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REVIEW

PROGRESS IN PHOTOVOLTAICS WILEY

Progress, challenges, and perspectives on polymer substrates for emerging flexible solar cells: A holistic panoramic review

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Abstract

In pursuit of a renewable, inexpensive, sustainable, and compact energy source to replace fossil fuels, solar photovoltaic devices have become an ideal alternative to meet human needs for environmentally friendly, affordable, and portable power sources. It is due to their excellent mechanical robustness and outstanding energy conversion efficiency. Concerning the increasing demand for flexible and wearable electronic devices with standalone power sources, much attention has been paid to photovoltaics' flexibility and lightweight developments. Along with high mechanical flexibility and lightweight, flexible photovoltaic devices have the advantages of conformability, bendability, wearability, moldability, and roll-to-roll processing into complex shapes that can produce niche products. Emerging solar cells, among other photovoltaic technologies, have been exalted for their high conversion efficiency, low cost, and ease of production, making them a viable new-generation photovoltaic technology. The main commercialization choice for cutting-edge solar cells is flexible dye-sensitized and perovskite solar cells since they can be made using a roll-to-roll printing technique and are appropriate for mass manufacturing. More significantly, flexible evolving solar cells may be created on ultrathin and light substrates to fulfill the demands of the developing flexible electronics industry and discover uses that are not possible with traditional photovoltaic technology. In any flexible device, the substrate is a backbone on which further materials rely. A flexible substrate reduces the installation and transportation charges, thereby reducing the system price and increasing power conversion efficiency. In this review, we comprehensively assess relevant materials suitable for making flexible photovoltaic devices. Several flexible substrate materials, including ultra-thin glass, metal foils, and various types of polymer materials, have been considered. For conducting materials, transparent conducting oxides, metal nanowires/grids, carbon nanomaterials, and conducting polymers have also been comprehended. Progress on various flexible foils, fabrication and stability issues, current challenges, and solutions to those challenges of using conductive polymer substrate is endorsed and reviewed in detail. The originality of this holistic study lies in its ability to offer a thorough overview of recent advancements in flexible dye-sensitized and perovskite solar cells on polymer substrates, which is conceivable and worthy as a roadmap for future research work.

KEYWORDS
flexible dye-sensitized solar cell, flexible perovskite solar cell, flexible solar cell, metal substrates, polymer substrates, transparent conducting layers, ultra-thin glass substrates

1 | INTRODUCTION

The role of electricity in developing human civilization to power electrical appliances such as heating and cooling devices, lighting systems, domestic, and industrial appliances is indisputable.[1] The world's population is rapidly increasing along with the amelioration of socioeconomic conditions, increasing electricity consumption per capita. Due to the ever-increasing global energy demand, fossil fuels are mined and burned to produce electricity, causing severe environmental problems such as greenhouse gas emissions, air pollution, and water pollution that ultimately affect the ecosystem and human health.[2] Nonetheless, humans must accept that fossil fuels are finite and will soon run out.[3] In times of crisis, energy and environmental issues will become the most critical future issues. Creating clean, renewable, and sustainable energy sources is an essential strategy for the sustainable civilization of our society.[4] Renewable energy sources such as hydropower, biomass, geothermal, wind, and solar have tremendous potential to replace traditional fossil fuels in the future eventually.[5] Among various renewable energy sources, sunlight has long been recognized as the most promising sustainable source of inexhaustible, clean energy capable of meeting the ever-increasing demand for greener and more sustainable energy sources in the human environment.[6] The solar energy reaching the earth's surface is 3.8 million exajoules per year (1 EJ $= 10^{18}$ J), enough to meet the world's annual energy demand in a short period.[7] Given the abundant solar resources on Earth, solar energy provides a compelling reason to replace fossil fuels and can potentially be a widely available biodegradable energy source.[8] However, the main requirements are directed toward technological progress in terms of cost reduction, environmental friendliness, and flexibility. As known, solar energy from the sun is the cleanest, most reliable, and cheapest source of energy that can be converted into any other desired form of energy.[9,10] Various technologies are being developed using solar energy, such as solar heaters, thermal architecture, solar power generation (photovoltaics [PVs]), artificial photosynthesis, and photocatalytic water splitting.[11] Among these technologies, PV systems remain at the forefront of renewable power alternatives that, unlike burning fossil fuels, do not emit carbon dioxide. PV systems operate on the principle of the PV effect. The PV effect was discovered in 1839 by Edmond Becquerel, a French scientist, by observing the light-dependent voltage between electrodes immersed in an electrolyte.[12] By the PV effect, PV systems use the electronic properties of certain semiconductors to convert sunlight energy directly into electrical energy.[13] Therefore, PV technology is the best way to produce power from the natural source (sunlight).

The development of PV systems began in the 18th century and continues to the present. There have been several stages of development, starting with the observation of the PV effect, followed by the development of materials exhibiting the PV effect, the development of apparatus for solar energy conversion, and lastly, the implementation of such units on a large scale. So far, numerous types of solar cells have been developed by successive generations. The first-generation solar cells are based on crystalline silicon (Si), including single-crystalline silicon solar cells and polycrystalline silicon solar cells.[14,15] With a high efficiency of about 27% and a long lifetime, these solar cells dominate the PV market.[16] However, these solar cells have certain drawbacks, including an expensive, convoluted, energy-intensive manufacturing process, poor esthetics, and high environmental impact. Further, these solar cells perform poorly in low-light circumstances, which limits their prominence in the terrestrial PV market.[17] The solar cells based on amorphous silicon, thin films, and semiconductors derived from group III and V elements of the periodic table constitute the second-generation solar cells. The best-suited active materials of second-generation solar cells are copper zinc tin sulfide (CZTS), copper indium gallium diselenide (CIGS), cadmium telluride (CdTe), and gallium arsenide (GaAs).[18] The thin film technology has helped to reduce the costs compared to crystalline silicon-based technology because of lower material requirements, can be manufactured in large sizes, and can be mounted on curved surfaces using suitable substrates. In contrast, amorphous silicon and III-V materials have significantly shown improved efficiencies.[19] However, the second-generation technology has some downsides, such as the lower efficiency of thin film solar cells compared to wafer-based cells and elements such as indium that are becoming scarce and expensive. At the same time, cadmium is highly poisonous, causing production challenges. Also, manufacturing III-V-based solar cells is costly.[20] As a result, third-generation solar cells also referred to as emerging PV have been created as a substitute for Si-based and thin-film solar cells. These emerging solar cells tend to obtain high-efficiency devices at a lower cost and use nontoxic and abundant ingredients.[21] Examples of third-generation PVs include dye-sensitized solar cells (DSSCs), quantum dot-sensitized solar cells (QDSSCs), organic solar cells (OSCs), and perovskite solar cells (PSCs).[22–25]

The rapid development of electronic technology has significantly increased the demand for portable, functional, and wearable electronic devices. In this situation, converting solar cells into a flexible form can effectively and enormously be used in various applications as an integrated power device. Figure 1 shows applications that use flexible solar cells as integrated power devices. Therefore, in recent years, high-performance flexible solar cells have sparked strong research interest in innovative materials and device designs with flexible and stretchable properties for PV technologies.[27] Emerging solar cells with additional capabilities, ease of processing, and minimal manufacturing costs can be converted into efficient, flexible solar cells by replacing the substrate, electrode, and other solar cell components with more flexible and cost-effective materials (see Figure 2). Flexible

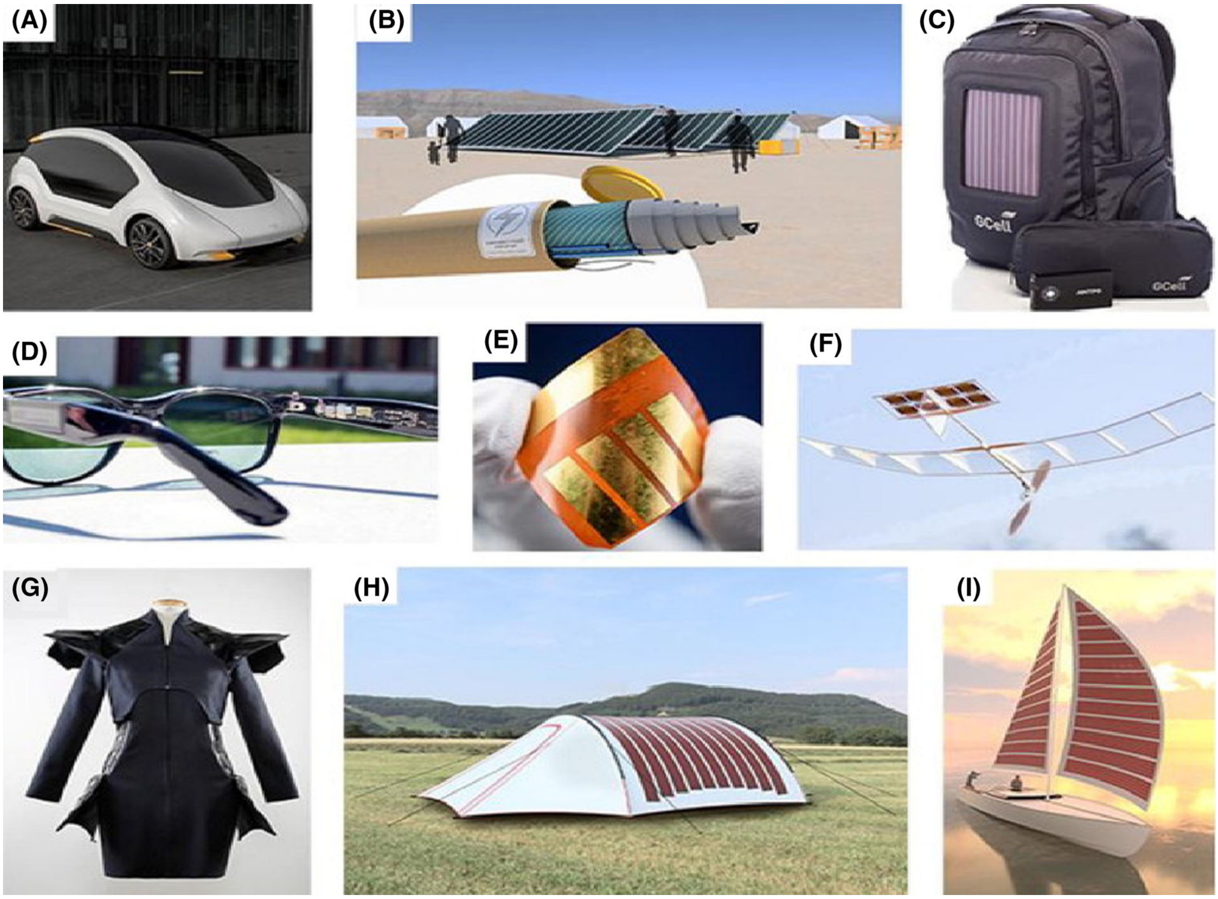

FIGURE 1 Integrated applications of flexible solar cells. (A) A battery-powered electric vehicle that uses flexible solar panels, (B) flexible cells in tubes waiting for further deployment, (C) a solar-powered bag, (D) electric sunglasses with solar cells for lenses, (E) a photograph of a typical lab-made flexible solar cell, (F) an uncrewed aerial vehicle with flexible solar cells, (G) solar wearable clothing, (H) a tent embedded with flexible solar cells, and (I) a solar-powered boat. Reproduced with permission from Zhang et al.[26] Copyright 2020, Elsevier.

solar cell technologies necessitate highly functional materials, appropriate equipment, and compatible methods. Therefore, researchers worldwide have been exploring new materials that match these requirements. The bendability, stretchability, foldability, compatibility with curved surfaces, and reduction in the overall weight of the cell distinguish flexible PV devices from rigid PV devices.[26] In addition, flexible solar cells can further reduce production costs by using a low-temperature energy-saving method and can be manufactured on flexible substrates using continuous roll-to-roll (R2R) fabrication technology.[28] These characteristics make flexible PVs more cost-effective in terms of transportation and installation. Their benefits are considerably more impressive when used in aircraft, stratosphere, and space applications.[29] As a result, research into developing high-efficiency flexible emerging solar cells is essential.

This review presents a complete assessment of flexible substrate materials suited for fabricating emerging solar cells. The synergistic effects of various substrate materials (ultrathin glass, metal foil, polymers) are investigated by elucidating their distinct roles in enhancing the property and performance of the solar cell. In further sections, various polymer substrates that can be used in PV applications are reported with their advantages and solutions to their limitations. As an ideal substrate, polymers exhibit suitable insulating properties. The substrates are made conductive when used in electronic devices by applying a transparent conductive layer. Such layers are thoroughly investigated here. With the efficient properties of conductive polymer-based substrates, a literature investigation on existing dye-sensitized and PSCs is summarized in subsequent sections.

2 | FLEXIBLE SUBSTRATES

A good substrate plays a pivotal role in defining PV device properties. Solar cells were first researched and developed on rigid glass substrates because they are resistant to heat, optically transparent, and establish good interaction with transparent conductive films. In contrast, the flexible PV cells on lightweight substrates represent new scientific and technological challenges for subsequent generation applications. The flexible substrate is the most vital component of any flexible device. It is because the deposition of successive materials on it is highly dependent on the mechanical and chemical properties of

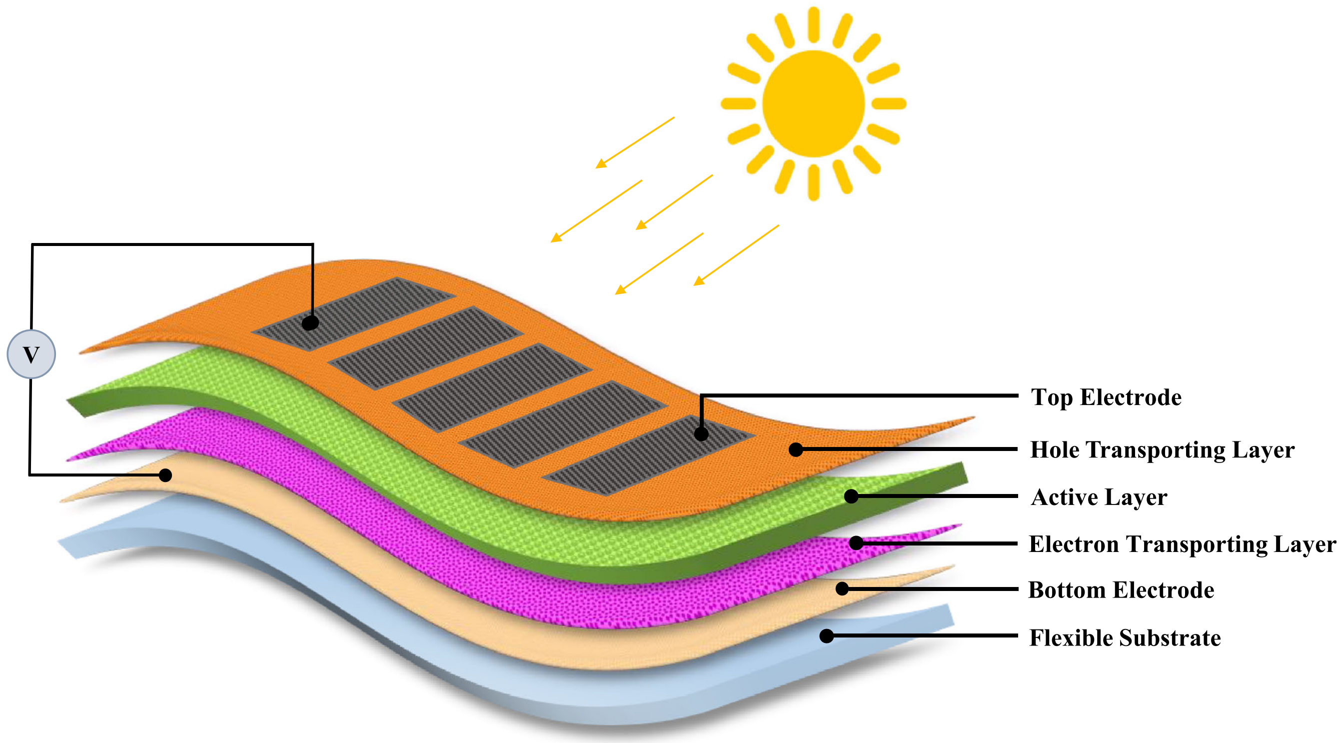

FIGURE 2 Schematic representation of flexible solar cell.

the substrate.[30,31] Appropriate substrate materials are required for flexible solar cell devices with stable and excellent performance.

The following requirements must be met to have an ideal flexible substrate for a PV cell:

(1) High flexibility: First and foremost, the substrate material should be flexible. This means the flexural rigidity for a given configuration and flexing axis should be as low as possible. This boils down to having a low Young's modulus for a substrate of given dimensions or the ability to create ultrathin versions of the substrate, given a material.[28] *(2) High optical transmittance*: As solar cells are light-absorbing devices, consideration should be given to the substrate's optical characteristics as it determines the design/structures of solar cells. In optical properties, reflectance and transmittance are two quantitative parameters. Light passes through the substrate in transmittance, while in reflectance, the substrate tends to reflect the incoming photon. For a flexible PV application, an ideal substrate should have either high optical transmittance (particularly more than 90% of the visible light spectrum) or high reflectance.[26] *(3) High elasticity*: The substrate material should have a high elastic strain limit to account for large strains in the top and bottom layers associated with the bending motion. This will ensure an adequate minimum bending radius for various applications. Upon applying stress to the substrate, an internal force known as restoring force generated inside the substrate should be high enough to allow the substrate to retain its original shape when stress is removed.[24] Thus, the substrate material should be elastic to the minimum bending radius required. *(4) High thermal tolerance*: Various materials are deposited on substrates in any flexible device. Some bonding materials require high temperatures. Therefore, high thermal stability is a characteristic property of a substrate to produce high-efficiency devices. *(5) Robust mechanical durability*: When various layers are deposited on substrates, it experiences extreme stress and strain, which can change their functions. Whereas an ideal flexible substrate should be able to deformably transform and effectively release tension without affecting its original functionalities.[32] Therefore, a substrate should have good dimensional stability to maintain its properties. *(6) Low coefficient of thermal expansion (CTE)*: The thermal characteristics of the substrates are primarily determined by two parameters, that is, *CTE* and the thermal transition (T). To obtain an effective solar cell, the ideal temperature of the substrate material, often referred to as the glass transition temperature (T_g), must be compatible with the maximum temperature (T_{max}) for the fabrication process of subsequent layers. When the temperature changes, the substrates will either expand or contract. The lattice would experience additional stress because of a significant mismatch in thermal characteristics between the adjacent layers, which would cause strain and crack during the fabrication process. To avoid the build-up of stress because of temperature cycling over its lifetime, it is ideal to have a substrate with a low CTE or with CTE matching that of other materials in the integration.[33] *(7) High resistance to chemical solvents*: During the fabrication process of solar cells, substrates are exposed to many different substances, including gas and solvents. Hence, the substrate should be stable and inert against chemicals, and the substrate should not release contaminants.[19] *(8) Low surface roughness*: Since a solar device is a stack of

thinner films, the electrical function is more sensitive to surface roughness. The substrate's rough surface precluded conformal layer coating and preventing electrical short circuits in the device. To attain an effective solar cell, the surface roughness should be low. Different planarization processes like polishing and coating will increase the processing cost when the roughness is very large. *(9) Good oxygen and water barrier properties*: The majority of electrical equipment are sensitive to oxygen or moisture, which significantly reduces performance. The substrate should function as a barrier layer to prevent the passage of oxygen and water vapor to provide a long-term steady performance.[34] *(10) Higher conductivity*: The substrate should be insulated to ensure that different parts of the integrated system are electrically isolated.[33] However, depending upon the application, there may be some merit in having an electrically conductive substrate that can provide an easily accessible common terminal to interconnect various parts of the circuit or shield the circuit from electromagnetic noise. In solar cell devices, conduction plays an important role in improving device performance. Typically, TCO, a charge-collecting layer, is placed on a substrate to serve as a transparent conducting electrode (TCE). The PV performance, particularly the fill factor and photocurrent density of the solar cell, is highly correlated with the surface resistance of TCE.[28]

In addition to these properties, an ideal flexible substrate should be easy and inexpensive to manufacture, allowing the device to be scaled. For example, substrates can be manufactured in bulk using large chemical baths or through the R2R process.[35] The substrates, particularly in the case of wearable electronics, should be non-toxic and biocompatible. From a usability point of view, ideal substrates should be lightweight and easy to handle independently.[24] These properties have been summarized in Figure 3. These qualities define a perfect substrate and a superstrate. However, it is impossible to obtain a material with all of these ideal properties because many of these are conflicting; that is, having one desirable property implies the loss of another from a fundamental physics perspective. For example, it is stated that an ideal substrate should be flexible, which requires Young's modulus of a material to be low, given that the dimensions and axis of flexure are fixed. However, low Young's modulus leads to a low spring constant for a substrate of given dimensions, leading to a low restoring force. Thus, there needs to be a balance between the requirement for high restoring force and low flexural rigidity.[36] Similarly, high thermal conductivity and electrical insolation results in a very narrow set of materials. Highly crystalline materials such as diamonds demonstrate high thermal conductivity due to effective phonon transport through the lattice while being electrically insulating because of the lack of free carrier concentration at room temperature owing to the tight binding of electrons in the lattice. However, such materials are neither flexible because the tightly bound lattice leads to a high Young's modulus nor easy to fabricate. After all, highly crystalline materials require sophisticated fabrication techniques such as the epitaxial process.[28] Hence, the search for ideal substrates for flexible PV applications leads to compromises based on the application at hand, functional materials being used, the conditions of deployment, and so on. Indeed, it is interesting to note that most of the properties described here are fulfilled by the most commonly used

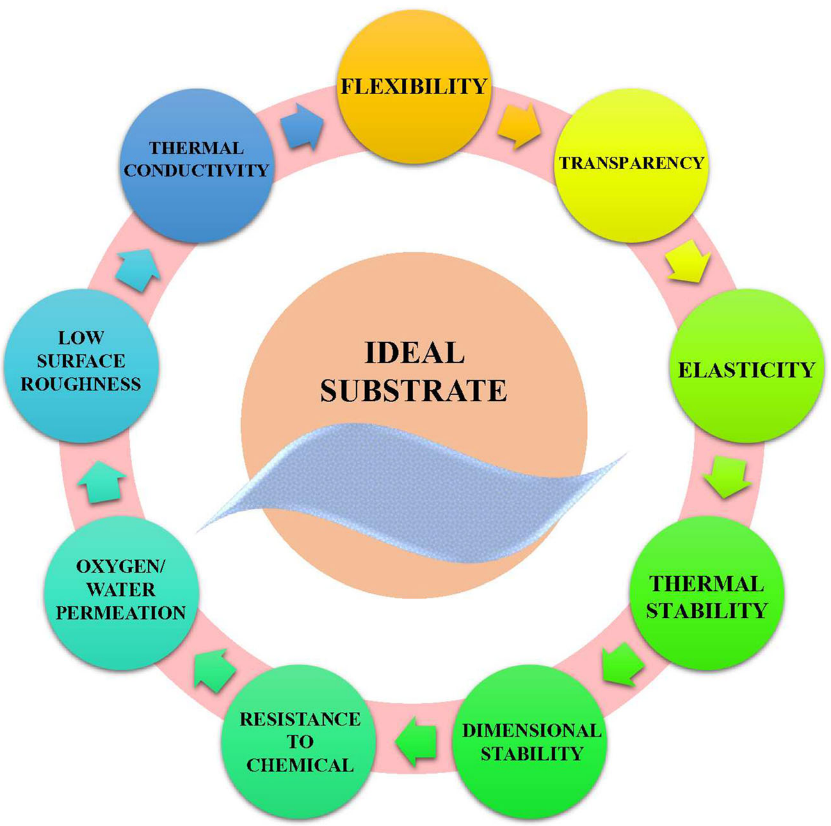

FIGURE 3 Properties associated with an ideal substrate material for flexible photovoltaics.

substrate in the electronic industry—the single crystalline silicon substrate. Silicon is mechanically, thermally, and chemically stable; has low CTE and high thermal conductivity; has low electrical conductivity (in the intrinsic state); is easy to manufacture and handle; and can have an atomically smooth surface finish that can withstand high temperatures. However, flexibility does not qualify as the first and foremost property required from a flexible electronic substrate. Although reducing the thickness of silicon can be made flexible, it leads to other problems related to ease of handling. Thus, to date, flexible PV devices primarily use three types of substrates—ultrathin glass, metal foils, and polymer/plastics.[27] These substrates meet all the requirements of an ideal substrate with ease of fabrication and cost-effectiveness. Additionally, the following sections describe each substrate material's advantages, disadvantages, and prospects.

2.1 | Flexible glass substrates

The glass substrate is the most commonly used in the field of solar cells. It is a type of light-trapping ceramic material. The use of glass substrates is attributed to their beneficial properties. In a PV device, glass substrates can be used as front and back layers. The front glass layer allows incident light to travel before reaching the solar cell.[37] With today's glass-making technology, the glass substrate can be highly flexible by reducing the thickness below 100 μm (as shown in Figure 4A). This type of flexible substrate could meet most of the requirements for an ideal solar cell. The ultrathin glass substrates have high optical transmittance (over 90%), enabling more photons to reach the absorber. Glass is already a popular substrate choice for the fabrication of PVs because, apart from being transparent, ultrathin glass is also resistant to almost all chemical solvents, has low CTE of $\sim 10^{-6}/^{\circ}C$, and can withstand pretty high temperatures (>600°C). This makes it easy to transition these integration strategies onto a flexible glass substrate.[40] Ultrathin glass substrates can also be made transparent to selected wavelength ranges by changing their chemical composition and processing method (changing the “color” of the glass). Glass sheets also have very low water/oxygen permeability that is an essential property for PV cell since it is oxygen or moisture sensitive.[28] Flexible glass substrates can mass-produce PV devices using R2R technology (see Figure 4B,C).[35] Further, glass substrates/sheets are easy to produce because of the existence of a highly scaled infrastructure from the extraction of ore to purification and processing. Glass sheets can also be very low-cost because the two base elements used in glass, silicon, and oxygen are the two most

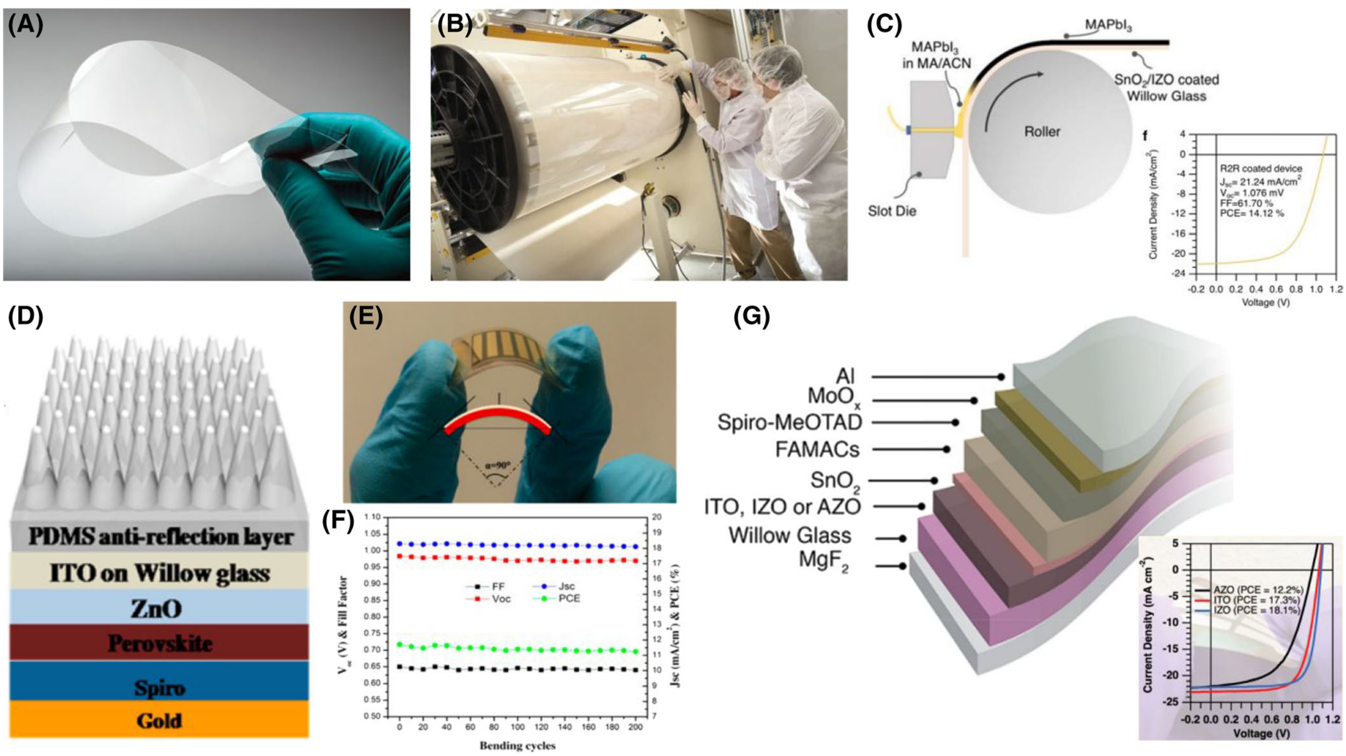

FIGURE 4 (A) Flexible and bendable ultra-thin glass sheet (photo courtesy of corning.com). (B) Flexible willow glass wound on a spool, compatible with R2R device manufacturing (photo courtesy of corning.com). (C) Schematic representation of slot-die coating on flexible glass with R2R coater. Reproduced with permission from Dou et al.[35] Copyright 2018, American Chemical Society. (D) Schematic structure of a solar cell based on a flexible glass substrate with a PDMS anti-refection layer. (E) Bending ability of flexible willow glass-based perovskite solar cell. (F) Efficiency stability depending on the bending cycle in perovskite solar cells based on a willow glass substrate. (D–F) Reproduced with permission from Tavakoli et al.[38] Copyright 2015, American Chemical Society. (G) Device architecture of flexible perovskite solar cells with current density—voltage (J—V) characteristics (reverse scans) under 100 mW/cm^2 AM1.5G. Reproduced with permission from Dou et al.[39] Copyright 2017, American Chemical Society.

TABLE 1 The photovoltaic performance of various flexible solar cells based on ultra-thin glass substrates.

Substrate	Device structure	PCE (%)	Special characteristic	Reference
Ultra-thin flexible glass (Willow glass)	Flexible glass/TiO_2/Ag/AZO/ZnO/PTB7: $PC_{71}BM$/MoO_3/Ag	6.6%	ITO-free polymer solar cell.	Formica et al.[43]
	Flexible glass/ITO/ZnO/$CH_3NH_3PbI_3$/Spiro-OMeTAD/Au	12.1%	On further applying a PDMS antireflection layer, the efficiency increased up to 13.1%.	Tavakoli et al.[38]
	Flexible glass/modified-PEDOT:PSS/ $CH_3NH_3PbI_{3-x}Cl_x$/PCBM/Ag	4.3%	ITO-free solar cell.	IEEE Staff[44]
	Flexible glass/AZO/Au/AZO/c-TiO2/mp-TiO2/ $CH_3NH_3PbI_3$/Spiro-OMeTAD/Au	9.6%	The utilization of AZO/Au/AZO multilayer electrodes instead of FTO.	Dang et al.[45]
	Flexible glass/IZO/SnO2/$Cs_{0.04}$ $MA_{0.16}$ $FA_{0.80}$ $Pb_{1.04}$ $I_{2.6}$ $Br_{0.48}$/Spiro-OMeTAD/MoOx/Al	18.1	-	Dou et al.[39]
	MgF_2/Willow glass/ITO/PTAA/$MAPbI_3$/C_{60}/ BCP/Cu	19.72	15.86% PCE on an area of 42.9 cm^2 with the blade-coating method	Dai et al.[46]
	Willow glass/ITO/TiO_2/N719: THF dye/Pt	4.53	-	Sheehan et al.[37]
	Flexible glass/PEDOT:PSS/organic material/ metal cathode	8.0	Higher power conversion efficiency than ITO or PEDOT:PSS on a plastic substrate.	Wang et al.[47]
	Willow glass/ITO/PTB7:PC70BM/ZnO/MoO_3/ Ag	6.3	-	Adiga et al.[48]
	Willow glass/ITO/PC70BM/$MAPbI_3$/PEDOT: PSS/Al	11.9	-	Adiga et al.[48]
	Willow glass/AgNW/ZnO/PTB7-Th: PC70BM/ MoO_3/Ag	7.3	Higher intensity of light was observed in combustion reacted ZnO/AgNW hybrid electrode.	Park et al.[49]
	Ultra-thin glass/Ti/TiO_x/mp-TiO_2/perovskite/ Spiro-OMeTAD/MoOx/IZO	13.61	Cost-effective and lightweight non-FTO device.	Chiang et al.[50]

abundant elements in the Earth's crust by weight. Further, glass substrates can have very smooth surfaces (RMS roughness around 1 nm), depending on the fabrication process.[37] The low surface roughness reduces the post-fabrication planarization step required for most metal substrates.

Corning Company has manufactured an ultrathin flexible glass called Willow® glass. Many researchers have used willow glass to fabricate flexible solar cells. By nature, willow glass is an alkali-free, alkaline earth boro-aluminosilicate glass and is highly transparent. Because the bending stress and stiffness of a flexible glass substrate directly depend on its thickness,[41] the small thickness of willow glass dramatically reduces the stiffness of the substrate. It reduces the bending stress to less than 100 MPa at a bending radius of 5 cm, along with an average surface roughness < 0.5 nm. Ultra-thin glass substrates tend to have higher surface energy than metal foil and polymer counterparts. Surface energy is the property of a surface that determines how well another material can deposit and adhere to it. High surface energy substrates are desirable for the material being deposited to "wet" the surface and eventually form a stable thin film.[33] A classic example of a low-energy surface is the surface of a lotus leaf that makes the water droplets bead up instead of allowing the water to form a thin layer on top. Possessing high surface energy will enable materials to create a thin film on the glass substrate. This is essential because most integration strategies for PV devices rely on deposition and adherence. Flexible glass is compatible with various existing and emerging device manufacturing methods, including R2R photolithography, printing, coating, laminating, vacuum deposition, patterning, etching, and transferring.[39] The abovementioned properties make flexible glass an ideal substrate for flexible PV applications. Also, compared to rigid commercial glasses, flexible glass substrates fabricated solar cell generates competitive efficiency. The fabrication of various solar cells on flexible glass substrates has been started since 2013.[42] In 2015, Tavakoli et al. first used willow glass (50 μm) as flexible substrates to prepare flexible PSCs and achieved an efficiency of up to 12.06%.[38] Similarly, Sheehan et al. first reported the flexible DSSC on a willow glass substrate giving a power conversion efficiency of 7.42%.[37] Figure 4D–G and Table 1 summarize various solar cells fabricated on ultra-thin flexible glass.

However, there are some limitations to the use of glass. The biggest problem with flexible glass substrates is their flexural rigidity. Although lowering the thickness of the sheet increases flexibility, the rigidity of a glass substrate is much higher than that of polymer sheets of the same thickness. This requires glass substrates to be substantially thinner than polymer sheets for the same flexibility, thus, reducing their ability to be handled independently, making its upscaling a challenge to achieve.[28] In addition to the flexible glass substrate, zirconia substrates have also been created for solar cells.[51] The zirconia substrates also exhibit properties such as high thermal stability (up to 1000°C), low thermal expansion, im-permeability to oxygen or moisture, and corrosive resistance. In 2015, Todorov et al. fabricated a copper zinc tin sulfide-selenide (CZTSSe) solar cell on a zirconia-based flexible substrate and reported a PCE of 11.5%.[52] Compared to the flexible glass substrate, zirconia substrates are heavy, expensive, and have a high surface roughness (~25 nm).

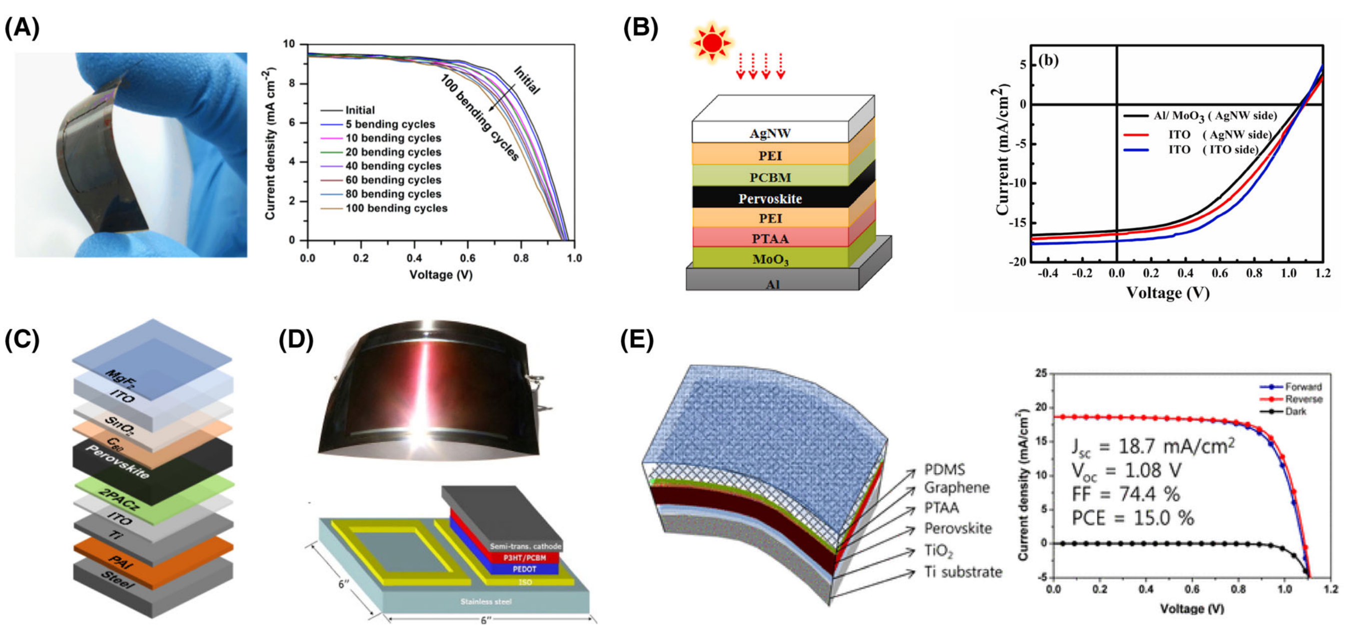

FIGURE 5 Metal foil substrate-based flexible solar cells. (A) Photograph of a solar cell fabricated on metal foil. Bending cycles of the flexible solar cell. Reproduced with permission from Wang et al.[57] Copyright 2014, Elsevier Ltd. (B) Schematic diagram of perovskite solar cells based on Al substrate and J-V curves. Reproduced with permission from Sun et al.[58] Copyright 2022, Elsevier B.V. (C) Device architecture of stainless steel-based flexible perovskite solar cell. Reproduced with permission from Feleki et al.[59] (D) Flexible organic solar cell fabricated upon stainless steel. Reproduced with permission from Galagan et al.[60] Copyright 2012, Elsevier. (E) Ti-based flexible solar cell owing efficient performance. Reproduced with permission from Heo et al.[61] Copyright 2018, American Chemical Society.

Further reducing the thickness and surface roughness can make zirconia substrate efficient for PV devices.

2.2 | Metal foil substrate

Metals have unexceptionable conductivities, high ductility, and malleability. Due to these excellent properties, metal sheets/foils less than 125 μm are supposed to be a promising flexible substrate for fabricating flexible solar cells.[28] A metal foil retains excellent mechanical properties, has high thermal conductance, low CTE, resistance to fire and process chemicals, and excellent properties against humidity and oxygen.[53] Most importantly, metal substrates can withstand high temperatures (up to 1000°C). Therefore, metal substrates can be sintered at high temperatures to remove the organic residues and promote particle interconnections, especially in DSSCs.[54] The metal foil can simultaneously function as a flexible substrate, bottom electrode, and transport layer, greatly simplifying the manufacturing process. But unlike glass substrates, metal substrates can only serve as back contacts due to their opacity (i.e., concerning the visible spectrum, it possesses significant optical reflectivity). To allow photons to enter the active materials, the top electrode of the solar cell must be optically transparent.[55] Several materials have been used as substrates to produce flexible solar cells. Each metal substrate has its advantages and shortcomings. The examples of metal foil that have received the most attention among metals are stainless steel (SS), aluminum alloy (Al), titanium (Ti), copper (Cu), and molybdenum (Mo).[56] Figure 5 shows flexible metal foils used as a substrate for flexible PV cells.

SS has been the most used in research because of its low weight, excellent corrosion resistance, and easy fabrication of substrates of various shapes.[62] An SS can withstand process temperatures of up to 1000°C, is dimensionally stable, and provides an ideal barrier to moisture and oxygen penetration. SS substrates are generally more durable than plastic and glass substrates.[28] Compared with other metal substrates, steel foils can also be produced in bulk through an R2R production process, significantly reducing their cost. In 2006, Kang and co-workers reported the first SS-based flexible DSSC, leading to a much higher overall solar conversion efficiency of 4.2%.[63] Further, the efficiency was improved by roughening the SS substrate and optimizing the counter electrode (CE).[64] There are few reports on PSCs on SS substrates, but all works show a p-i-n configuration (i.e., light is illuminated from the p-side). Recently, a PSC with this configuration was fabricated on a polymer-coated steel substrate and showed an energy conversion efficiency of 16.5%.[59] A PSC with an n-i-p structure (i.e., light is illuminated from the n-side) exhibited a power conversion efficiency of 5.03%.[65] Despite the ideal characteristics, a long-time stable solar cell is difficult to achieve with SS substrates.[62]

In recent years, other metals, such as titanium (Ti) foil, have also been used to make flexible solar cells. Although the sheet resistance of Ti is higher than that of general metals such as Ag and Cu, it shows low sheet resistance when compared with conductive glass. In addition, its conductivity is higher, and its cost is cheaper than conductive glass. After sintering at 500°C, the sheet resistance of conductive glass increased from 14.4 to 66.7 $\Omega\ sq^{-1}$. In contrast, the sheet resistance of Ti-foil just increased from 0.40×10^{-3} to $0.70 \times 10^{-3}\ \Omega\ sq^{-1}$, showing meager sheet resistance and high ability to withstand heat, as well as no need for pre-treatment to prevent surface oxidation.[54] Flexible Ti metal foil-based DSSCs were first demonstrated in 2006, resulting in a power conversion efficiency of 7.2%.[77] With a respectable power conversion efficiency of 8.31%, the first effort at a flexible PSC based on a Ti metal foil substrate was reported in 2015. Furthermore, titanium foil solar cells continue to function well even after 100 mechanical bend cycles, demonstrating their high flexibility.[57] With high efficiency, good flexibility, and simple manufacturing technology, flexible titanium foil solar cells open a promising future for power supplies for rooftop solar power generation and wearable devices. Table 2 represents flexible solar cells fabricated on various metal substrates. While metal foils have some advantages as flexible substrates, they have some significant drawbacks that make them less popular. One such problem is their surface roughness. The typical surface roughness of metal foils is in the order of a few microns. Thus, the defects on the surface can be significant compared to the stack being formed. This can lead to shorts in the device resulting in lower yield and reliability issues.[33] The process must be carried out with specialized rollers with high-quality surfaces to obtain a better surface finish. The rollers should be free of dust particles or other contaminants to get an ultra-smooth metal foil surface. These constraints increase the cost of production of the foils for applications in PVs, making their commercial use impractical. To reduce the cost of manufacturing, metal foils are sometimes post-processed to obtain a high-quality surface finish. For example, a cold-rolled steel foil can be subjected to chemical mechanical polishing (CMP) process to get a better surface finish before deposition or transfer of electronic materials. The CMP process for metal foils is typically carried out with silica or alumina slurry, leading to a post-process surface roughness of a few nanometers. Another commonly used process for increasing metal foil surface quality is spin-on passivation layers.[56] For example, a layer of spin-on-glass (SOG), a colloidal solution of silica particles, is spin-coated on a metal foil substrate and annealed to form a thin film. The thin film is significantly more planar than the underlying substrate, providing a smooth surface for fabricating electronic circuits. These layers also provide an insulating layer to separate one electronic device from another. If the substrate is used as a common terminal, the passivation layer can be patterned using lithography and etched to create metal vias. However, the substrate's flexibility is the trade-off with the use of passivation layers for planarization and electrical insulation. Thick passivation layers lead to better planarization and electrical insulation; however, they are susceptible to cracking when the flexible substrate is bent.[26] On the other hand, thin layers that can withstand bending strain do not offer good surface planarization. The absence of superior top transparent electrodes with greater conductivity and flexibility, which is a significant issue with employing metal foil substrates, is impeding the development of metal foil-based flexible solar cells.

TABLE 2 The photovoltaic performance of various flexible solar cells based on metal substrates.

Substrate	Device structure	PCE (%)	Special characteristic	Reference
Titanium (Ti)	Ti/TiO_2/perovskite/PTAA/graphene/PDMS	15%	-	Heo et al.[61]
	Flat titanium sheet with micro holes (FTS-MH)/TiO_2/dye-electrolyte	7.25%	FTS-MH has been utilized to fabricate TCO-less DSSCs in back contact device architecture, which offers several advantages in terms of simplicity and ease of fabrication.	Hayat et al.[66]
	Ti/TiO2 + CH3NH3PbI3/Spiro-MeOTAD/ITO	9.1%	Insertion of varying thicknesses of ultra-thin Ag films to the top electrode improves the efficiency to 11.01%.	Lee et al.[67]
	$Ti/TiO_2/Al_2O_3$/perovskite/PEDOT:PSS/Spiro-OMeTAD/TCO/PET+Ni mesh	10.3%	-	2015_Highly efficient, flexible, indium-free perovskite solar cells employing[68]
	Ti/TiO_2 flowers/N719 dye/PEDOT/ITO-PEN	6.26%	-	Lei et al.[69]
	Ti foil/AgNP-TiO_2/N719 dye-electrolyte/Pt/FTO glass	4.7%	Back-illuminated DSSC	Shamsudin et al.[70]
	Ti foil/TiO_2/N719 dye – electrolyte/Pt/ITO-PET	3.74% 4.98%	H2O2-treated Ti foil resulted in higher PCE	Rui et al.[71]
Copper (Cu)	$Cu/CuI/CH_3NH_3PbI_3/ZnO/Ag$	12.8%	The cheap and low-weight conductive substrate has a close work function to ITO.	Abdollahi et al.[72]
Aluminum (Al)	Al/MoO_3/PTAA/PEI/perovskite/PCBM/PEI/AgNW	7.09%	Low-cost perovskite solar cells are obtained using aluminum foil.	Sun et al.[58]
Stainless steel (SS)	SS/Si_3N_4/(Cr/Au/Cr)/PEDOT:PSS/$CH_3NH_3PbI_3$/PCBM/BCP/Ag	5.03%	n-i-p configuration	Kumar et al.[65]
	Steel/Ti/ITO/SnO_2/PCBA/perovskite/TCTA/MoO_3/ITO/MgF_2	15.2% 14.9% 14.1% 13.8%	The performance gradually decreased with increasing surface roughness (Rp ≈ 200, 500, 1500, 2500 nm) using planarization on a layer coated on steel substrates.	Feleki et al.[73]
	SS/Si_3N_4/(Cr/Au)/SnO_2/perovskite/Spiro-OMeTAD/MoO_3/Ag	5.3%	-	Kumar et al.[74]
	SS (type-304)/SiO_2/Au/TiO_2/perovskite/Spiro-OMeTAD/Au	3.45%	-	Kumar et al.[75]
	SS/TiO_2/N719-dye/SS/Pt	2.25%	Stainless steel is used as both the top and bottom electrodes.	Bonilha et al.[76]

2.3 | Polymer substrate

Polymers are fast becoming the substrate material of choice when it comes to flexible PV applications. Because of the vastness of synthetic organic chemistry, there are almost unending polymers to choose from. Further, a change in a particular ligand or moiety can lead to specific changes in the properties of the final polymer. Thus, there is a scope to tune the properties of the polymers according to the requirement of the final application. While metal foil and glass sheet properties can be changed by changing additives and processing methods, the range of properties achievable using polymers cannot be matched. For example, we have polymers with reversible stretchability (elastic limit) from a few percent to 10s of times (1000s of a percent) their original length. There is similar diversity in chemical resistance, optical transparency, hermetic barrier properties, surface energy, and all the other properties that are important for consideration in flexible substrates.[78] In addition, polymer substrates are attracting much attention in the field of flexible solar cells due to their high flexibility, lightweight, low cost, and R2R processing ability. However, the main disadvantages of polymers are thermal stability (less stable than glass substrates), high CTE, and easy permeability to oxygen and water.[28] Specifically, depending on the temperature change, the polymer substrate may contract or expand because of its low T_g and high CTE. This necessitates a thorough rethink of all production procedures in comparison to those that can be heated to high temperatures. Most polymer substrates have a glass transition temperature below 200°C, so they cannot be used to directly deposit thin films or layers using CVD or other high-temperature processes.[79] Therefore, to fabricate a flexible solar cell on polymer substrates, low-temperature processability of the charge transport layer is required. Figure 6 represents a qualitative comparison of polymer substrates with other flexible substrates. Further, polymer substrates are classified as natural polymer (paper) substrates and plastic substrates.

2.3.1 | Paper substrate

Several factors make natural, plentiful, recyclable, and ecologically benign materials particularly appealing for usage in the field of flexible electronics.[80] The rising demand for ecologically friendly and biocompatible goods is thought to make cellulose an infinite supply of raw materials. Therefore, wood remains one of the essential raw materials for obtaining cellulose, but other sources (such as hemp, cotton, sisal, ramie, etc.) can also be used.[81] Natural cellulose-based polymers are one of the greatest inventions in human history that have existed for thousands of years. Their use continues worldwide, such as paper substrate.[70] Moving toward green flexible emerging PVs, the supporting substrate is the most accessible component to replace with a biodegradable material. High flexibility, low weight,

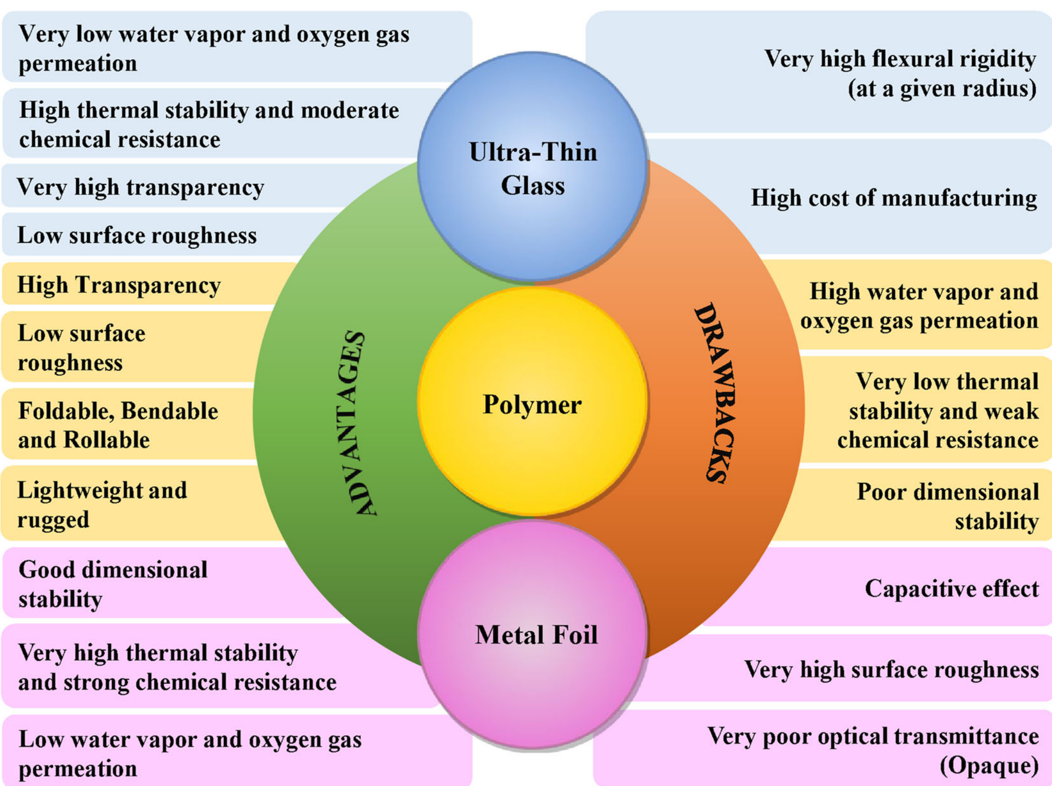

FIGURE 6 Comparison of polymer, glass, and metal-foil substrates for flexible photovoltaic devices.

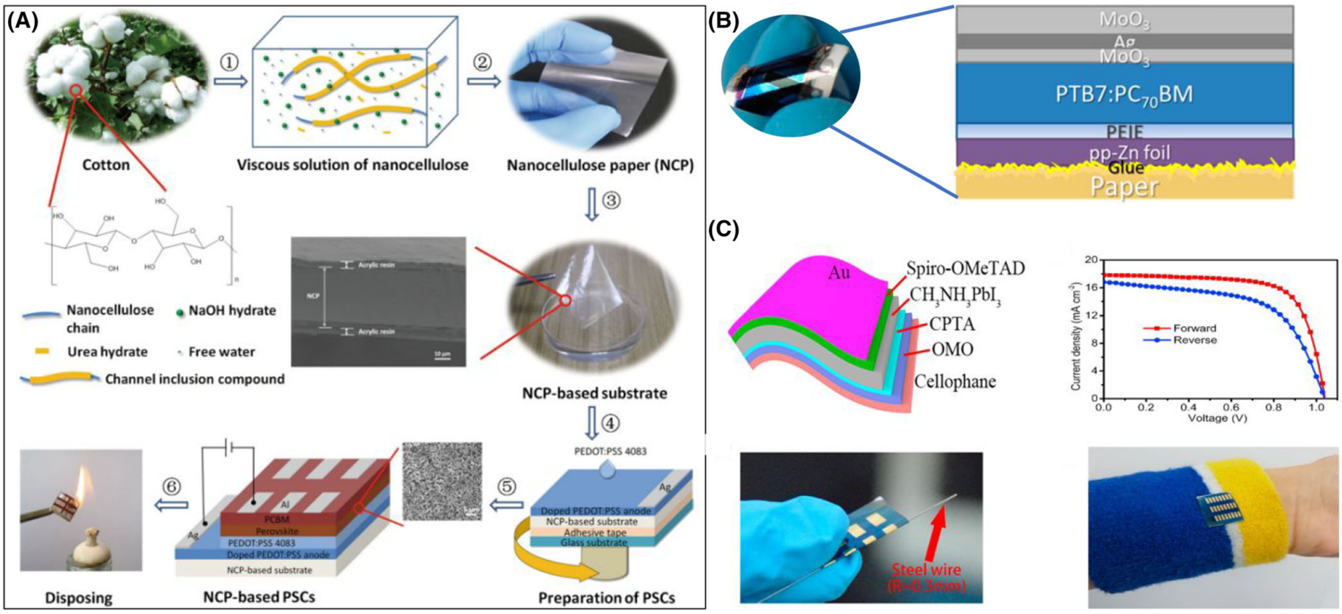

FIGURE 7 (A) Preparation process of nanocellulose paper (NCP)-based substrate and NCP-based perovskite solar cell (PSC). Reproduced with permission from Gao et al.[83] (B) Flexible paper-based organic solar cell with the schematic structure of the device. Reproduced with permission from Leonat et al.[84] Copyright 2014, American Chemical Society. (C) A schematic structure of a cellophane paper-based perovskite solar cell with a J–V curve. Photograph of the flexible PSCs on cellophane bent around cylindrical steel wire with a radius of 0.3 mm and mounted on a human wrist. Reproduced with permission from Li et al.[85] Copyright 2019, Elsevier.

availability, biodegradability, and various adjustable physical qualities are just a few of the possible benefits that cellulose paper as a substrate material might provide.[82] Figure 7A represents the preparation of nanocellulose paper substrate and the fabrication process of solar cell on it. In addition, cellulose-based paper substrates are often more thermally stable than commonly used flexible plastic foils.[86] The higher the crystallite size of cellulose higher the thermal stability.[81]

Among other flexible substrate materials, the paper substrate is much cheaper. The main benefit of employing paper as a substrate is that it can fabricate large-scale devices using low-cost methods like printing.[87] The paper surface, however, poses several difficulties for the fabrication of solar devices. These include excessive roughness, poor chemical and mechanical barrier qualities, and capillary action of liquid molecules into their porous structure. In the case of impermeability to air/moisture, lamination is a standard technology to protect the paper substrate against humidity. Therefore, if solar cells are made on paper substrates, lamination technology can be quickly adapted to improve solar cells' resistance against moisture.[88] Additionally, to fill the holes left by the gaps between the cellulose fibers, mineral fillers, sizers, and clays are employed. After that, various coatings of pigments and fluorescent whitening agents are used to enhance the roughness and whiteness of the paper surface. All of these additives, especially if solution procedures are involved, can have a significant impact on the manufacture of any device on a paper substrate. Since only dry methods or a protective covering have been used, PV systems on paper have proved effective.[89–91] Commercially available paper with coatings from the polyvinyl family of materials is suitable for electronic-device fabrication. Smoothing polyvinyl formal (PVF) layers with a knife-edge coater give them an acceptable root mean square roughness of around 2.6 ± 0.2 nm.[92] Flexible solar cell substrates need high optical transparency in the case of optical transmittance. Still, they also favor high optical haze since it increases light scattering and, as a result, the absorption in the active components. A revolutionary transparent paper with improved transmittance and haze performance that can be produced at a significantly reduced cost, nano-paper is made of wood-based cellulose nanofibers (CNFs).[93]

Additionally, the majority of paper products are opaque, necessitating clear top-contact device designs.[96] Because of its resistance to degradation and ability to undoubtedly lessen the negative effects of electronic waste, high-performance, low-cost, transparent paper is thus potentially a revolutionary material that may influence a new generation of environmentally friendly PV technology. This would be accomplished by addressing the problem of plastic pollution, which has developed into a threat to global ecology. In 2011, the first DSSC on paper substrates was reported with a power conversion efficiency of 1.21%.[88] In 2017, the first PSC fabricated directly on a paper substrate was reported to deliver a maximum power conversion efficiency of 2.7%.[87] The first hole transport material (HTM)-free flexible PSCs were produced on a plentiful, inexpensive, and biocompatible cellulose paper substrate, improving the stability of the PSC. The paper-based flexible HTM-free PSC achieves the maximum PCE of 9.05%.[94] Figure 7B,C shows paper substrate-based flexible solar cells. Various other flexible solar cells on cellulose paper are summarized in Table 3.

TABLE 3 The photovoltaic performance of various flexible solar cells based on paper substrates.

Substrate	Solar cell architecture	Efficiency	Reference
Cellulose paper	Paper/pp-Zn foil/PEIE/PTB7:PC70BM/MoO_3/Ag/MoO_3	4%	Leonat et al.[84]
	Cellulose paper/PEDOT:PSS/perovskite/PCBM/Al	4.25%	Gao et al. Gao et al.[83]
	Paper/carbon/$MAPbI_3$/C60/BCP/Cu@Au	9.05%	Gao et al.[94]
	Paper/PVF/Ag/ZnO/P3HT: PCBM/PEDOT:PSS	3.37%	Rawat et al.[92]
	Paper/PVF/Ag/ZnO/PTB7: PCBM/PEDOT:PSS	6.44%	Rawat et al.[92]
	Paper/Au/SnO_2/meso-TiO_2/$CH_3NH_3PbI_3$/Spiro-OMeTAD/MoO_x/Au/MoO_x	2.7%	Castro-Hermosa et al.[87]
	Wood-based cellulose paper/ITO/TiO_2/perovskite/spiro-OMeTAD/Au	16.8%	Li et al.[95]
	Cellophane/OMO/ZnO/PTB7-Th: $PC_{71}BM$/MoO_3/Al	5.94%	Li et al.[96]
	Cellophane/OMO/AZO/p-i-n-Si/Ag	5.7%	Wang et al.[97]
	Cellophane/TiO_2/Ag/TiO_2/ C60 pyrrolidinetris-acid (CPTA)/$CH_3NH_3PbI_3$/Spiro-OMeTAD/Au	13%	Li et al.[85]

2.3.2 | Plastic substrate

In recent years, a synthetic organic polymer known as plastic has attracted significant attention in the field of flexible electronics over paper-based polymers mainly due to its high durability and longevity. The present research has discovered many biocompatible plastic polymers just like paper. The substrates made up of plastic foil have ubiquitous properties permitting R2R processing. Plastic substrates, incredibly transparent plastic substrates, possess good optical transmittance like that of thin glass; the excellent flexibility and toughness are comparable to those of metal foils.[98] The high flexibility of plastic is due to lower Young's moduli (~3 GPa) and higher elastic elongation (10^{-2}) compared to other substrates (~300 GPa, 10^{-2}).[99] Also, plastic substrates are less expensive than laminating paper. However, they are thermally and dimensionally less stable than glass substrates due to their low glass transition temperature. They have a much higher oxygen and water vapor transmission rate, which is detrimental to PV applications.[28] To overcome the thermal stability problem, the solar cell can be fabricated by a suitable method, such as solution processing that utilizes low temperature. At the same time, the high permeation of oxygen and water in plastic substrates is due to the loosely packed chains leaving many voids inside the material. Through these voids, gas molecules or moisture vapor can enter and instantly interact with the device's active components. Some polymers also tend to absorb water, while others do not. Therefore, it can be avoided by coating barrier layers on both sides of the plastic substrate using a technique such as atomic layer deposition (ALD) that is ideally suited as it produces conformal pin-hole-free coatings.[100–102] For example, metal oxides and nitrides such as aluminum oxide (AlO_x), silicon oxide (SiO_x), titania (TiO_x), silicon nitride (SiN_x), and other ceramic materials are often coated on the plastic substrate as barrier layers using ALD to reduce the permeation of the oxygen and moisture. In addition, a multilayer organic/inorganic barrier stack coating is generally required to obtain a satisfactory transmission rate.[102,103] Unlike natural polymers, the typical plastic foils are shrunk by heating and cooling cycles, but they shrink less if pre-stabilized by prolonged annealing. Therefore, considering the ubiquitous properties of plastic substrates and their potential in advancing photovoltaics, it is worth exploring their application in enhancing the performance of solar cell devices. Further, detailed information on various flexible plastic substrates is discussed.

3 | CLASSIFICATION OF PLASTIC SUBSTRATES

3.1 | PET and PEN substrates

Polyethylene naphthalate (PEN) and polyethylene terephthalate (PET) are excellent commercial semi-crystalline thermoplastic polymer resins of the polyester family. PET is the ethylene terephthalate polymer that is an ester of ethylene glycol and terephthalic acid. In contrast, PEN is a polymer of ethylene naphthalate that is an ester of ethylene glycol and naphthalene dicarboxylic acid.[33] Like glass and metal, they have a long history of industrial usage, which means the infrastructure to produce vast quantities of these materials is already present. Both are considered flexible substrate materials in various electronics applications, including PV technology. The most important reasons are that they exhibit good optical properties with optical transmission higher than 85% in the visible region, high ductility up to 70% elongation at break, and mechanical flexibility under bending or buckling circumstances due to the low value of Young's modulus.[104] Also, they are hydrophobic polymer, have good resistance to a wide range of chemicals and solvents, have excellent dielectric properties, and absorb relatively little water (~0.14%). The water vapor transmission rate of 100-μm-thick PET substrate is ~9 $gm^2\ day^{-1}$, and the PEN substrate is ~2 $gm^2\ day^{-1}$, which is lower than other polymer substrates.[105,106] PET and PEN substrates having low water and oxygen permeation still cannot meet the transmission rate requirement for flexible solar cells. Therefore, the substrate is coated with barrier layers. For instance, the PET substrate is coated with stacks of aluminum oxide-polyacrylate pair that show a reduced transmission rate of $\sim 10^{-5}$ gm^2 day^{-1}.[103] However, multilayer coatings are more expensive compared to plastic substrates. However, their crucial disadvantage is their thermal stability. PET and PEN substrates' glass transition temperatures (T_g) are ~78°C and ~120°C.[107] Both materials deform severely when exposed to temperatures above 200°C, even after pre-stabilization by annealing. At a sustained temperature above 120°C, PET undergoes substantial shrinkage and decreased optical transparency and flexibility. But PEN has a slightly better response to high temperatures because the presence of naphthalene rings stabilizes the polymer.[108] Comparing both, PEN has much lower surface roughness and is more expensive than PET substrates. Both substrates are degradable under ultraviolet (UV) exposure. Therefore, developing efficient and low-cost barrier coating techniques will improve the PV device's demand and performance.

3.2 | Polyether ether ketone (PEEK)

PEEKs, also referred to as polyketones, are a colorless thermoplastic polymer with extraordinary mechanical properties. It is the highest-performance semi-crystalline material available.[109] Young's modulus of elasticity is 3.6 GPa, and its tensile strength is 140–170 MPa, making this polymer highly flexible.[110] The chemical structure of PEEK confers excellent thermal stability up to 350°C and is highly resistant to thermal degradation, which makes it unique from other polymers.[111] The PEEK is compatible with a variety of reinforcing agents, resistant to radiation, chemical, and hydrolysis damage. It is more durable than many metals, which makes flexible electronic uses for it quite appealing. Among the polymers, it has one of the best biocompatibilities. Thus, the stability, biocompatibility, and mechanical properties make PEEK a suitable substrate for PV applications.[112]

3.3 | PC and PES substrates

Polycarbonates (PCs) and polyether sulfone (PES) are a group of non-crystalline (amorphous) thermoplastic polymers containing carbonate groups and diphenyl sulfone in their chemical structures. They are rigid transparent amorphous materials with better impact strength and lower weight than other polymer substrates. Both are interesting due to their high optical transmittance and high glass transition temperature.[113,114] PC has a glass transition temperature T_g of ~150°C. It is a well-known, commercially available optical polymer used in various optical applications due to its combination of chemical stability and optical and mechanical properties.[115] The flexural modulus of PC is ~2.4 GPa, which shows good flexibility and possesses a high impact strength of 960 J/m.[116] However, due to its unfavorable surface characteristics, which include low hardness, low resistance to abrasion and scratching, and low surface energy, it loses some of its desirable bulk properties. As a result, the films that are directly deposited on it typically adhere poorly and have low wettability.[115,117] Over the years, several methods have been developed to modify polymer surfaces for improved wettability, adhesion, and so on. These methods include plasma treatment, chemical treatment, UV/ozone treatment, plasma-enhanced chemical vapor deposition (CVD), and surface grating. Additionally, it has a high permeation rate to water vapor/oxygen (~50 gm^2 day^{-1}) that requires an enormous amount of coating to reduce the permeation rate. While PES polymer is dimensionally stable over a wide temperature range. It can be easily processed and exhibits low mold shrinkage. It has a high temperature at which glass transitions (~220°C). It can also be used continuously under load at temperatures of up to about 180°C and in some low-stress applications to 200°C. PES polymers show significant resistance to acid or base environments in a considerable range of concentrations and temperatures and present optical transparency (greater than 90%) and compatibility with most semiconductor processing chemicals.[118] Although it is biocompatible and has high thermal resistance, the bare PES substrate has the drawback of having higher surface roughness than glass substrates.[119] These polymers' high-impact strength, heat resistance, durability, and high-quality finish justify their expense.

3.4 | Polyimide (PI) substrate

PI is another polymer commonly considered an ideal substrate for many flexible electronic applications. PI is a polymer of the imide monomer, a functional group consisting of two acyl moieties attached to a nitrogen atom. Imides are generally formed from dicarboxylic acids through a reaction with ammonia. PIs are polymers with the central nitrogen molecule attached to an organic group with another attached to the acyl group. PIs share common properties derived from their base structure and composition; however, they can be tuned based on the functional groups R_1 and R_2. The most important property of PIs is their relative stability at moderate temperatures compared to other polymer materials.[33] Kapton is a commercially available PI material, which is stable up to 400°C. Thus, Kapton sheets can be used directly as a substrate for carrying out fabrication processing steps. Kapton is also resistant to many chemicals routinely used in the semiconductor fabrication industry. However, it absorbs light in the visible blue region, thus giving it a yellow tinge. This can limit its applications in the flexible display/PVs space. Another problem with using PI as a substrate is its limited elastic strain, which is reported to be less than 1% for Kapton. This is very low compared to most polymer substrates and can be problematic in using Kapton for stretchable electronics applications or applications requiring extensive strains. This does not limit the flexibility because fragile sheets of Kapton can be fabricated and handled reliably. PI is also commonly used as a substrate for manufacturing flexible solar cells. However, the PIs show good solubility in polar solvents, indicating that the PI substrate may be destroyed in the solution-processed layer-by-layer fabrication process. Therefore, the contradiction between transparency, solution-process, and solvent tolerance is the key issue that should be addressed.[120]

3.5 | Polyetherimide (PEI) substrate

PEI is a member of the PI family of high-performance materials that also includes polyamide-imide (PAI). An amorphous thermoplastic, PEI's polymeric structure has ether linkages to the PI molecular structure. The ether linkage in the PEI provides sufficient flexibility yet retains the aromatic imide characteristics of excellent mechanical and thermal properties.[121] Their advantage over other aromatic PIs is that they are melt-processable and can be molded or extruded into various shapes, parts, and films. Their high aromatic content also makes them radiation stable. A typical flexible substrate of PEI polymer has characteristics that include increased strength and modulus (~2.9 GPa), high dielectric strength, hydrolysis-resistant (~0.25%), high thermal stability due to high glass transition temperature (~217°C), and biocompatibility. Like PI substrates, PEI is transparent but has a yellow tint.[122] Its characteristics are like the related plastic PEEK. When comparing PEI and PEEK, the former is cheaper but has low impact strength and a tighter temperature range.

3.6 | Silicone substrate (PDMS)

Silicones are polymers consisting of a siloxane group attached to other organic moieties. The siloxane group consists of an oxygen atom attached to two silicon atoms attached to three different groups. Silicone polymers typically include a -Si-O-Si- chain with the Si atoms attached to other functional groups. The simplest siloxane polymer is polydimethylsiloxane (PDMS), in which the silicon atom is connected to two methyl groups. Thus, the PDMS chain consists of a repetition of the structure [-Si $(CH_3)_2$-O-] $_n$. Polymers from the silicone family are typically transparent, rubber-like materials with very low oxygen/moisture permeability. Apart from these, they possess very high reversible stretchability (elastic strain limit) and high optical transmittance; they are resistant to chemical reagents and can be formed into thin films that can be handled independently. This makes it an ideal substrate for stretchable electronics applications. However, the surface energy of PDMS is such that it is not wet easily, causing problems with stiction with other thin films, and the glass transition temperature is shallow (~−50°C).[79] Further, PDMS does not respond well to temperatures above 150°C.[33]

3.7 | Polyarylate (PAR) substrate

PAR resins are a family of thermoplastic aromatic polyesters with excellent toughness, heat resistance, UV stability, flexural recovery, dimensional stability, and electrical properties. PAR polymer properties can be tailored by compositional variation, alloying with other high-performance thermoplastics, and reinforcement. This flexibility renders PAR one of the more versatile high-temperature polymers for current and emerging markets served by engineering thermoplastics.[123] PARs are transparent and range in color from light yellow to amber (optical transmittance ~ 87%). The transparent character can be maintained in alloying with other reactive polyesters, while mechanical properties can be tailored to end-use applications. Additional property modification is achieved by targeting mechanically compatible blends. PARs exhibit glass transition temperatures of 180–185°C. However, it has poor chemical resistance.[124]

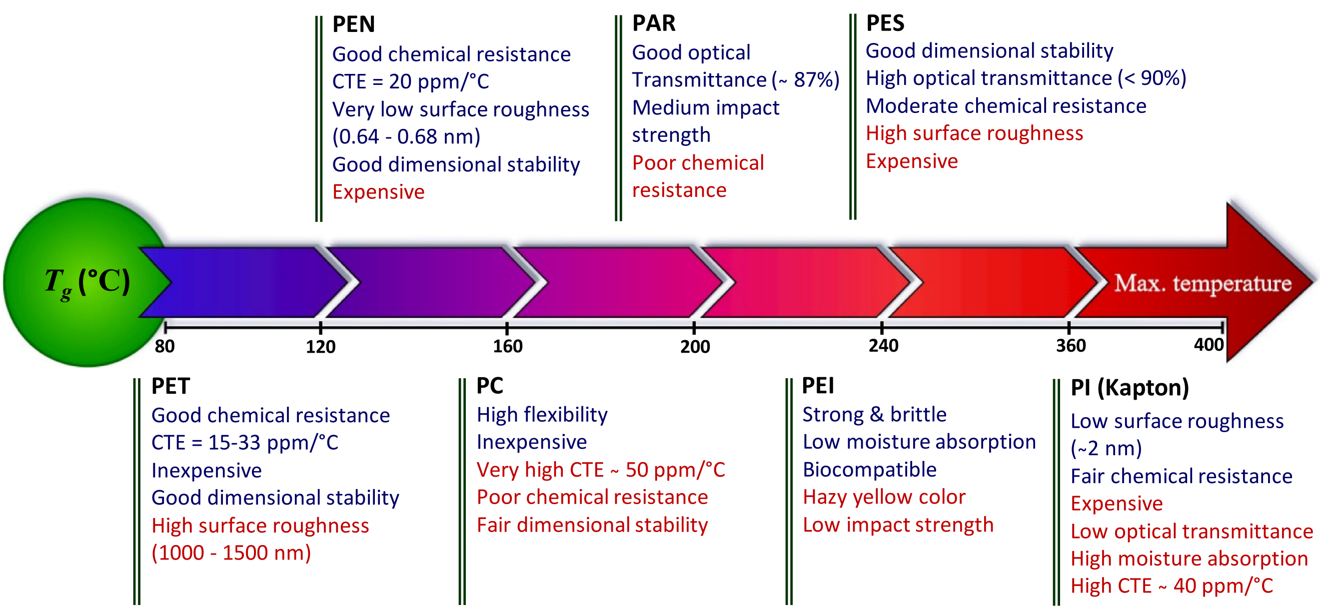

FIGURE 8 Comparison of various plastic substrates for flexible photovoltaic devices.

TABLE 4 Comparison of properties for various plastic-based polymer substrates.

Polymer	Young's modulus (GPa)	Optical transmittance (%)	Dielectric constant	Bandgap (eV)	Glass transition temperature (°C)	Tensile strength (MPa)	Elongation (%)
PET	3	85	3–3.2	3.2	75	120–180	70
PEN	3.3	80–90	2.7–3.2	3.4	120	200	60
PEEK	3.6	20–50	3.2–3.5	3.5	350	140–170	20–40
PC	2.5	85–90	2.9–3.4	3.3	150	60–90	70
PES	2.8	<90	3.2–3.5	3.1	220	70–100	20–50
PI	3.1	60–70	3.3–3.5	3–3.4	300	100–200	5–7
PEI	2.9	85–90	3.2–3.5	3.2	217	80–120	3–5
PDMS	0.87	90–95	2.5	-	−50	3–15	430–640
PAR	2.3	85–87	3.4–3.8	3.5	180–185	50–150	5–20
PVDF	2.9	80–90	7–8	5.6	−38.5	50–57	20–25

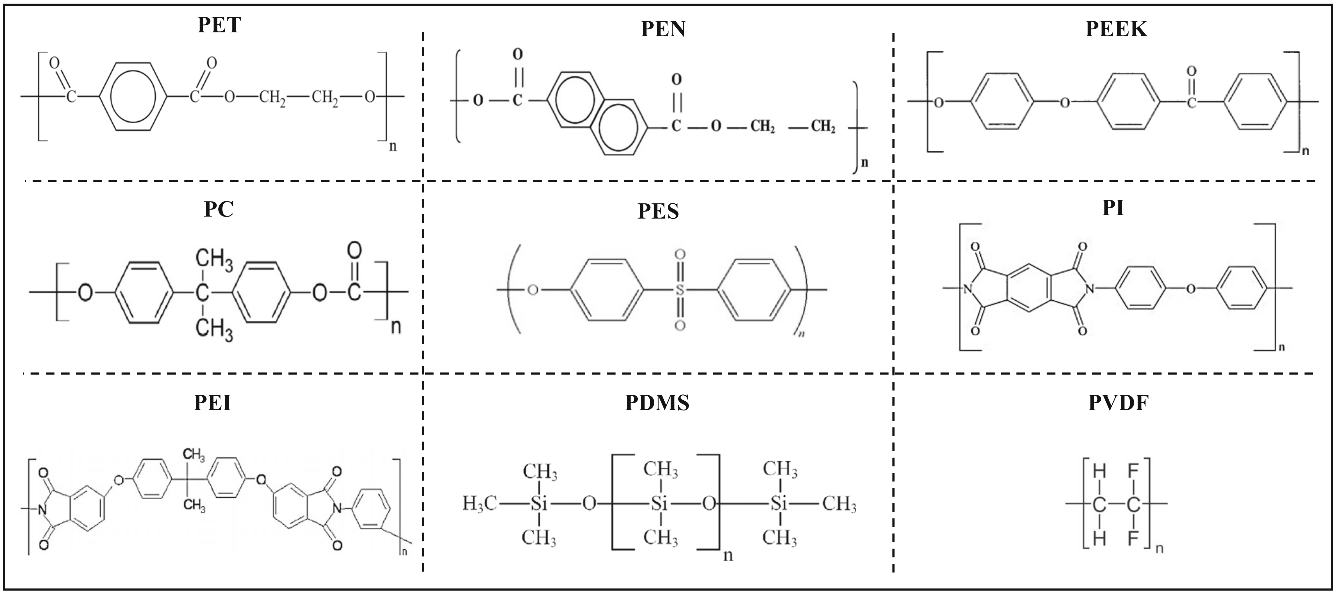

FIGURE 9 Chemical structure composition of various polymers used as substrate.

3.8 | Polyvinylidene fluoride (PVDF) substrate

PVDF is a semi-crystalline thermoplastic fluoropolymer. It is an emerging class of new engineering polymers that combines high mechanical strength with good processability. In recent years, considerable interest has been expressed in the polymer PVDF.[125] The interest was received because it exhibits the most robust piezoelectric properties compared to any other commercial polymer. Also, it retains ambitious standards of purity and is highly resistant to most chemicals, including solvents, acids, and hydrocarbons. Some other characteristic features of PVDF are excellent abrasion resistance, better thermal stability with a glass transition temperature (∼−38.3°C), resistance to UV light and high energy radiation, high dielectric strength, and low water absorption (absorbs less than 5% water).[126] Due to low glass transition temperature, the fabrication on such substrates requires low-temperature processing techniques. One of the most common low-temperature processing techniques for fabricating solar cells on PVDF substrates is solution processing, while other techniques include spray coating, doctor blading, and R2R processing. In the future, such polymers with properties of a flexible substrate can also be used in PV and piezoelectric applications. A detailed comparison of various polymer substrates is represented in Figure 8 and Table 4. Further, the chemical structure composition of various polymer materials is illustrated in Figure 9.

4 | CONDUCTING POLYMER SUBSTRATES

An ideal polymer substrate is electrically insulating and optically transparent. However, the application of PVs needs an electrically conductive substrate that can provide an easily accessible common terminal to interconnect various parts of the cell. Unlike metal substrates, polymer substrates are not conductive and cannot act as cell electrodes. Therefore transparent conductors (TCs) are deposited as a thin-film layer on the polymer substrates. The transparent conducting films (TCFs) enable their use as current collectors (electrodes) in solar cells without blocking light, that is, letting light into the solar cell to convert sunlight into energy while acting as collectors for converted energy.[127] Transparent conductive materials possess wide bandgaps with tremendous energy than visible light. As such, these materials do not absorb photons with energies below the bandgap value, and visible light passes through. The solar cell application often requires a broader range of transparency beyond visible light to efficiently use the entire solar spectrum. Therefore, it is evident that TCs have several diverse applications in solar energy utilization and efficiency.[127]

Transparent conductive materials fabricated on polymer substrates for PV applications can be inorganic and organic. The inorganic transparent conductive material mainly includes Active oxides (TCOs). Metal nanoparticles/nanowires, carbon nanotubes (CNTs), graphene, and conducting polymers are examples of organic transparent conductive materials with great mechanical flexibility, low cost, and high infrared light transparency. All of the contenders support solution-method processing, which is essential for creating flexible transparent conducting layers (TCFs) on polymer substrates at low cost and over a large surface area. Slot-die, spray, dip, and spin coating are among the straightforward coating techniques that may be used to fabricate large-area flexible TCFs on an R2R basis.[128] Flexible TCFs may also be produced on a wide scale using a variety of printed processes, including inkjet printing, screen printing, offset printing, and gravure printing.[129] In subsequent sections, the properties of various TCFs compatible with polymer substrates are discussed.

4.1 | TCOs

The most used TCFs are TCOs. These are transparent conducting metal oxides called wide-bandgap semiconductors (bandgap > 3.2 eV).[130] TCOs have made a special place in the field of flexible PVs because of their unique combination of transparency and electronic conductivity. Common TCO base materials include indium oxide (In_2O_3), tin oxide (SnO_2), and zinc oxide (ZnO).[131] Doping the base material with specific metal ions significantly increases transparency and conductivity due to the external doping effect. Common TCOs include indium tin oxide or tin-doped indium oxide (ITO), fluorine-doped tin oxide (FTO), indium zinc oxide (IZO), aluminum-doped zinc oxide (AZO), and W-doped In2O3 (IWO).[39,132–134] Among the TCOs, ITO and FTO are commercially and widely used as TCFs in PV devices. Since the optical and electrical properties of the TCOs are highly dependent on the substrate type and deposition conditions. Polymer substrates are not a good fit for the FTO because of their high-temperature needs. For instance, IZO can be deposited at 100°C, whereas indium oxide (ITO), azo oxide (AZO), and indium tungsten oxide (IWO) can be produced at ambient temperature. FTO is specifically formed at substrate temperatures exceeding 350°C.[134,135] In the case of AZO, researchers have shown that AZO chemically interacts with the active layer, reducing cell productivity.[39,133] For IZO, the sheet's low resistance is advantageous, but it is brittle and breaks when bent along small radii.[128,132] Kim et al. produced IWO for flexible solar cells using a room-temperature arc plasma ion plating process. Due to improved optical transparency, especially in the near-infrared (NIR) range, and a larger work function (4.85 eV) compared to ITO (4.65 eV), the IWO-based flexible solar cell exhibited a superior power conversion efficiency of 11.33% to the ITO-based (10.6%).[134]

Nevertheless, the most commonly utilized TCF in flexible solar cells is ITO. It is an advantageous metal oxide exhibiting high conductivity and transmittance. ITO can also be ascribed to low-temperature fabrication methods, chemical resistance to active substances, well-aligned band structures, and, most importantly, an established mass-production approach. In recent studies, flexible solar cells with more than 20% performance are usually based on commercially available polymer/ITO substrates.[136,137] Despite its widespread use, there are many obstacles. An ideal flexible solar cell is expected to retain about 90% of its original efficiency after being bent 1000 times at a radius of 4 mm to meet the requirements of flexible and wearable electronics. It is difficult for ITO-based flexible solar cells to meet these requirements due to their lack of flexibility and breakage characteristics. Cracks in the ITO film and increased sheet resistance during bending cycles degrade device performance.[36,138–140] The most costly component of flexible solar cells is ITO, which limits their ability to be mass-produced due to the shortage of indium and the high cost of vacuum deposition processes including sputtering, evaporation, and electroplating.[141] ITO films have high surface roughness due to the sputtering effect, leading to current leakage and poor flexibility. ITO reduces solar energy use due to its low transparency in the NIR region. ITO becomes chemically unstable when exposed to the active layer.[142,143] In addition, during the coating process, ITO is unable to get the best optoelectronic characteristics on flexible plastic substrates. Sputtering for ITO coating is often carried out at high temperatures when high-resistance substrates like glass are utilized. At 90% transmittance (T [%]), it produces sheet resistances of under 20 Ω sq^{-1}. When a flexible plastic substrate is utilized, ITO is often deposited at lower temperatures to protect the substrate, raising the corresponding sheet resistance to 200 Ω sq^{-1} at 90% transmittance. The brittleness of ITO and its propensity to shatter under mechanical stress are only a couple of the shortcomings listed above that have severely restricted its use in flexible devices. However, ongoing advancements to ITO's optical, electrical, and mechanical characteristics are being made. At the trade-off of greater virgin resistance, decreasing the thickness allows substrates to be more flexible and to shatter less during the bending test.[144] Besides, the deposition method can improve the photoelectrical quality of the ITO conducting layer. Amorphous structure results in high sheet resistance, low optical transmittance, a rough surface, and inadequate flexibility in ITO films produced by direct current sputtering at room temperature.[38,135,145] Therefore, an alternative method, such as the vertical plasma arc ion plating method, has been introduced to produce a smooth surface of ITO on polymer substrates with low sheet resistance and high optical transmittance.[145] Additionally, the temperature while annealing is important. In contrast to PET substrates, which are more temperature-sensitive due to the activation of the Sn dopant and the crystallization of the ITO layer, ITO films on colorless PI substrates showed reduced sheet resistance at annealing temperatures up to 300°C.[135] In addition, structural engineering optimization of ITO electrodes is also beneficial to improve light utilization.[146]

4.2 | Metal conductors

4.2.1 | Metal Nanowires

Metal nanowires are also considered an extremely competitive candidate for use as TCEs in emerging flexible solar cells. It is because of their high electrical and optical properties, low resistivity, high flexibility, high figure of merit, relatively good chemical stability, and bending durability.[147] Metals such as copper (Cu), silver (Ag), and aluminum (Al) have been reported as highly conductive materials and low sheet resistance materials.[148,149] Metal nanowires may readily be deposited on films using typical solution-based techniques including spin coating, vacuum filtering, and R2R compatible methods like spray coating, bar coating, and slot-die coating, which are advantageous for large-scale production.[150–153] The potential of silver nanowires (Ag NW) as TCEs was shown by Lee et al. in 2008. According to their findings, the optical transparency (>80%) of the solution-processed silver nanowire mesh electrodes was on par with or more than that of metal-oxide thin films, which had a sheet resistance of around the same value (<20 Ω/sq). When utilized in OSCs, the electrodes displayed a performance that was comparable to that of gadgets built using a regular transparent metal-oxide electrode.[154] Among metal nanowires, silver nanowires (Ag NWs) are widely used as transparent electrodes in flexible solar cells due to the superior electrical properties of silver (Ag) materials and upscaling synthesis. Generally, the conductivity of Ag NWs could be improved when the length and diameter of each wire increases.[155] Ag NWs can be synthesized by hard-template and soft-template methods. The conductivity and transmittance could even be better than those of ITO electrodes. The mechanical flexibility of Ag NWs has received increased attention. Figure 10B shows the sheet resistance under bending for three different types of conductive materials. The Ag NWs showed a lower sheet resistance in comparison to other TCFs. One of the main advantages of Ag NWs compared to metal oxides, such as ITO and FTO, is their total optical transmittance. As shown in Figure 10A, the total optical transmittance of Ag NWs, compared with metal oxides in the NIR region, is significantly lower. Thus, Ag NWs can be used to fabricate transparent or semi-transparent solar cells.[156] To create semi-transparent solar cells in the visible range, low-diameter silver nanowires can be used to create a low-haze network and a high-transparency conductive film. Due to the significant NIR spectrum absorption of this cell, the average visible wavelength range is transmitted at a rate of more than 60%.

Poor chemical stability, high junction resistance, high surface roughness, limited coverage (less than 40%), and low adherence of Ag NWs to polymer substrates are among the serious problems with Ag NWs, though.[157] Moreover, in the case of PSCs, the halides in perovskites would chemically react with Ag and lead to severe degradation of device performance.[153,157,158] Additionally, when the Ag NWs network is exposed to air or severe conditions, corrosion and oxidation can readily occur. A short circuit and electrode deterioration of the perovskite and Ag NWs result from the rough surface produced by the agglomeration of nanowires during the solution process.

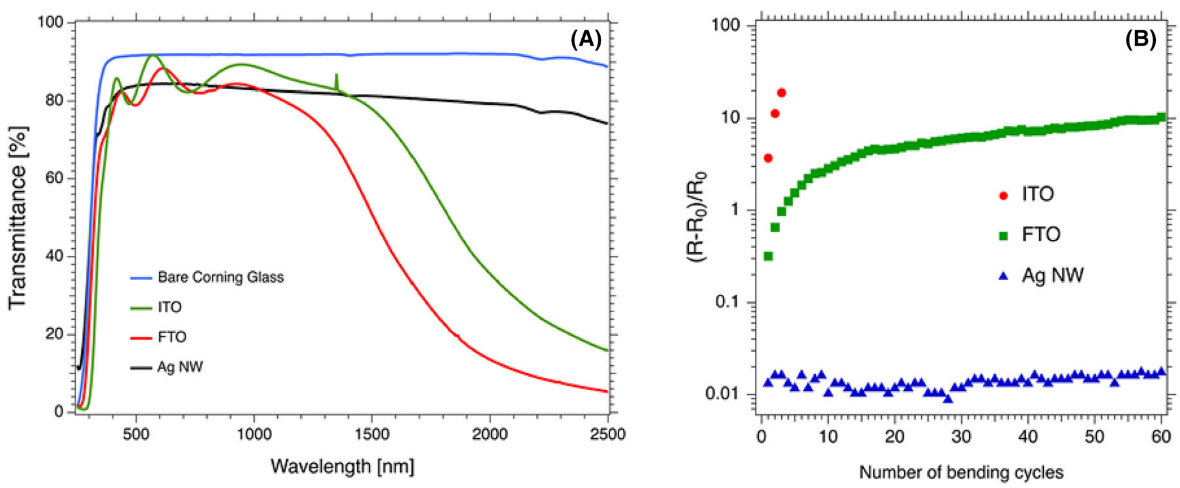

FIGURE 10 (A) the total optical transmittance for three types of transparent conductive materials: ITO, FTO, and Ag NWs network; and (B) the sheet resistance under bending. Reproduced with permission.[156] Copyright 2017, MDPI.

There have been several published methods for obtaining a smooth surface, including barrier layer deposition (transparent metal oxides, poly(3,4-ethylene dioxythiophene):poly(4-styrene sulfonate) [PEDOT:PSS], graphene), as well as the use of unconventional deposition techniques such as capillary printing.[155,159–161] To minimize extra energy loss, the protective barrier should also have superior ion permeability, mechanical resilience, and high optical transparency. TCO components such as ITO, AZO, and ZnO have been placed between the metal electrode and the halide perovskite as a protective layer. As an instance, fluorine-doped ZnO (FZO), which was deposited over the Ag NWs layer, showed increased conductivity as well as chemical and mechanical stability.[160] Similar to commercial ITO/PET transparent electrodes, ATO placed on Ag NWs film (Figure 11A) demonstrated 76–82% visible area transmission with low sheet resistance.[140] As the layer between Ag NWs and perovskite, conducting polymer PEDOT:PSS with good mechanical flexibility is advantageous. By filling the open spaces and joining the silver nanowire junctions, the incorporation of PEDOT:PSS and Ag NWs has benefits in increasing surface flatness, adhesion to substrates, and electrical conductivity. Ag NWs/PEDOT:PSS transparent electrodes are fabricated on polymer substrates (e.g., PET) via a scalable, R2R slot-die process.[165] Combining with graphene is one of the most efficient ways to preserve Ag NWs because of its strong chemical and thermal stability, high optical transparency, and easy fabrication method. However, because of the challenging manufacturing process, these approaches with extra protective layers are incompatible with the large-scale chemical method.

Furthermore, the random Ag NWs network created using traditional solution approaches performs poorly due to its high surface roughness and low optical transmittance. Whereas Ag NWs employed on polymer substrate via capillary printing process resulted in smoother surface morphology and higher optical transmittance (92.3% at 550 nm) compared to random Ag NWs (90.8% at 550 nm) as demonstrated in Figure 11D.[163] Also, this method prevented direct contact between active material and electrode, resulting in no electrical conductivity loss during the entire fabrication process. Contact transfer methods and applying pressure are various other methods to overcome the problem of high surface roughness. To achieve a surface roughness reduction of less than 1 nm in contact transfer methods, it is recommended to transfer Ag NWs from a rigid

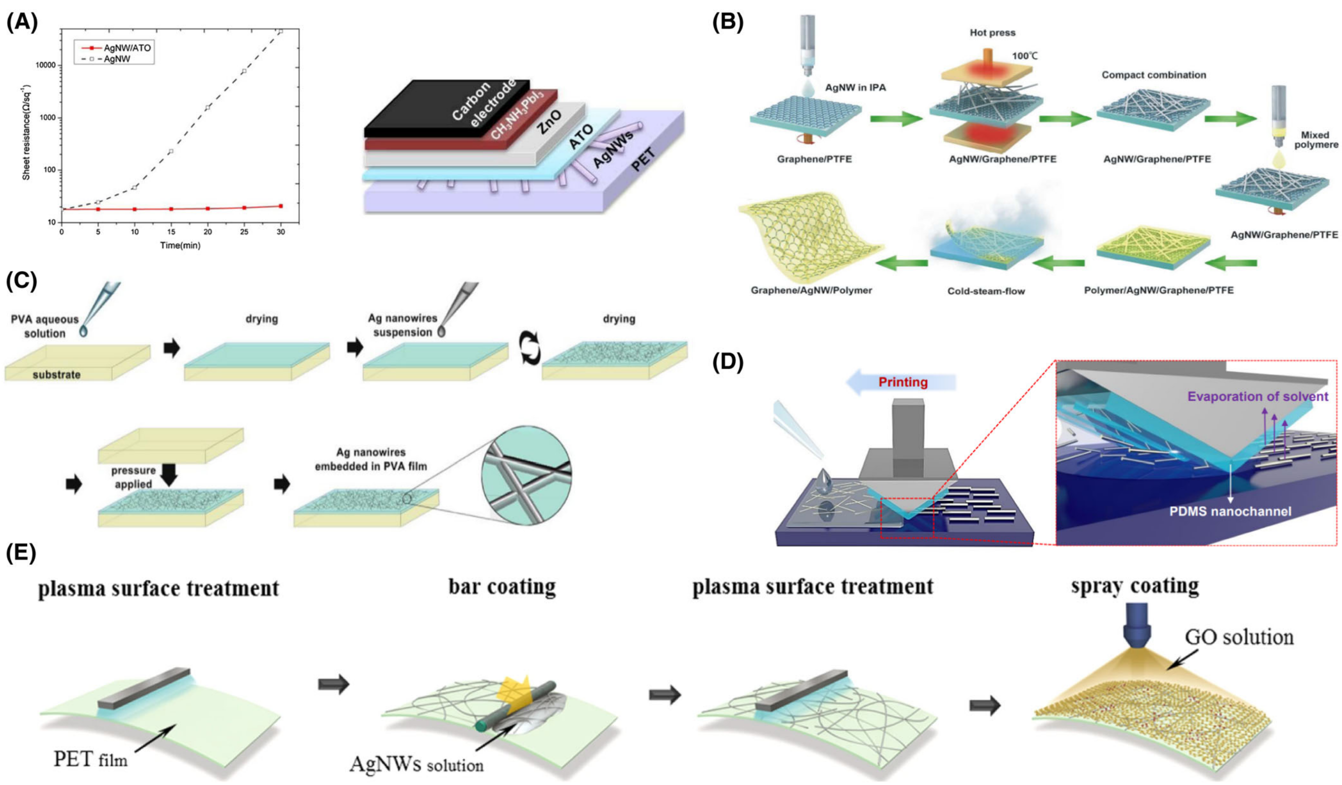

FIGURE 11 (A) Chemical stability of Ag NW and Ag NW/ATO composite transparent electrodes. The device structure of FPSCs is based on PET/Ag NWs/ATO substrate. Reproduced with permission from Jon et al.[140] Copyright 2020, Elsevier. (B) Schematic illustration of the procedures in fabricating composite PET/graphene/Ag NWs electrode. Reproduced with permission from Dong et al.[158] Copyright 2016, American Chemical Society. (C) Process of embedding Ag NWs into their initial polymer substrates by applying pressure to the fabricated transparent conductive composite. Reproduced with permission from Lian et al.[162] Copyright 2017, Elsevier. (D) Schematic of fabricating orthogonal Ag NWs via capillary printing. Reproduced with permission from Kang et al.[163] Copyright 2015, American Chemical Society. (E) Schematic illustration of the plasma surface treatment in the GO/Ag NW/PET film fabrication. Reproduced with permission from Moon et al.[164]

substrate (such as glass) and implant them into a polymer substrate.[166,167] Also, this method solves the problem of low adhesion on polymer substrates. On the other hand, applying pressure on Ag NWs, it gets embedded into the initial polymer substrate (Figure 11C) and achieves an RMS of ∼4.6 nm.[162]

The poor adhesion of Ag NWs may bring down the device's efficiency. Various methods to improve the adhesion between Ag NWs and substrates are (1) the adhesion can be increased by heating the substrate and the Ag NWs, which can strengthen their interaction. Additionally, intense pulse-light (IPL) can be used to improve the connection between the Ag NWs and the plastic substrate.[168,169] It is noteworthy that the Ag NWs and substrate may both be rapidly heated to high temperatures using the IPL approach, boosting their contact levels. These techniques can strengthen adhesion, although they are used less frequently than the techniques that follow since they are more appropriate for larger areas. (2) It is desirable to use polymer binders with high oxygen content, including carboxylate and hydroxyl groups, which have a great affinity for binding to the Ag NW surface. cellulose polymers (such as hydroxypropyl methylcellulose [HPMC]), CNFs, and other oxygen-rich polymers with strong solubility have all been shown to have the ability to increase adhesion.[170–172] (3) An effective way to increase adhesion strength is to overcoat with a second layer made of metal oxides using either ALD or sol–gel techniques. (4) Surface modification of the substrate: Corona and plasma treatments are common ways of producing hydrophilic surfaces that can improve material adherence on polymer substrates (Figure 11E).[164] Despite the problems of using Ag NWs as a TCF, it has one and more solutions. Therefore, shortly, Ag NWs are anticipated to replace ITO-based TCFs as the dominant technology in the flexible solar cell industry. In addition to Ag nanowires, gold nanoparticles (Au NPs) also have unique optical and electrical properties that make them suitable for use as transparent conducting materials. The Au nanoparticles are highly conductive, and their small size means that they can be dispersed evenly throughout a transparent material without affecting its transparency. Also, gold nanoparticles exhibit what is known as a surface plasmon resonance, which allows them to absorb and scatter light in a way that can be controlled by changing their size and shape. This property can be exploited to create materials with specific optical properties, such as selective transmission or reflection of certain wavelengths of light.[173–175]

4.2.2 | Metal grids

Researchers have investigated and considered another group of TCFs made up of metal, the metal mesh (grid). Homogeneous metal mesh structures fabricated by nanostructure lithography techniques such as photolithography and nanoimprint lithography are also attracting much attention as another alternative to ITO because light transmittance and electrical conductivity can be easily controlled by controlling the line width and film thickness.[32,176] However, this approach produces classical two-dimensional metal mesh structures with a common trade-off between light transmission and electrical conductivity. Wider and denser networks have higher electrical conductivity but lower optical transmission costs and vice versa.[177,178] The three-dimensional metal grid mesh structure produced by secondary sputtering meets the majority of the requirements for transparent electrodes, including excellent optoelectronic performance (i.e., low sheet resistance with a transmittance of ∼85%), a flat surface (RMS roughness of approximately 5 nm), no haze (about 0.5%), high stretchability (no significant change in resistance for applied strain 15%), and strong adhesion to polymer substrates.[179] Metal grids, such as Ag, Au, Cu, and Ni, are typically coupled with other transparent conductive materials to create highly efficient composite electrodes. PET/Ag-mesh/ITO is a unique flexible hybrid electrode that combines silver (Ag) mesh-embedded poly(ethylene terephthalate) film (PET/Ag-mesh) with a thin ITO layer. It has excellent photoelectric characteristics, chemical stability, and mechanical endurance. The ITO film is a barrier between the active layer and metal-induced deterioration.[180,181] Cu-based electrodes are less expensive than Au and Ag-based electrodes and have reduced diffusion into active layers of cells.[182] Furthermore, the Ni mesh-based electrode is more stable because of the quickly generated dense nickel oxide on the surface of the Ni mesh, which prevents further degradation. Wang et al. created a transparent bottom electrode with a PET/Ni-mesh/PH1000 configuration, an outstanding optical transmittance of about 85–87%, and a low sheet resistance of about 0.2–0.5 Ω/sq, which is superior to the Ag-mesh counterpart.[183] Metal meshes made using the photolithography approaches are expensive and time-consuming, as it entails evaporating metal films on a pre-patterned photoresist film that has been selectively exposed with a photomask and then peeling the photoresist.[176,182,184] Hexagonal silver gratings are efficiently fabricated through R2R nanoimprinting lithography. Nanoimprinting lithography is an effective method for fabricating metal meshes. The first step in nano printing is to groove the substrate through a patterned mold. Metallic nanoparticle ink is used to fill the grooves, and extra ink is repeatedly scraped off. Using this strategy, they fabricated highly flexible and ultrathin PET/Ag-mesh/PH1000 hybrid electrodes exhibiting transmittances of 82–86% at surface resistances as low as 3 Ω/kV, corresponding flexible PSCs exhibiting high efficiency has been achieved 14%.[32] Due of pinholes between metal nanoparticles, top-down nanoimprinting procedures use little Ag ink and even have inadequate conductivity.

4.3 | Carbon nanomaterials

4.3.1 | CNTs

Carbon-based nanomaterials, that is, graphene and CNTs, exhibit excellent physical and electrical properties, including enhanced conductivity and mobility. The earth's abundant carbon nanomaterial is attracting more and more attention due to its high optical transparency and low surface resistance. The high optical transmittance makes these materials particularly attractive for use as TCEs in PV cells. Moreover, carbon nanomaterials also have mechanical flexibility and environmental resistance and deliver low-cost, R2R ease of production. Transparent carbon nanostructures outperform ITO equivalents in electrical and optical characteristics. Due to these exciting properties, research on manufacturing processes, modification methods, and patterning methods of various CNTs and graphene-based transparent electrodes for optoelectronic and PV devices is increasing. Furthermore, because of their simple synthesis and simple transfer approach, CNTs have high reproducibility. The common synthesis methods to grow CNTs are CVD, electric arc discharge, laser ablation, pulsed laser deposition, and floating catalyst chemical vapor deposition (FCCVD). The FCCVD technique has remarkably progressed as it reduces production costs.[57] However, these techniques require very high temperatures for direct deposition, which a polymer substrate cannot withstand. In such cases, a film transfer technique can be employed to place the synthesized CNTs on polymer substrates. The polymer substrate is wetted with a few drops of ethanol or chlorobenzene during the transfer process to ensure adherence and better electrical contact. Although a high conductive substrate can be obtained using the transfer method, it is incompatible with large-scale production due to complex procedures. To fabricate CNTs directly onto the polymer substrates, techniques such as spray coating, dip coating, and inkjet printing have been acknowledged. These techniques are most popular since they are simple, cost-effective, and can easily scale up to large production.[185]

Furthermore, CNTs can be described as cylindrically arranged graphene sheets. Based on the number of concentric graphene cylinders, CNTs are divided into single-walled, double-walled, multi-walled, and few-walled CNTs. A single layer of graphene makes up single-walled carbon nanotubes (SWCNTs).[186] SWCNT conducting layers present remarkable advantages compared to other materials used as TCFs. It exhibits better electrochemical properties, high resistance to acids, a wider potential window than ITO, ease of fabrication, and superior mechanical strength.[187,188] Additionally, the hydrophobic nature of SWCNTs prevents moisture from penetrating them, which greatly improves device stability. But achieving a homogeneous coating is hampered by this innate hydrophobicity.[188] Yoon et al. embedded SWCNTs in a polymer substrate (PI) and created a CNT polymer matrix that exhibits excellent mechanical strength, bendability, and stability. The ultrathin CNT polymer substrate served as an electrode to a flexible PSC with a sheet resistance of $82\ \Omega\ \mathrm{sq}^{-1}$ and optical transmittance of 80% at visible range. The flexible solar cell obtained a maximum PCE of 15.2% using a CNT-PI substrate.[189] There are also double-walled CNTs (DWCNTs) and multi-walled CNTs (MWCNTs). Unlike hydrophobic SWCNTs, the MWCNTs are solution processable. Composed of two concentric graphene cylinders, DWCNTs combine the advantages of MWCNTs and SWCNTs, exhibiting excellent transparency and conductivity while maintaining processability and chemical stability in solution.[190] The light transmittance of MWCNT films is lower in the visible light region than that of SWCNTs.[191] In addition, the PSCs based on modified DWCNTs achieved higher efficiencies of up to 17.2% with smoother film morphology and more favorable alignment of energy levels than SWCNTs, indicating the potential for using DWCNTs in flexible devices.[190]

4.3.2 | Graphene

Although individual CNT has high conductivity, their conductivity is constrained by the high inter-tube surface energy of CNTs.[192] In a wide wavelength range, single-atomic-thick graphene is more conductive, smoother, and more transparent than CNTs.[193] For instance, the two-layered graphene/PET electrode exhibits 87.3% optical transmittance at 550 nm and a resistance of 300 Ω/sq sheet.[194] Graphene is a desirable choice for the TCF/electrode since its transmittance is higher than ITO's, particularly in the NIR range.[195,196] Additionally, graphene is mechanically and chemically stable. It is often produced on Cu, Ni, or Pt foils using the chemical vapor (CVD) process and is then deposited onto substrates either by dry or wet transfer. But the process of transferring graphene poses a challenge and reduces its reproducibility.[192,197] It's encouraging that the polymer/graphene substrates are now on the market. Due to its irregular form, tiny grain sizes, and high-resistance grain boundaries, the pristine graphene layer of the single-layer graphene created by CVD exhibits high sheet resistance (over 1 kΩ/sq).[192] The conductivity of graphene can be increased by lowering the sheet resistance to tens to hundreds of Ω/sq using two main techniques: stacking the graphene sheets and chemical doping. The ideal number of graphene layers must be managed since there is a trade-off between electrical conductivity and optical transmittance when stacking graphene sheets.[61] Therefore, proper chemical doping is an effective way of adjusting the conductivity and fermi level. The widely used dopants include $AuCl_3$, MoO_3, and bis(trifuoromethanesulfonyl) amide (TFSA). In comparison, Jeon et al. replaced ITO with MoO-doped SWCNTs or MoO-doped graphene, TCO-free inverted flexible PSCs were demonstrated, and their efficiency and mechanical stability were compared. Due to higher optical transmittance and better film morphology, graphene-based flexible PSCs exhibited 13.3% higher efficiency than SWCNT-based flexible PSCs with a PCE of 11.0%.[195]

However, SWCNT-based flexible PSCs showed higher stability compared to graphene-based cells due to randomly oriented SWCNT entanglement and fewer defect sites. Overall, they decided that graphene was the best choice as a flexible electrode in PSC. Because of the physical transfer process and the lack of chemical bonding, graphene and substrates have a weak adhesion, which causes inadequate contact and significant mechanical deformation during flexural experiences.[198] To create ultra-flexible devices, more adequate contacts for graphene must be developed. By creating a chemical link between graphene and substrates, 3-aminopropyl triethoxysilane (APTES) can serve as an interlayer to enhance the adherence of graphene sheets, resulting in dramatically increased mechanical robustness of substrates.[199,200] A cross-linkable olefin-type polymer interlayer has also been employed to reduce the substrate's surface roughness and increase the adherence of graphene electrodes.

As a result, both CNTs and graphene have exceptional chemical stability and mechanical flexibility, which is advantageous in developing low-cost, extremely stable, and long-lasting conducting polymer substrates for flexible solar cells.

4.4 | Transparent conducting polymers

Organic substances with high electrical conductivity, transparency, and flexibility are known as conductive polymers. Due to these properties, polymers are applied as transparent conductive layers (electrodes) in various optoelectronic devices. Due to their low weight, mechanical flexibility, and exceptional compatibility with plastic substrates, transparent conductive polymers offer benefits over conventional electrode materials.[201] The two primary types of conductive polymers are coordination polymers and organic polymers. Cationic metal centers bound by ligands make up the distinctive structure of coordination polymers. High absorption in the visible range limits the creation of optically transparent electrodes, even though the self-assembly of nanoscale building blocks to construct highly ordered coordinated polymers improves conductivity. Conductive organic polymers are another promising replacement material for ITO. Polyaniline (PANI), polypyrrole (PPY), and PEDOT:PSS are the three most important conducting organic polymers. PEDOT:PSS has demonstrated its ability to replace conventional ITO. These polymers are highly transparent, flexible, good conductors, and easy to fabricate, making them suitable for optoelectronic devices.[27] Transparent conductive polymers can be a better alternative to ITO as TCEs. It is readily available and provides a very flexible film compared to ITO. PEDOT:PSS is 20 times more conductive than ITO before deformation. However, these polymers still need to exhibit better high-temperature stability and a disproportionate combination of sheet resistance and transparency (without composite formation) but perform better in composites. Conductive polymers require further improvements to achieve high-performance devices compared to ITO and other TCE materials. Additionally, the essential advantage of polymeric TCs is that they can be processed in solution, making them easy to process on R2R machines using many coating and printing methods.[202]

The various transparent conducting materials have been summarized above with their benefits and solutions to their drawbacks. Since transparent conductive materials have enormous properties (high optical transmittance and energy collector) and are compatible with polymer substrates with low sheet resistance and good adhesion. Depositing a layer of a transparent conductive material upon polymer substrates makes it conductive and produces a good-performance PV device.

5 | POLYMER SUBSTRATES BASED FLEXIBLE EMERGING PVs

Solar cells on flexible polymer substrates have become a promising direction in PVs among developed technologies for collecting sunlight. The flexibility and compatibility to various materials provided by polymer substrates further enhance the effectiveness of PV systems due to their compatibility with R2R production, capacity to be included into curved surfaces, resilience to complex deformations, lightweight design, and simplicity of storage and transportation. Although flexible silicon-based solar cells and other thin film PV systems have previously been created, their high cost and small bending radius still need to live up to people's expectations.[203] The emerging solar cells, including dye-sensitized and PSCs, have proven appealing due to their tunable colors, significantly less thickness, and low-temperature fabrication (below 150°C).[204] Emerging PVs' low-temperature fabrication property brings various polymer substrates into its account. This section reviews emerging solar cells with their benefits and drawbacks. In addition, a literature review of different polymer substrate-based flexible solar cells is provided.

5.1 | Flexible DSSCs

To address the present and foreseeable energy issues, a lot of researchers have concentrated on creating high-efficiency and affordable solar devices. In this regard, the DSSC reported by O'Regan and Gretzel in 1991 is one promising candidate.[205] A DSSC is a thin film solar cell that uses a photo-electrochemical system to convert visible light into electricity.[206] It is a relatively breakthrough PV technology that produces electricity for indoor and outdoor use, providing users with a platform for converting both natural and artificial light to power various electrical devices.[207] A DSSC's synthesis is economical and environmentally friendly, and the thin film structure is light and suitable for automated manufacturing. These solar cells outperform thin films but fall short of crystalline solar cells in terms of efficiency.

Furthermore, DSSC can be designed into flexible sheets, has a low-cost sensitization material for low-cost fabrication, is easy to manufacture, has a low process temperature, and can perform effectively under partial light conditions. Because of the lower overall manufacturing cost, DSSC can be predicted to have a higher return on investment than silicon-based solar cells.[208] A DSSC is a sandwich structure made up of a photoanode, CE, electrolyte, and dye sensitizer in addition to a conductive substrate. DSSCs are designed to capture sunlight to generate electrons and mimic the process of chlorophyll photosynthesis. For this reason, nanostructured semiconductors are usually deposited on conductive substrates and then sensitized with molecular dyes capable of absorbing in the visible light range. The semiconductor dye system is the heart of the device.[209] DSSC generally has a rigid construction since it uses glass materials with TCFs as the conductive substrate to withstand high production temperatures.[210]

These structures have several drawbacks because of their weight, stiffness, and lack of flexibility despite having several advantages, including high conductivity and transparency, as well as high efficiency and stability. Later, R2R bulk manufacture and incorporating DSSCs in numerous portable devices were prohibited.[217] As a result, in recent years, a flexible type of DSSC has been developed, enabling the qualities of low weight and flexibility, thus increasing DSSC's utility and keeping them abreast of solar cell technological advancements. All functional layers, particularly the conductive substrates, which also serve as electrodes for active layers and devices, must be made flexible to construct a flexible DSSC. The conductive substrate, which often comes in pairs, makes up the majority of the DSSC structure and is sandwiched between the other components. They help prepare the photoanode and counter-electrode for device assembly by supporting the photoanode materials and counter-electrode catalyst.[218] Flexible DSSCs are often made from a plastic/polymer conductive substrate (such as ITO-PET, ITO/PEN). Figure 12 shows various DSSC fabricated on conductive polymer substrate. The device is adaptable to high throughput operations including R2R forming, screen printing, and others while still being lightweight and versatile.[219] The flexibility of the plastic (polymer) substrate allows the device to be shaped more freely and integrated into a curved shape for mobile and wearable applications.[220] As a result, producing the DSSCs on flexible substrates would allow for significant cost savings and R2R mass production. Metal substrates have been employed as conducting substrates in flexible DSSC, in addition to plastic substrates.[221] On the other hand, metal substrates have various downsides, such as metal mesh having a low effective area to take sunlight and metal foil having little transmittance and affecting the chemical stability in the electrolyte. Furthermore, polymer-based photoelectrode (PE) is frequently mixed with plastic-based CEs because combining polymer-based PEs with metal-based CEs may potentially reduce resistive losses due to highly conductive CE.[222] Hence, for flexible DSSC, a plastic substrate with TCFs is more typically used. The mesoporous PE of the solar cell requires a heat treatment higher than 450°C in conventional DSSC preparation to achieve good adhesion between the PE particle-particle and PE particle-polymer substrate as well as to remove organic surfactant in the PE paste or suspension. On the other hand, flexible plastic substrates (such as PET, PEN, PVDF, and PEEK) can only withstand temperatures below 150°C.[223] To overcome this limitation, several low-temperature fabrication techniques have been addressed.

The fabrication procedures that coat the conductive substrate influence the device's performance. The technology used should be determined by the device's size, form, flexibility, and other aspects such as energy requirement, production cost, and volume.[225] There are two types of fabrication techniques: physical and chemical deposition methods. The physical technique uses physical phenomena to manufacture and deposit materials, whereas the chemical method uses the chemical reaction of the substrate, precursor, and chemical medium. The physical fabrication techniques include doctor blade, screen printing, microwave sintering,[226] chemical sintering,[227] mechanical compressing,[228] pulsed laser deposition,[229] RF magnetron sputtering, hydrothermal necking, and ultrasonic spray-coating.[230,231] The chemical fabrication techniques mainly include electrophoretic/electrochemical deposition (EPD).[232,233] The common fabrication techniques among these techniques are doctor blades, screen printing, pulse laser deposition, electrospray deposition, and electrophoretic deposition. This is due to the cost-effectiveness, flexibility, and simplicity of depositions. In addition to the methods already mentioned, sintering methods like laser sintering and plate-fin heat sink-assisted elevated temperature sintering, as seen in Figure 13, can also be used without harming the plastic substrate. In the case of the latter, the ITO-PET substrate was still fine even after sintering at 250°C without thermal decomposition or loss of PET polymer integrity.[224] Furthermore, modifying the morphological characteristics and the interface of all functional layers, which has an impact on PV device performance, requires low-temperature operations that come into contact with active layers and flexible electrodes.[234,235] Researchers are particularly interested in flexible DSSCs because they provide enormous potential for cost-effective commercialization. As a result, many efforts were made to boost its potential. Despite this, no major gain in conversion performance has occurred, dampening researchers'

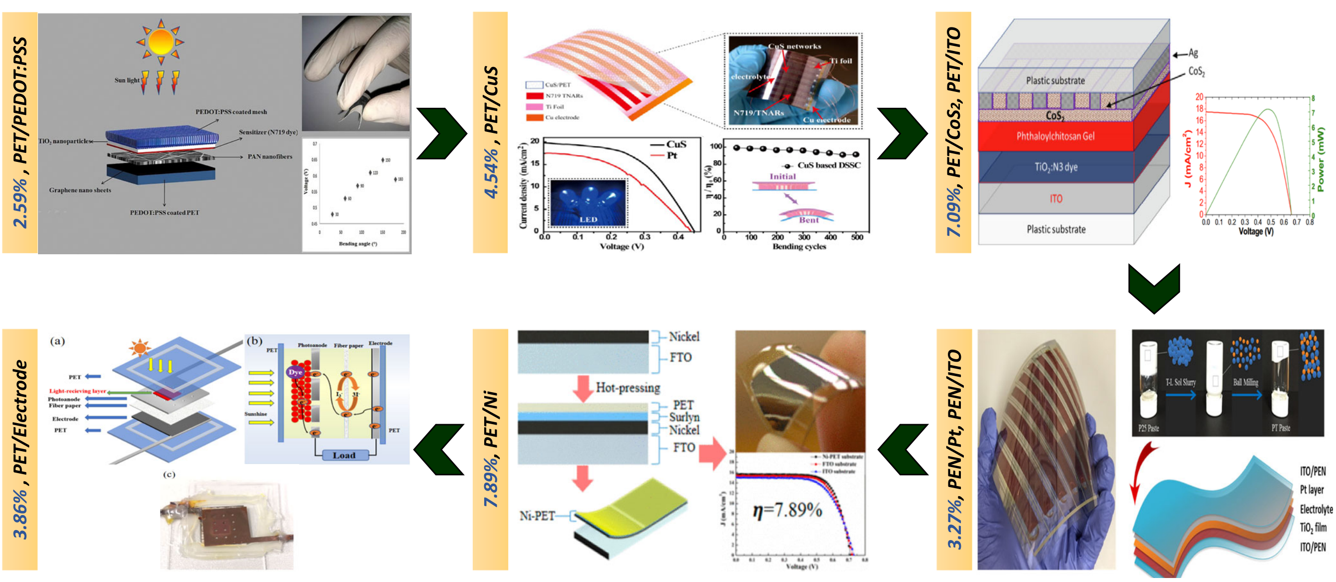

FIGURE 12 Flexible dye sensitized solar cells on polymer substrates. Reproduced with permission from the other studies.[211–216]

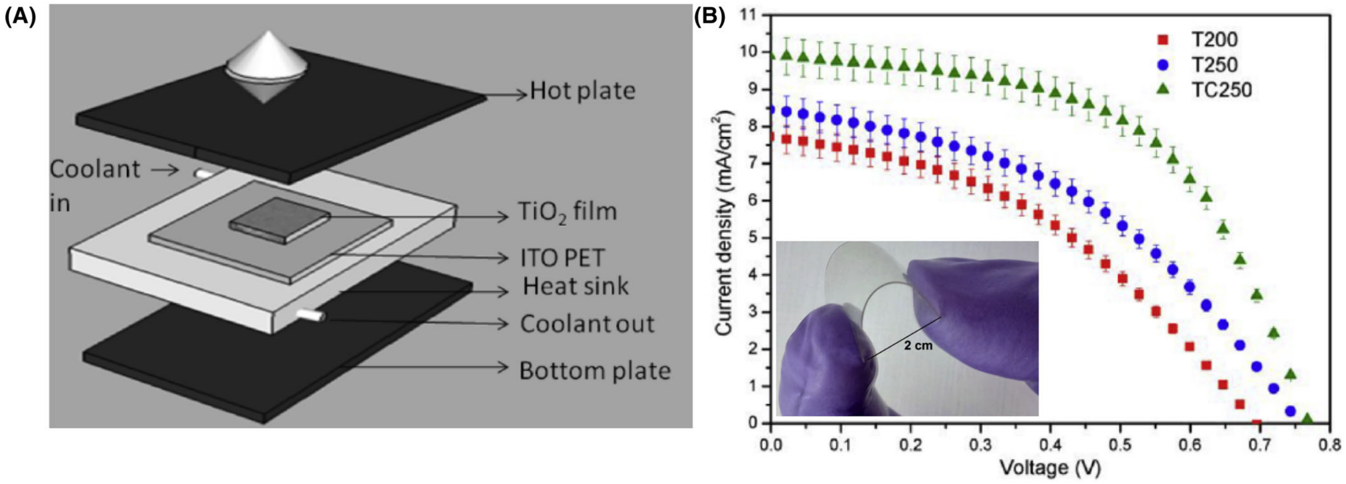

FIGURE 13 (A) Schematic illustration of heat sink assisted elevated temperature sintering of TiO2 coated onto the ITO PET polymer substrate (without applied load), (B) bare ITO PET film sintered at 250°C using heat sink. Photovoltaic J–V curves of flexible DSSCs fabricated using TiO2 and its composite films coated on to the ITO PET substrates sintered at various temperatures. Reproduced with permission from Baiju et al.[224] Copyright 2020, Elsevier.

enthusiasm for DSSCs. The low sintering temperature used in the fabrication of flexible DSSC photoanodes results in a slower electron transport rate that favors recombination reactions, a low charge collecting efficiency, and a higher inherent resistance, all of which reduce their efficiency. Flexible DSSCs receive minimal input relative to the effort put into DSSC research and development because of their poor conversion performance and ignorance of the possibilities of portable power supplies. Furthermore, flexible DSSCs have no actual conversion efficiency breakthrough, inadequate mechanical stability, and difficulty sealing while having a high theoretical conversion performance of about 32%.[236] Therefore, improving the characteristics of the sensitizers, substrate, redox electrolyte, and CE is another method for accelerating the application of flexible DSSC.[237] Over the years, several types of flexible DSSCs have been produced, either by introducing new materials to construct DSSC components or by investigating the specifications and operating circumstances when building solar devices. Table 5 summarizes the literature on several flexible DSSCs on polymer substrates.

Conducting plastic substrates such as ITO-PEI plastic substrate have higher thermal stability (i.e., it is capable of withstanding high sintering temperature) than PET and PEN. An optimal efficiency of 2.8% was achieved by fabricating a 100-nm layer deposited on a flexible PEI substrate. On the surface of PEI substrates, conductive oxide ITO was formed at various thicknesses using the RF magnetron sputtering process. The photoanode films were sintered at 200°C.[243] The utilization of high thermal stability polymer substrates has shown there is room to improve the performance of plastic-based flexible DSSC. In addition to polymer substrates, intensive research has been done to identify substitute materials for metal-based dyes, liquid electrolytes, metal CEs, and photoanodes to provide high-efficiency, low-cost devices with stable performance over time. To look into possible materials for efficient photoanodes, several metal oxides, including TiO_2, SnO_2, ZnO_2, In_2O_3, CeO_3, and NbO_3, have been used as photoanodes.[248] For DSSC experiments, metal-free organic photosensitizers based on coumarin, triarylamine, carbazole, indoline, and phenothiazine were used.[249–252] The leaking of liquid electrolytes is the cause of the stability problems for flexible DSSCs built on plastic substrates. To combat this, solid/gel electrolytes based on biopolymers, water-based synthetic polymers, and non-toxic solid/gel electrolytes like polyethylene oxide (PEO) and CNFs can be used in place of liquid electrolytes.[248,253–257] The high sunlight conversion efficiency of 7.03% was achieved using CNFs as electrolytes at simulated light intensities of 1.0. Additionally, after being subjected to harsh aging circumstances, the CNFs enhance the device's long-term stability and exceptional durability with over 95% retentive effectiveness.[258] To substitute platinum (Pt) CE for counter electrodes, carbon-based materials, various metal oxides, and polymer-based CE were also used.[259,260] Recently, Pervaiz et al. employed inorganic/organic composite ink as CE material for flexible DSSCs. The tetragonal $CuInS_2$ NPs and PEDOT:PSS polymer were used to synthesize the hybrid ink, which was then utilized to prepare an economically efficient CE on ITO-PET and cellulose paper substrates in flexible DSSCs using an ultrasonic spray deposition technique. Higher electrocatalytic performance has been shown in the inorganic/organic composite ink-based CEs.[238] Sabari Ginsun et al. employed molybdenum disulfide (MoS_2) as a CE for Pt-free DSSCs. MoS_2 electrode-based DSSC exhibits a higher photoconversion efficiency (7.96%) than platinum electrode-based DSSC (6.8%).[261] Fiber-shaped DSSCs are a possibility. Due to their easy preparation, light weight, and high wearability, fiber-shaped solar cells feature a one-dimensional linear structure as opposed to typical planar solar cells, making them a viable flexible power conversion technology for next-generation wearable electronics. The poor power conversion efficiency and flexibility of fiber-shaped DSSCs, however, are what essentially restrict their utilization. Therefore,

TABLE 5 Performance of different conductive polymer-based flexible dye-sensitized solar cells.

Year	Conductive substrate	Photoanode materials	Counter electrode material	Dye; electrolyte	Efficiency	Highlights	Ref.
2023	ITO-PET	TiO_2	$CuInS_2$/PEDOT:PSS/ITO-PET	N3 dye	3.99%	-	Pervaiz et al.[238]
2023	ITO-PET	TiO_2	PEDOT:PSS/ITO-PET	N3 dye	1.56%	-	Pervaiz et al.[238]
2023	ITO-PET	TiO_2	$CuInS_2$/PEDOT:PSS/cellulose paper	N3 dye	1.06%	-	Pervaiz et al.[238]
2022	ITO-PET	Bilayer TiO_2	Pt	N719 dye/Scattering layer; liquid	6.33%	-	Murali et al.[239]
2022	ITO-PET	TiO_2-P25	Pt/ITO-PET	N719; liquid	2.53%	The organic contamination of the TiO_2 surface was decreased by using UV-O_3 treatment.	Dawo et al.[240]
2022	ITO-PET	NiONP/Cdot	Pt-ITO/PET	N719 dye; electrolyte	4.43%	The substrate was a biodegradable, green cellulose nanofiber (CNF) sheet undergoing TEMPO oxidation.	Gemeda et al.[241]
	ITO/ODA-TOCNF	NiONP/Cdot	PPy/TOCNF		1.45%		
	NiO NW/TOCNF	NiONP/Cdot	Pt-TOCN		1.30%		
	ZnO NW/Cdot/TOCNF	ZnONP/Cdot	Pt-PPy/TOCNF		1.34%		
2021	ITO-PEI	TiO_2-P25	Pt/ITO-PEI	N3; gel	2.25%	The surface of the PEI (flexible substrate) was treated using plasma without chemical sintering.	Wante et al.[242]
2021	Ti foil	TiO_2	PEDOT:PSS/ITO-PET	N719; gel	1.7%	Spin coating	Wen et al.[236]
					1.33%	Screen printing	
2020	ITO-PET	TiO_2	Pt/ITO-PET	N719 dye	4.11%	-	Baiju et al.[224]
2020	ITO-PEI	TiO_2-P25	Pt/ITO-PEI	N3; iodide gel	2.8%	-	Wante et al.[243]
2020	FTO-Glass	TiO_2	Nanodiamonds/Zn/ITO-PET	N3-N719; liquid	6.23%	-	Fayaz et al.[244]
2020	FTO-PET	TiO_2-P25	MoS_2/FTO-PET	N719-BVIml; liquid	4.84%	-	Gurulakshmi et al.[245]
2020	ITO-PEN	TiO_2-TG LSL	Pt/ITO-PEN	N719; liquid	5.18%	-	Mustafa and Sulaiman[246]
2019	ITO-PET	TiO_2	CoS_2/Ag-PET	N3; phthaloyl chitosan gel	7.09%	-	Prasad et al.[213]
			Pt/ITO-PET		7.04%		
2019	ITO-PET	TiO_2	Carbon nanotubes + graphite + conductive carbon black + graphene/ITO-PET	N719 dye	4.32%	-	
2018	Cu-Ti foil	TiO_2 nanotube arrays	CuS/PET	N719; electrolyte	4.54%	CuS film served as conducting films (replacing ITO) and as superior catalysts for flexible DSSCs.	Xu et al.[212]
			Pt/PET		3.24%		

(Continues)

TABLE 5 (Continued)

Year	Conductive substrate	Photoanode materials	Counter electrode material	Dye; electrolyte	Efficiency	Highlights	Ref.
2018	Ag-Kapton	TiO_2	Pt/FTO glass	N719; liquid (Iodide)	7.03%	The Kapton-based devices showed good printed layer quality and uniformity due to the polyimide Kapton film's high chemical and temperature resistance.	Liu et al.[247]
2017	PET/PA/PEDOT:PSS	TiO_2	rGO/PEDOT:PSS-PET	N719; quasi-solid electrolyte (PAN)	2.59%	High flexibility	Berendjchi et al.[211]

introducing emerging materials and fabricating on polymer substrates improves its performance and stability along with its flexible nature.

5.2 | Flexible PSCs

In the history of PVs, no solar cell like the PSC has achieved an amazing power conversion efficiency exceeding 26% in such a short period. The abundance of precursor materials, which makes PSCs ideal for mass production, is a significant advantage they have over other thin-film solar cells. Emerging technologies include organic photovoltaics (OPVs), DSSCs, and quantum dot solar cells (QDSCs), all of which are hindered from being used commercially by either difficult fabrication procedures or poor power conversion efficiency. Thus, PSCs could become an encouraging candidate for the generation of emerging solar cell technologies in terms of manufacturing cost and high device efficiency.[262] PSCs have emerged from solid-state DSSCs, the DSSCs are very much restricted, and perovskite solid-state absorbers can increase efficiency. Perovskite has certain advantageous properties, including flexibility, lightness, and semitransparency. Therefore, due to its solid-state nature and low-temperature manufacturing methods, the perovskite PV platform offers a perfect path for fabricating flexible devices. Figure 14 represents various flexible PSCs fabricated on polymer substrates over the years.

An efficient PSC structure is a sandwich structure made up of an active layer (perovskite) stacked between ultrathin carrier transport materials with electrodes. The perovskite structure resembles the molecular structure of calcium titanate ($CaTiO_3$) having an ABX_3 chemical structure.[269] In the composition of perovskite, A corresponds to a group of monovalent cations, B corresponds to a metal ion, and X corresponds to the halide ion.[270] Hereby, the discovered perovskites are bright in color and have good conductivity.[271–273] The ultra-thin layers between which the perovskite is stacked have two different electron affinities and ionization potentials. The layer with low electron affinity and ionization potential serves as hole transporting materials (HTLs), while the layer with high electron affinity and ionization act as electron transporting material.[274–276] The HTL and ETL layer works as an electron-blocking and hole-blocking layer that prevents electron flow and release of holes to the front electrode.[277] The typical examples of hole-transporting layers are Spiro-OMeTAD, P3HT, CuI, NiO, CuSCN, and 2D materials such as MoS_2.[278–280] While examples of electron-transporting layers are ZnO, TiO_2, SnO_2, IGZO, PCBM, graphene oxide, and so on.[281,282] These charge carrier transport layers are deposited on electrodes (anode and cathode) based on the device structure. A PSC exists in two diverse categories, including p-i-n junction for HTL/perovskite/ETL type and n-i-p junction for ETL/Perovskite/HTL type. In the n-i-p configuration, the light passes through the TCF (electrode) in front of the ETL while the p-i-n configuration has the opposite arrangement. The most common structure used in PVs is the n-i-p type due to high optical transmittance through substrates and reports higher efficiency than the p-i-n type.[283] Fabricating the different perovskite solar structures on flexible substrates gives a flexible device that can be used in various portable and

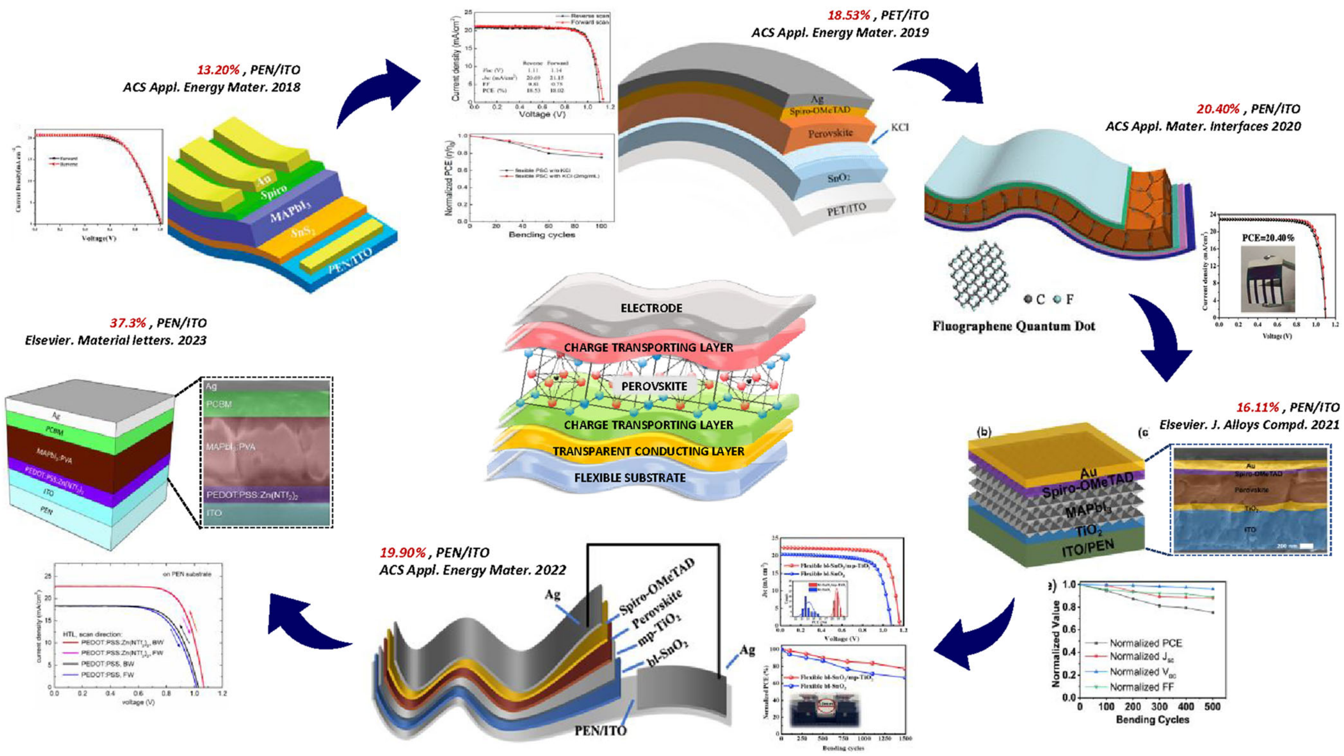

FIGURE 14 Polymer substrate-based flexible perovskite solar cells. Reproduced with permission from other studies.[263–268]

wearable applications. The various fabrication techniques for producing PSCs include solution-based processes that are compatible with substrates such as spin coating, spray coating, doctor-blading, inkjet printing, and R2R processing.[284] Table 6 summarizes recent perovskite PV devices on polymer substrates.

Since PSCs can be fabricated in low temperatures, polymer substrates are suitable for building up a device with ease of fabrication resulting in light weight, low cost, and higher efficiency. The most common fabrication techniques are doctor blading, slot-die coating, screen printing, inkjet printing, spray coating, and R2R coating. Using specific fabrication techniques, various PSCs are fabricated at low temperatures upon a polymer substrate. Due to its flexibility, portability, low weight, and ease of integration over small, big, and curved surfaces, developing polymer substrate flexible PSCs technology can be seen as a viable and intriguing field from an application point of view in this situation.

6 | SUMMARY AND PERSPECTIVE

Flexibility would be especially important for next-generation PV devices, which have low material requirements for photoactive layers and can be manufactured using R2R and other cutting-edge techniques. The essential components for creating flexible solar cells, such as substrates and TCFs, are thoroughly reviewed. Metals, glass, paper, and plastics are among the substrate materials discussed in this article. Although flexible substrate technology has advanced, a new and/or better substrate is still required to meet the demands of flexibility and affordable goods. The difficulty of manufacturing a flexible glass substrate on a big scale and at a reasonable cost and the opaque nature of metal substrates limits their application. Further polymer substrates solve the issue and exhibit high optical transmittance with low-cost manufacturing. Various polymer materials that can serve as substrates are discussed in this review. Among the polymers, PET and PEN are commercially used as flexible substrates due to high optical transmittance and mechanical flexibility but deform highly at high temperatures. Thus, low fabrication methods are employed. For high-temperature fabrication, polymers such as PES, PEI, and PI can be used. Polymers such as PEEK and PVDF are efficient substrates that combine high mechanical strength with good processability. The polymer substrates are further coated with transparent conducting materials for charge collection. A thorough summary of current advancements and advances in TCEs is provided in this article. Various transparent conducting materials that can be fabricated on polymer substrates are reviewed. The standard ITO is important in many optoelectronic applications, but because it is brittle and there is a shortage of indium, alternative TCEs are required to increase performance in terms of stretchability and flexibility and to address the issue of indium scarcity. ZnO is readily accessible and doped, as was previously mentioned. The most often used TCO for ITO replacement is ZnO. ZnO:Al is less adaptable when used alone and exhibits poor stability in oxidizing situations as well as etchability when these materials come into contact with acids. Another strong contender is carbon nanomaterials, which has excellent mechanical and electrical

TABLE 6 Recently developed polymer-based flexible perovskite solar cells.

Year	Substrate	Cell architecture	Efficiency	Reference
2023	PEN	PEN/ITO/PEDOT:PSS:Zn $(NTf_2)_2$/$MAPbI_3$:PVA/PCBM/Ag	37.3%	Chen et al.[268]
2022	PET	PET/IZO/PTAA/2D BOC Spacer (FPEAI)/$CsFAPbBrI_3$/C_{60}/BCP/Ag	18.66%	Dasgupta et al.[285]
2022	PET	PET/ITO/SnOx/PNPs@NiOx@PAM/Spiro-OMeTAD/Au	14.66%	Zhu et al.[286]
2022	PI	PI@GR/PEDOT:PSS/$MAPbI_3$/$FAPbI_3$/C_{60}/BCP/PI@GR	15.1%	Jeong et al.[287]
2022	PET	PET/ITO/MB-NiO/C_{60}/ALD-SnO2/Au/PEDOT:PSS/NBG perovskite/C_{60}/BCP/Cu	25%	Li et al.[288]
2022	PET	PET/ITO/PTAA/BAFACs/PCBM/BCP/Ag	11.96%	Lan et al.[289]
		PET/IZO/PTAA/BAFACs/PCBM/BCP/Ag	12.78%	
		PET/IZVO/PTAA/BAFACs/PCBM/BCP/Ag	14.57%	
2022	PET	PET/ITO/SnO_2/perovskite/spiro-OMeTAD/Au	11.4%	Dkhili et al.[290]
		PET/ITO/ZnO/SnO_2/perovskite/spiro-OMeTAD/Au	14.9%	
2022	PET	PET/PH1000/Al 4083/$MAPbI_3$/PCBM/Ag	9.49%	Xhie et al.[201]
		PET/ITO/Al 4083/$MAPbI_3$/PCBM/Ag	10.10%	
2022	PEN	PEN/ITO/bl-SnO_2/mp-TiO_2/perovskite/Spiro-OMeTAD/Ag	19.90%	Gu et al.[267]
2022	PET	PET/ITO/SnO_2/$MAPbI_3$/Spiro-OMeTAD/Au	14.90%	Sun et al.[291]
2021	PEN	PEN/ITO/TiO_2/$MAPbI_3$/Spiro-OMeTAD/Au	16.11%	Yang et al.[266]
2021	PET	PET/ITO/SnO_2/perovskite/spiro-OMeTAD/Au	12%	Taheri et al.[292]
2021	PET	PET/ITO/PEDOT:GO/$MAPbI_3$/$PC_{61}BM$/Ag	10.26%	Xue et al.[293]
2021	PEN	PEN/ITO/c-TiO_2/mp-TiO_2/$FAPbI_3$/spiro/Ag	14.52%	Yin et al.[294]
2021	PET	PET/ITO/nano-TiO_2/$MAPbI_3$/spiro/Au	13.33%	Wu et al.[295]
2021	PET	PET/IZO/PTAA/PVK/C_{60}/SnO_x/ITO/Ag/ITO	9.02%	Spinelli et al.[296]
2020	PI	PI/Cu grid/graphene/PEDOT:PSS/perovskite/PC61BM/ZnO/Ag	16.4%	Jeong et al.[181]
2020	PEN	PEN/ITO/SnO2/perovskite/Spiro-OMeTAD/Ag	16.08%	Huang et al.[297]
2020	PET	PET/IZO/PTAA/perovskite/PCBM:PMMA/Cr/carbon	15.18%	Babu et al.[298]
		PET/IZO/PTAA/perovskite/PCBM:PMMA/Cr/silver	15.71%	
2020	PEN	PEN/ITO/Au-TiO_2/TiO_2/FAPbI3/spiro/Ag	15.36%	Zhao et al.[299]
2020	PET	PET/ITO/R-Fu/Lt-TiO_2/FAMAPb $(IBr)_3$/spiro/Ag	18.06%	Wang et al.[300]
2020	PET	PET/ITO/NiO_x/perovskite/$PC_{61}BM$/BCP/Ag	18.0%	Wang et al.[301]
2020	PEN	PEN/ITO/PEALD TiOx/RIE-treated mp-TiO_2/perovskite/Spiro-MeOTAD/Au	17.29%	Kim et al.[302]
2020	PET	PET/Cu HC:dd-PH1000/Cu:NiO_x/$MAPbI_3$/PCBM/BCP/Cu	13.58%	Li et al.[182]
2020	PEN	PEN/ITO/modified SnO2/perovskite/spiro-OMeTAD/Ag	20.40%	Yang et al.[265]
2020	PEN	PEN/ITO/TiO_2/PCBM/Gual treated perovskite/Spiro-OMeTAD/Au	17.0%	Li et al.[303]
2019	PET	PET/ITO/C_{60}/TiO_x/FAMAPb $(IBr)_3$/spiro/Au	14.43%	Liu et al.[304]
2019	PET	ITO/PET/SnO_2/KCl/$MAPbI_3$/spiro-OMeTAD/Ag	18.53%	Zhu et al.[264]
2018	PEN	PEN/ITO/TiO_2/FAMAPb $(IBr)_3$/PTAA/Au	17.1%	Wilkes et al.[305]
2018	PEN	PEN/ITO/SnS_2/$MAPbI_3$/Spiro/Au	13.20%	Chu et al.[263]
2018	PET	PET/ITO/SnO_2/TiO_2/$FAPbI_3$/spiro/Au	14.8%	Dagar et al.[306]
2018	PES	Au/Spiro/$MAPbI_3$/ZnO/AZO/AgNW/AZO/PES	11.23%	Lee et al.[307]
2017	PEN	PEN/ITO/ED-TiO_2/BK-TiO_2/$MAPbI_3$/spiro/Au	15.76%	Lin et al.[308]
2017	PEN	PEN/Graphene/MoO_3/PEDOT:PSS/$MAPbI_3$/C_{60}/BCP/LiF/Al	16.8%	Yoon et al.[198]

characteristics as well as great flexibility and functional efficiency. However, it exhibits high sheet resistance and high roughness, which reduce carrier movement and reduce device lifespan. To take its rightful place as a superior TCF, carbon materials still require considerable refinement. A better alternative to ITO for TCE is graphene, which has a high degree of flexibility, conductivity, transparency, and ease of production. However, more efforts like mass manufacturing and a simpler synthesis procedure are needed to make it the top choice for use as TCEs in devices by the light and display sectors. Ag NW is the primary material for usage as a conducting substance in metal nanowires. Ag NWs may be a superior TCE depending on the sheet resistance, transparency, and flexibility. Ag NW films still have a number of shortcomings, including a high degree of surface roughness, high junction resistance, a tiny contact area, and poor chemical and mechanical endurance. To employ Ag NW as a more effective substitute for ITO, they need to be improved. However, because to its poor durability at high temperatures and an uneven combination of sheet resistance and transparency, PEDOT:PSS, a conducting polymer, is less ideal for TCE applications. Further, we examined dye-sensitizer and perovskites as active materials. In the near future, perovskites and dye sensitizers will rule the market. Perovskite and DSSCs are anticipated to experience an acceleration in commercial development due to their high efficiency and low-temperature manufacturing. At last, a literature review of conducting polymer substrate-based perovskite and DSSCs is presented. The application of polymer substrates enhances the performance of emerging solar cells along with their flexibility. The flexible solar modules giving higher performance can further be integrated into many curved devices making it portable and wearable.

AUTHOR CONTRIBUTIONS

Poonam Subudhi: Investigation; writing—original draft preparation; writing—reviewing and editing. **Deepak Punetha:** Supervision; writing—reviewing and editing.

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CONFLICT OF INTEREST STATEMENT

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

DATA AVAILABILITY STATEMENT

No data were used for the research described in the article.

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