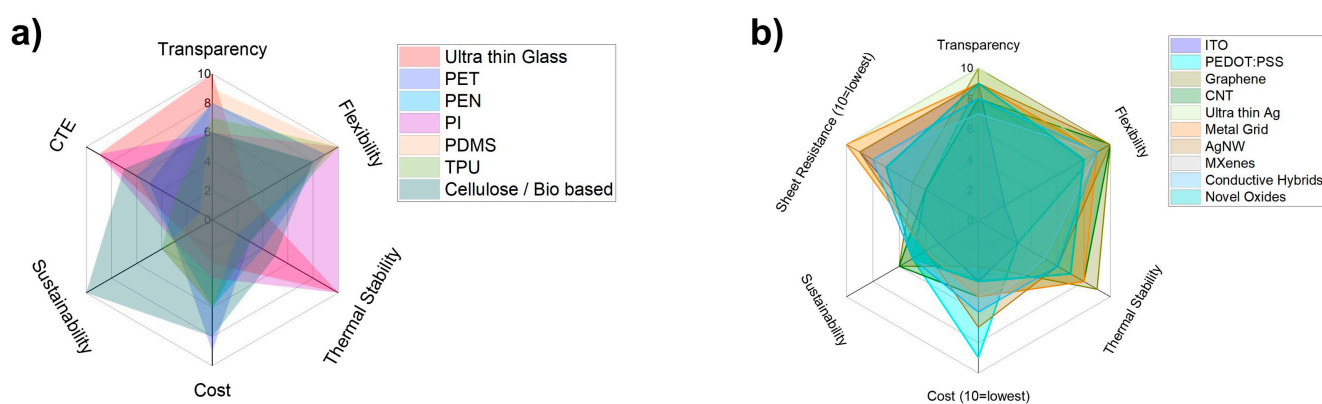


Figure 16. Radar chart comparing the performance of (a) flexible substrate materials and (b) flexible electrode materials across six key metrics.

4.1. Interface Engineering

The interface between the flexible substrate and the transparent electrode is a critical junction where mechanical, chemical, and electronic properties converge. Effective interface engineering is paramount to ensure strong adhesion, minimize contact resistance, and provide a stable foundation for subsequent layers. A weak interface can lead to delamination under mechanical stress, while a poorly matched electronic interface can create energy barriers that impede charge extraction.

One primary goal of interface engineering is to enhance adhesion and mitigate mechanical stress. The inherent difference in surface energy between a hydrophobic polymer substrate like PET and a hydrophilic conductive layer like PEDOT:PSS often results in poor adhesion. To address this, researchers have employed chemical strategies to bridge this gap. For instance, as highlighted by Ge et al., incorporating hydroxylated compounds such as D-maltose into a PH1000 solution facilitates the formation of strong hydrogen bonds and van der Waals interactions with the carbonyl groups on the PET substrate [62]. This molecular-level “bridging” significantly improves interfacial bonding, leading to enhanced mechanical flexibility and device stability. Similarly, for ultra-thin metal films, the substrate surface can be modified to promote favorable growth kinetics. The work by Lee et al. demonstrated that a thin polyethyleneimine layer on a PEN substrate provides ideal nucleation sites for silver atoms. This seed layer alters the growth mode of the subsequently deposited ultra-thin Ag film from an island-like (Volmer–Weber) to a more layer-by-layer (Frank–van der Merwe) fashion, yielding a smooth, continuous, and highly conductive electrode that would otherwise be unattainable [74].

Beyond adhesion, the interface dictates charge carrier dynamics. The work function of the electrode must be aligned with the energy levels of the adjacent charge transport layer to ensure ohmic contact and minimize recombination losses. Surface treatments on the electrode itself can be viewed as an extension of this interfacial strategy. The work by Ge et al. exemplifies this, where a gentle acid treatment of a PEDOT:PSS electrode on PET was used to modulate its work function to approximately 4.91 eV, optimizing hole extraction to the photoactive layer. This demonstrates that interface engineering is not passive; it is an active tool for tuning the device’s electronic landscape [27].

4.2. Mechanical Compatibility

The flexibility of an FOSC is not determined by its most pliable layer but by its most fragile interface or component. A mismatch in the mechanical properties of the substrate and electrode can lead to catastrophic failure under bending or stretching. Therefore, ensuring mechanical compatibility is a cornerstone of synergistic design. This compatibility must be considered across multiple mechanical parameters, including elastic modulus, ductility, and the CTE.

The most instructive example of mechanical incompatibility is the pairing of a brittle electrode like ITO with a flexible polymer substrate. As documented by Kim et al., ITO on PET suffers from crack formation and propagation under even modest bending cycles (e.g., a 40% efficiency drop after just five bends at an 8 mm radius). This dramatic failure occurs because the high elastic modulus and low fracture strain of ITO cannot accommodate the tensile strain imposed on the PET substrate. The cracks in the ITO layer not only increase its own sheet resistance but can also propagate into the overlying active and metal electrode layers, leading to catastrophic device failure. This fundamental limitation has been a primary driver for the development of inherently flexible electrode materials, such as conductive polymers and metal nanowires, which can better conform to the deformation of the substrate [58].

For a truly robust system, the CTEs of the substrate and electrode should also be closely matched. Temperature changes during processing (e.g., annealing of transport layers) or outdoor operation can generate significant thermal stress at the interface if the materials expand or contract at different rates. This stress can lead to delamination or micro-cracking, undermining long-term stability. In this context, cellulose nanofiber-based substrates offer a distinct advantage. As noted by Chueh et al. in their work on TOCN/AgNW composites, cellulose-based materials can exhibit a remarkably low CTE (e.g., $3.37 \times 10^{-6} \text{ K}^{-1}$), which is much closer to that of metals and metal oxides than typical petroleum-based polymers like PET. This intrinsic CTE compatibility helps to minimize thermomechanical fatigue, contributing to the enhanced durability of the device [50].

4.3. Optical Coupling

In a conventional FOSC, light must traverse both the substrate and the electrode before reaching the photoactive layer. A simplistic view would aim to maximize the transmittance of each component independently. However, a synergistic approach recognizes that these layers can interact optically to produce a net effect greater than the sum of their parts. This interaction can be harnessed through advanced light management strategies, such as anti-reflection, light scattering, and even the recycling of parasitic losses.

One powerful strategy is to engineer the substrate itself to function as an optical element. Instead of being a passive window, the substrate can be patterned to reduce surface reflection and enhance light incoupling. A prime example is the work by Yu et al., who fabricated an inverted moth-eye nanopattern on a PDMS substrate. This sub-wavelength structure creates a graded refractive index profile, effectively suppressing reflection across a broad spectrum and increasing the solar-weighted transmittance to ~94.2%. The substrate thus actively contributes to enhancing the photocurrent of the device, demonstrating a direct synergy between its physical form and optical function [34].

Optical synergy can also be achieved within the electrode structure itself. For instance, the work by Han et al. on a ZnO/AgNW hybrid electrode revealed an additional optical benefit. The nanopores present in the combustion-derived ZnO layer were found to scatter light, particularly in the 450–550 nm range. This scattering increases the optical path length of photons within the active layer, boosting the probability of absorption. In this case, the ZnO layer serves a triple function: as a seed layer for the AgNWs, as an electron transport layer, and as a light-scattering medium, all synergistically integrated [22].

Pushing the boundaries of optical synergy further, recent research has sought to reclaim energy typically lost to parasitic absorption. Metal electrodes, particularly AgNW networks, suffer from plasmonic absorption losses. In a groundbreaking example of synergistic design, Tang et al. proposed a “plasmon energy recycling” strategy. By incorporating a green fluorophore into the nickel oxide hole transport layer atop an AgNW electrode, they enabled a plasmon–exciton energy exchange. The energy typically lost to heating the electrode was instead transferred to the fluorophore and re-emitted as light, which could then be absorbed by the active layer. This sophisticated approach transforms a parasitic loss mechanism into a beneficial gain channel, representing the ultimate level of optical-functional synergy [84].

4.4. Process Compatibility

The final and practically most critical dimension of synergistic design is process compatibility. The choice of substrate fundamentally dictates the manufacturing processes that can be used to deposit and pattern the electrode and subsequent layers. A design that is elegant in theory but impossible to manufacture at scale has limited practical value. Therefore, the thermal, chemical, and mechanical tolerances of the substrate and

electrode must be co-optimized with an eye toward scalable production techniques like R2R processing.

Thermal stability is a primary constraint. Many high-performance metal oxide transport layers, and even some advanced electrode treatments, require elevated temperatures for annealing or crystallization. Standard PET substrates, with a T_g around 78–120 °C, cannot withstand these processes. This limitation was explicitly addressed by Hou et al., who developed a low-temperature (130 °C) sol–gel route for ZnO [30]. However, even this reduced temperature proved too high for PET, forcing the use of a more thermally robust (and expensive) PEN/ITO substrate. This case illustrates how the substrate's thermal budget directly dictates the permissible processing window. Conversely, materials like polyimide offer a high-temperature pathway, enabling unique fabrication strategies. The work by Park et al. leveraged the >400 °C thermal stability of PI to directly integrate it with CVD-grown graphene, eliminating the need for a separate, contamination-prone transfer process and resulting in an ultra-clean, high-performance electrode [66].

Chemical compatibility is equally important for solution-processed devices. The substrate must be resistant to the solvents used in subsequent coating steps, such as chlorobenzene or chloroform for the active layer, to prevent swelling, cracking, or dissolution. Here, ultrathin glass offers a distinct advantage due to its complete solvent resistance, as noted by Yu et al., simplifying the processing sequence [21]. In contrast, as Yu correctly pointed out, PI can be susceptible to swelling in certain polar solvents, presenting a significant challenge for all-solution-processed device fabrication on this otherwise attractive substrate [33]. This challenge necessitates further synergistic strategies, such as the development of cross-linked PI or the introduction of protective barrier layers, to reconcile material advantages with processing realities.

4.5. Operational and Environmental Stability

Operational stability under continuous illumination and environmental stability against moisture, oxygen, and thermal stress are among the most critical barriers to the practical deployment of FOSCs. While mechanical flexibility has been extensively studied, a systematic comparison of operational and environmental stability across different substrate–electrode combinations is equally essential.

A direct comparison between AgNW and PEDOT:PSS front electrodes was conducted by Krebs et al., who evaluated roll-to-roll-printed flexible OPV modules under multiple ISOS protocols over 140 to 360 days [92]. The results showed that replacing PEDOT:PSS with AgNW as the front electrode improved operational stability by up to 1000%, with the AgNW front electrode combined with a PEDOT:PSS/Ag grid composite back electrode exhibiting the best overall performance.

The choice of substrate material also plays a decisive role. Min et al. systematically compared ultra-flexible OSCs based on PI/ITO and PET/ITO integrated electrodes under thermal and bending stress [93]. The PI-based devices consistently outperformed their PET-based counterparts, demonstrating superior stability under both stress conditions. Yu et al. directly compared ITO-free flexible OSCs on ultra-thin flexible glass, PEN/ITO, and PEN/PEDOT:PSS substrates [20]. The flexible glass devices achieved a PCE of 8.0%, outperforming PEN/ITO (6.4%) and PEN/PEDOT:PSS (5.8%). Critically, the authors pointed out that the acidity of PEDOT:PSS can corrode polyester substrates such as PET and PEN, leading to device degradation, whereas flexible glass completely avoids this issue. Park et al. further demonstrated that polyimide, with its exceptional thermal stability, can be directly integrated with CVD-grown graphene to form a PI@GR hybrid electrode. The resulting flexible OSCs achieved 15.2% efficiency and maintained stable performance under

thermal stress, highlighting the advantage of PI over conventional polymer substrates for high-temperature compatibility [67].

Encapsulation strategy is another critical factor governing environmental stability. Urbina et al. performed a systematic lifetime study of P3HT:O-IDTBR OSCs following ISOS protocols, directly comparing glass-to-glass versus plastic-to-plastic encapsulation [94]. The glass-encapsulated devices exhibited an exponential decay with a T_{80} lifetime of 86 h under ISOS-T3 accelerated thermal testing, while plastic encapsulation showed distinct decay behavior with attenuation constants of $k = 0.01$ to 0.002 h^{-1} , suggesting that glass encapsulation is preferable for moderate climates whereas plastic encapsulation may suffice for colder or buffered environments.

Electrode structure and interface engineering also significantly influence stability. Zhou et al. embedded AgNWs into a crosslinked polyvinyl alcohol matrix (AgNWs-em-cPVA), achieving a transparent electrode with excellent damp-heat stability that can withstand up to 200°C and exhibits only 1.12% moisture adsorption over 168 h at 60% relative humidity and 25°C [95]. Large-area flexible OPV modules (52.3 cm^2) based on this electrode retained 92.76% of their initial efficiency after 1008 h under AM 1.5 illumination with UV filtering. Cheng et al. developed AgNW/carbon nanotube composite transparent electrodes, and the resulting flexible devices retained 90.64% of their initial PCE after 2000 bending cycles at a radius of 5 mm [96]. Zhou et al. addressed the thermal stability challenge of ultra-thin silver transparent electrodes ($<10 \text{ nm}$) and developed high-thermal-stability ultra-thin Ag electrodes for large-area flexible OPV modules [97]. After over one month of outdoor tracking tests, the modules demonstrated operational stability comparable to silicon reference devices. Ma et al. employed an amorphous ITO-modified printed metal grid electrode, and the flexible OSCs retained excellent long-term stability under damp-heat conditions (85°C , 85% RH) for 200 h [98].

These systematic comparisons demonstrate that substrate selection, electrode material, encapsulation strategy, and interface engineering collectively determine the operational and environmental stability of flexible OSCs. Among the various pairings, PI/AgNW or PI/graphene combinations offer superior thermal and mechanical stability; ultra-thin glass paired with PEDOT:PSS avoids acid-induced substrate corrosion; and AgNW electrodes with appropriate embedding or composite strategies significantly outperform PEDOT:PSS in long-term operational tests. These findings provide clear guidance for selecting substrate-electrode combinations tailored to specific stability requirements in practical applications.

5. Conclusions and Future Perspectives

5.1. Optimal Substrate-Electrode Pairings

The preceding analysis underscores a central tenet of synergistic design: there is no singular “best” substrate or electrode in isolation. Instead, the optimal combination is dictated by the specific performance priorities and application scenarios of the target device. By examining the successful integrations reported in the literature, several prototypical pairings emerge, each offering a distinct balance of properties tailored to different functional demands (the optimal substrate-electrode pairings are benchmarked in Table 5).

Table 5. Benchmarking of optimal substrate–electrode pairings for flexible electronics.

Optimal Pairings	High-Performance Solution	Stretchable Solution	Eco-friendly Solution	Low-Cost Printing Solution
Substrate Type	Ultra-thin Glass/PI	TPU/PDMS	Nano-cellulose/Silk Protein	PET
Electrode Type	Embedded AgNWs/Metal Mesh	AgNWs/PH1000 Composite/PEDOT:PSS	Embedded AgNWs + PEDOT Overcoat	Coated AgNWs/Modified PEDOT:PSS
Transparency	>90%	>85%	>80%	>85%
Conductivity	Sheet resistance <10 Ω sq ^{−1}	Sheet resistance 10–30 Ω sq ^{−1}	Sheet resistance 20–50 Ω sq ^{−1}	Sheet resistance 15–40 Ω sq ^{−1}
Flexibility	Bendable, non-stretchable	Stretchable >20%	Bendable, partially stretchable	Bendable, non-stretchable
Thermal Stability	>300 °C	<120 °C	<150 °C	<100 °C
Solvent Resistance	excellent	good	Requires crosslinking	Moderate
Interfacial Adhesion	Excellent (Enhanced by embedding/seed layer)	Good (Surface treatment needed)	Moderate (Hydrophilic treatment needed)	Good (Enhanced by UV/O ₃ treatment)
Process Compatibility	Photolithography/transfer, suitable for R2R	Solution coating/printing	Solution coating, low-temperature drying required	Suitable for R2R, spray coating, Meyer rod coating
Environmental Stability	Excellent (Moisture/heat and oxidation resistant)	Good (Encapsulation needed for moisture resistance)	Moderate (Waterproofing treatment needed)	Moderate (PEDOT needs moisture protection)
Cost	High	Medium	Low (Bio-based materials)	Low
Recommended Applications	High-efficiency flexible modules, high-temperature process devices	Wearables, curved surface lamination, stretchable electronics	Eco-friendly disposable electronics, biocompatible devices	Large-area flexible photovoltaics, low-cost printed electronics
Considerations/Notes	Avoid sharp bending, prevent brittleness/cracking	Avoid high-temperature processes, enhance interface	Enhance water resistance via crosslinking, avoid polar solvents	Avoid high-temperature annealing, control surface roughness

For applications prioritizing record-high efficiency and operational stability, where the goal is to close the performance gap with rigid devices, ultrathin glass combined with metal grids or AgNWs represents the most compelling choice. UTG provides an ideal platform for high-performance electrodes due to its exceptional transmittance (>90%), thermal stability (>600 °C), and complete solvent resistance. This robust foundation allows for the implementation of demanding electrode fabrication processes. Metal grids, often fabricated via photolithography, can achieve ultra-low sheet resistance on the dimensionally stable UTG surface without the risk of substrate deformation [77,78]. Alternatively, UTG's high-temperature tolerance permits aggressive annealing or chemical sintering of AgNW networks to minimize junction resistance, while its flawless smoothness mitigates the risk of nanowires shunting the thin active layer [22]. This pairing is best suited for high-performance portable power sources and modules where efficiency cannot be compromised.

When the ultimate goal is extreme flexibility and ultrathin form factors (e.g., total device thickness < 10 μ m), the combination of polyimide with AgNWs or graphene emerges

as the superior strategy. PI offers unmatched thermal stability ($>400\text{ }^{\circ}\text{C}$) and mechanical strength among polymers, providing a resilient foundation at sub-micron thicknesses. The inherently flexible nature of AgNWs and graphene overcomes the brittleness of traditional oxides like ITO. The “semi-embedding” strategy, where AgNWs are partially anchored into the PI surface, is particularly effective [31]. It harnesses the high conductivity of the nanowire network while the PI matrix provides mechanical protection and mitigates surface roughness. For graphene, PI serves a dual role as both a flexible substrate and a high-temperature stable carrier, enabling the direct integration of CVD-grown graphene for an ultra-clean, high-performance interface [67]. This pairing is the optimal choice for wearable electronics, implantable devices, and other “skin-like” applications where weight and bendability are paramount.

For devices demanding intrinsic stretchability and conformability to curved or moving surfaces, elastomeric substrates like PDMS and TPU must be paired with embedded AgNWs or highly ductile PH1000. The key challenge here is maintaining percolation pathways under tensile strain. A purely surface-coated electrode would delaminate or fracture. Therefore, an “embedded” or “composite” architecture is essential. Embedding AgNWs into the surface layer of PDMS or TPU creates a mechanically interlocked structure where the elastomer matrix absorbs the strain while the nanowire network deforms and slides to maintain conductivity [42,43]. Similarly, intrinsically conductive polymers like PH1000 can be optimized for stretchability on these substrates through surface treatments or by forming hybrid composites [35,43]. This combination is uniquely suited for soft robotics, artificial skin, and bio-integrated electronics.

Where the primary drivers are low cost and scalability for large-area manufacturing, the workhorse combination of PET/PEN substrates with solution-processed AgNWs or PEDOT:PSS remains the most viable. Both substrates are commercially mature, offer good transparency, and are compatible with high-volume R2R processing. AgNWs can be deposited via slot-die or Meyer rod coating directly onto PET, and their performance can be enhanced through chemical welding or the use of polyelectrolyte additives to reduce junction resistance [24,83]. PEDOT:PSS, particularly the high-conductivity PH1000 grade, offers an all-solution, vacuum-free electrode route when combined with acid treatments or interfacial adhesion layers to boost conductivity and bonding on PET [27,63,64]. This pairing is the frontrunner for building-integrated photovoltaics, consumer electronics accessories, and other cost-sensitive large-area applications.

Finally, for applications guided by the principles of a circular economy and environmental sustainability, the pairing of cellulose-based substrates with AgNWs is gaining significant traction. The key challenge for cellulose—its surface roughness—has been effectively addressed through the use of cellulose nanocrystals, which can achieve a smoothness comparable to PET [51]. Furthermore, functionalized celluloses like TEMPO-oxidized cellulose nanofibers offer abundant surface chemistry that can interact favorably with AgNWs, producing smooth, highly conductive composites with the added benefits of a low CTE and biodegradability [57]. While still in development, this pairing holds immense promise for eco-friendly disposable electronics and implantable devices where material degradation is a feature, not a failure.

Translating laboratory-scale FOSC demonstrations into commercially viable products requires careful consideration of manufacturing scalability, production costs, and technology readiness [99]. Roll-to-roll processing is widely regarded as the most promising route for high-throughput and low-cost manufacturing of flexible OPVs, enabling the rapid fabrication of large-area thin-film modules on flexible substrates. However, transferring high-performance lab-scale devices to industrially relevant module sizes remains a major challenge, and effective strategies to bridge this gap are still lacking [100]. From a cost

perspective, detailed manufacturing analyses based on state-of-the-art printing and coating processes for fully encapsulated, ITO-free flexible OPV modules have shown that material costs can drop below €10 per square meter in optimized production scenarios, positioning OPV as a competitive alternative to established power generation technologies. It is widely recognized that a cost target of under \$0.20 per watt is essential to compete with incumbent technologies, requiring continued innovations in both materials and manufacturing processes. Replacing cost-intensive layers, particularly ITO which relies on the rare metal indium, along with process optimization, represents a key pathway to achieving these targets [101]. In this context, solution-processable alternatives such as AgNWs and PEDOT:PSS offer distinct economic advantages over vacuum-deposited ITO, as they eliminate high-energy deposition steps and are compatible with high-speed R2R coating. The energy payback time (EPBT) is another critical metric for assessing the environmental and economic viability of FOSCs. Replacing ITO with alternative transparent conductors has been shown to significantly reduce both the energy payback time and the overall cost of OPV modules. For ITO-free R2R-fabricated modules, the EPBT is strongly influenced by module efficiency and the energy intensity of manufacturing processes such as sputtering. With continued efficiency improvements and the adoption of low-energy printing techniques, FOSCs are expected to achieve substantially shorter EPBTs, further enhancing their competitiveness [102].

In conclusion, the selection of an appropriate substrate–electrode pair is a decision that must be guided by the ultimate application. The most successful FOSCs are not merely assemblies of high-performance parts but rather integrated systems where the substrate and electrode are harmonized across multiple dimensions, including mechanical, thermal, optical, and chemical compatibility, to collectively meet the demands of their intended use.

5.2. From Compatibility to Functional Integration

The preceding discussion on the optimal pairing of substrates and electrodes fundamentally remains grounded in the core paradigm of current FOSC research, namely achieving compatibility between these two components through material screening and interface engineering, under the premise of clearly distinguishing their respective functions. However, as device performance continues to improve and application scenarios expand, this traditional paradigm is facing challenges. A more forward-looking perspective is that the functional boundary between substrates and electrodes is gradually blurring, with both evolving toward an integrated, multifunctional direction that can be described as functional integration.

This trend is evidenced by several emerging strategies. First, substrates are being engineered to take on active optical roles, as seen in moth-eye nanopatterned PDMS films that function as built-in anti-reflection layers [35]. Second, substrates are being used to enhance electrode performance through structural integration, such as the semi-embedding of AgNWs into polymer matrices to improve mechanical robustness and surface smoothness [31,42]. Third, electrodes themselves are assuming multiple functions, with examples like ZnO/AgNW hybrids serving simultaneously as transparent conductors, electron transport layers, and light-scattering media [22]. Finally, the direct integration of materials like graphene onto PI substrates points toward a future where the distinction between substrate and electrode disappears entirely during fabrication [70].

From a design methodology perspective, the concept of functional integration necessitates the adoption of a holistic optimization strategy. The traditional “bottom-up” approach to material screening and process optimization will be complemented by a “top-down”, target-oriented design method: starting from the desired final device performance, the required multi-dimensional performance parameters (mechanical, optical, electrical, and

thermal) are derived. These parameters, in turn, guide the synthesis and construction of an integrated material system with graded functionality. This shift in design philosophy holds the potential to fundamentally resolve the issue of interfacial failure inherent in traditional layered structures, simplify device fabrication processes, and open new pathways toward realizing truly ultra-thin, ultra-flexible, and highly stable flexible optoelectronic devices.

5.3. Conclusions

In summary, this review has shown that achieving compatibility between flexible substrates and transparent electrodes through deliberate co-design across interface, mechanical, optical, and processing dimensions is a practical approach for improving the performance of flexible organic solar cells. The most effective device demonstrations to date are not simply combinations of high-performance individual materials but rather integrated systems in which the substrate and electrode are matched across multiple physical and chemical parameters to meet the requirements of a given application. At the same time, several limitations remain. Interfacial adhesion under repeated mechanical stress, long-term operational stability under ambient conditions, and the inherent trade-offs among transparency, conductivity, and scalable fabrication continue to constrain the broader applicability of these devices. A more systematic understanding of failure mechanisms at the substrate–electrode interface is still needed.

Looking forward, a shift from compatibility toward functional integration may offer a way to address some of these limitations. In this paradigm, the substrate is no longer a passive support but can contribute to optical management, mechanical resilience, or charge transport, while the electrode becomes embedded rather than simply deposited. Such integrated architectures have the potential to reduce interfacial failures, simplify processing, and enable thinner and more mechanically robust devices. Whether this approach can be realized in practice will depend on continued progress in materials design, processing control, and device characterization. The long-term goal is not to replace existing strategies entirely but to expand the design space available for building flexible photovoltaics that are both efficient and fit for practical use.

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