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REVIEW

Foldable solar cells: Structure design and flexible materials

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Abstract

Foldable solar cells, with the advantages of size compactness and shape transformation, have promising applications as power sources in wearable and portable electronics, building and vehicle integrated photovoltaics. However, in contrast to mild bending with curvature radius of several millimeters, folding generates the crease with extreme curvature radius of sub-millimeter, resulting in the appearance of large strain and stress. As a result, it is highly challenging to realize robustly foldable and highly efficient solar cells. Here, we summarize the recent progress on the photovoltaic performance and mechanical robustness of foldable solar cells. The key requirements to construct highly foldable solar cells, including structure design based on tuning the neutral axis plane, and adopting flexible alternatives including substrates, transparent electrodes and absorbers, are intensively discussed. In the end, some perspectives for the future development of foldable solar cells, especially the standard folding procedure, improvement in the folding endurance through revealing failure mechanism, are provided.

KEYWORDS
ductile absorber, flexible transparent electrodes, foldable solar cells, structure design, ultrathin substrates

1 | INTRODUCTION

Solar energy is considered to be one of the competitive alternatives to fossil fuels in the future due to its abundance, cleanness, and sustainability.[1,2] Solar energy can be utilized in many ways, among which the solar cell that converts sunlight into electricity is the most convenient route. Recently, flexible solar cells, with the advantages of low cost, light weight, foldability, roll-to-roll fabrication, have attracted wide attention. The deformation of

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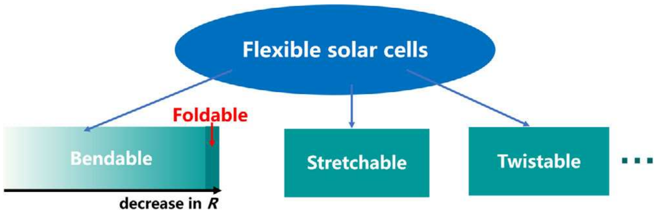

FIGURE 1 Schematic illustration of the deformation of flexible solar cells

flexible solar cells mainly includes bending, folding, stretching, twisting and crumpling (Figure 1). It is widely accepted that folding is the extreme condition of bending which generating crease with extreme low curvature radius of sub-millimeter. Thus, foldable solar cells meet the requirements of size compactness and shape transformation for many emerging applications. They can not only be used as electrical power generating in wearable and portable electronics to form self-powered systems, but also can be integrated into the complex-shaped building and transportation vehicle to offer integrated functionality.

Recently, flexible solar cells have experienced fast progress in respect of the photovoltaic performance, while the attention on the mechanical stability is limited.[3–10] By now, most reported flexible solar cells can only tolerate bending with curvature radius of several millimeters. The investigation on foldable solar cells is only a few. It is well known that folding induces the crease with a curvature radius of sub-millimeter, resulting in the appearance of large strain and stress. As a result, cracks will be formed in the functional layers or delamination will be occurred at the interface as soon as the strain exceeds the crack onset strain, leading to the degradation or even failure of the solar cells under repeated folding. Therefore, it is highly challenging to realize robustly foldable solar cells. Moreover, the performance of foldable solar cells on foldable conductive substrates has not been optimized, which is far behind the normal bendable solar cells, not to mention the rigid ones. Thus, it is urgent to improve both the photovoltaic performance and mechanical stability of foldable solar cells for the further practical applications.

Here, we summarize the recent progress on photovoltaic performance and mechanical robustness of foldable solar cells. Then, we discuss the key requirements to construct highly foldable solar cells, including structure design based on tuning the neutral axis plane, and flexible alternatives including substrates, transparent electrodes, and absorbers (Figure 2). In the end, we discuss the promises of foldable solar cells and provide some perspectives for the future development. We hope this review will shed light on the design and fabrication of foldable solar cells for many emerging applications.

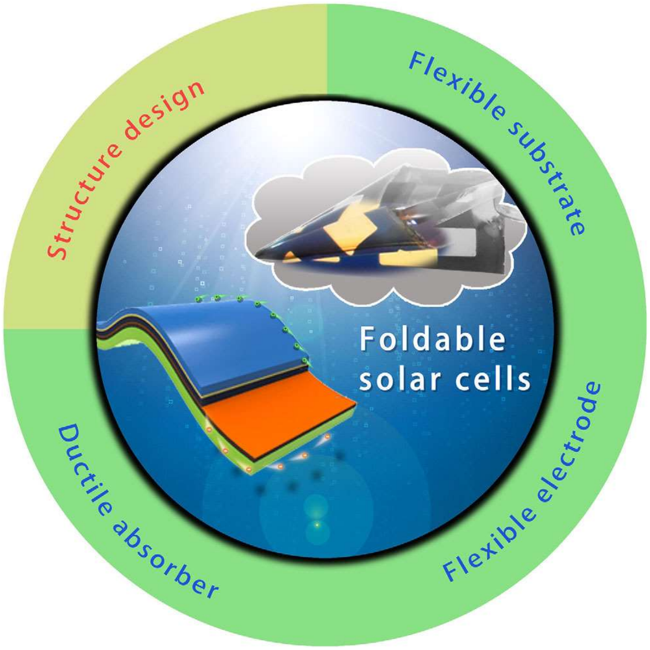

FIGURE 2 Key points for achieving highly foldable solar cells

2 | STATUS OF FOLDABLE SOLAR CELLS

Compared to the normal bendable solar cells which can endure flexion with a smooth curve with radius of several millimeters, foldable solar cells can tolerate the crease at the edge with a curvature radius of sub-millimeter. It is apparent that foldable solar cells experience large strain or stress, resulting that it is difficult to realize highly foldable solar cells. In addition, compared to the normal bending procedure which can be precisely determined by the curvature radius and cycle, there is a lack of the accurate definition of folding procedure by now. In some papers, the authors fold the devices around cylinder with fixed curvature radius of sub-millimeter.[11,12] In the other papers, the authors directly fold the devices with or without force, while no accurate characterization on curvature radius is given.[13–15] In the latter condition, the folding radius which strongly depending on the sample thickness and imposed force, differs from samples to samples. In this paper, we make the following definition that the devices possess foldability if they can endure flexion with curvature radius of sub-millimeter, direct folding, or crumpling, which will be included in the following discussion. Based on our understanding, the reported foldable solar cells can be mainly divided into the following two types.

On one hand, folding is done in the predesigned place which can endure large strain and stress, such as the flexible substrates or flexible transparent electrodes.[16–18] Nogi et al. demonstrated foldable organic solar cells using nanofiber paper as substrates and silver nanowire as

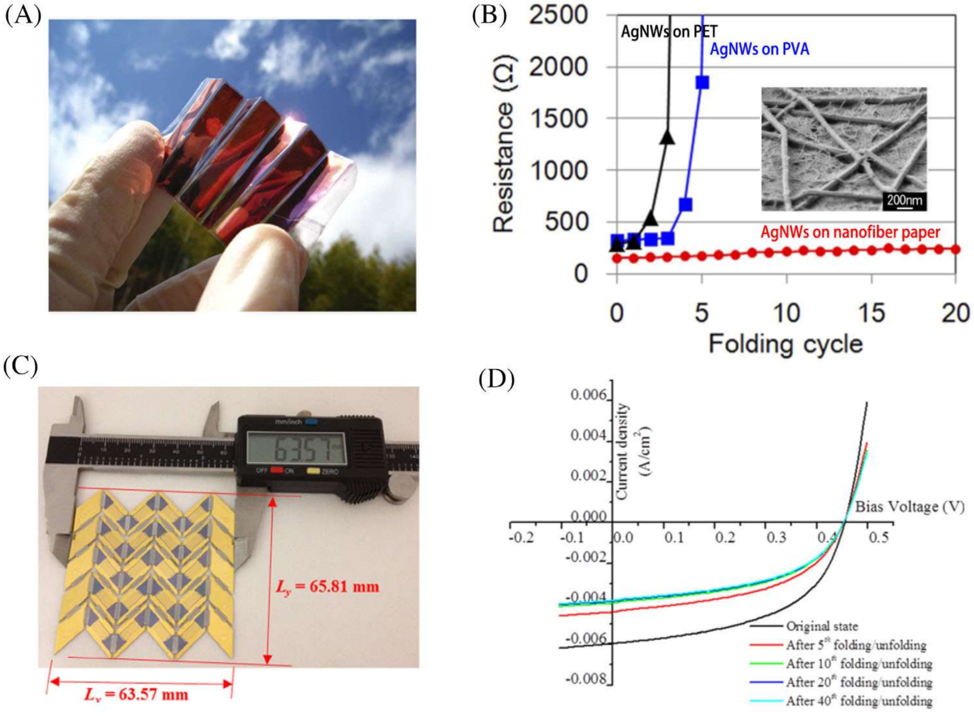

FIGURE 3 Foldable solar cells with crease in the predesigned place. A, Organic solar cells folding in the transparent conductive nanofiber paper. B, Electrical resistance of AgNWs on different substrates as a function of folding cycle, the inset is the SEM image of AgNWs on nanofiber paper.[16] Copyright 2015 Springer Nature. C, Foldable c-Si solar modules enabled by origami design. D, *J-V* curves of origami c-Si solar modules after cyclic folding.[17] Copyright 2014 American Institute of Physics

electrodes with power conversion efficiency (PCE) of 3.2%. However, the folding was done in the transparent conductive nanofiber paper instead of organic absorber layers, as shown in Figure 3A. It was proved that silver nanowires on nanofiber paper exhibited high electrical stability in repeated folding tests due to the dual advantages of hydrophilic affinity and entanglement between cellulose nanofibers and silver nanowires, as shown in Figure 3B, imparting the foldability of paper-based organic solar cells.[16] Yu et al. realized foldable crystalline silicon (c-Si) solar modules enabled by origami structure. In this approach, high-performance devices placed in the parallelograms did not experience large strain during deformation, while the serpentine-shaped metal interconnects placed at the creases suffered large strain during folding and unfolding, as shown in Figure 3C. At the original state, the c-Si solar modules exhibited open circuit voltage (V_{oc}) of 0.455 V, short circuit current density (J_{sc}) of 5.95 mA cm^{-2}, and filling factor of 52.1%. When the solar modules subjected to folding, the J_{sc} started to decrease and gradually saturated at around 4 mA cm^{-2} after 10 cycles of folding/unfolding, while the V_{oc} almost remained constant throughout 40 times folding/unfolding, as shown in Figure 3D.[17]

On the other hand, the researchers try to fabricate real foldable solar cells in which creases locate in the active layers.[11–15,19–22] Due to its low-cost, lightweight, foldable, degradable and recyclable nature, paper is a promising substrate for foldable solar cells. B. A. Korgel et al. reported foldable CuInSe$_2$ nanocrystal solar cells on paper substrates composed of bacterial nanocellulose, as shown in Figure 4A. The inorganic solar cells exhibited PCE as high as 2.25%. More importantly, the performance of solar cells was stable even after five cycles of folding on the active layers. While further folding would lead to the failure due to the formation of large cracks in indium tin oxides (ITO) and nanocrystal CuInSe$_2$ layers.[13] Song et al. applied the strategies of using ultrathin substrate to decrease stress in the functional layers during folding, combined with using ultrathin silver films to replace brittle ITO electrodes, to construct robustly foldable solar cells. Polymer solar cells using 25 μm cellophane paper as substrates and zinc oxide/ultrathin Ag/zinc oxide (OMO) as electrodes exhibited PCE of 5.94%. Furthermore, the polymer solar cells maintained 92% of initial PCE after folding at 180° angle for 35 cycles, as shown in Figure 4B.[14] Using the same strategies, they demonstrated perovskite solar cells on 25 μm cellophane paper substrates and titanium oxide/ultrathin Ag/titanium oxide electrodes. The solar cells exhibited PCE of 13.19%, the highest among all the paper-based solar cells. Moreover, perovskite solar cells retained 97.6% of the initial PCE after bending with a radius of 0.3 mm and even preserved 95.8% of the initial

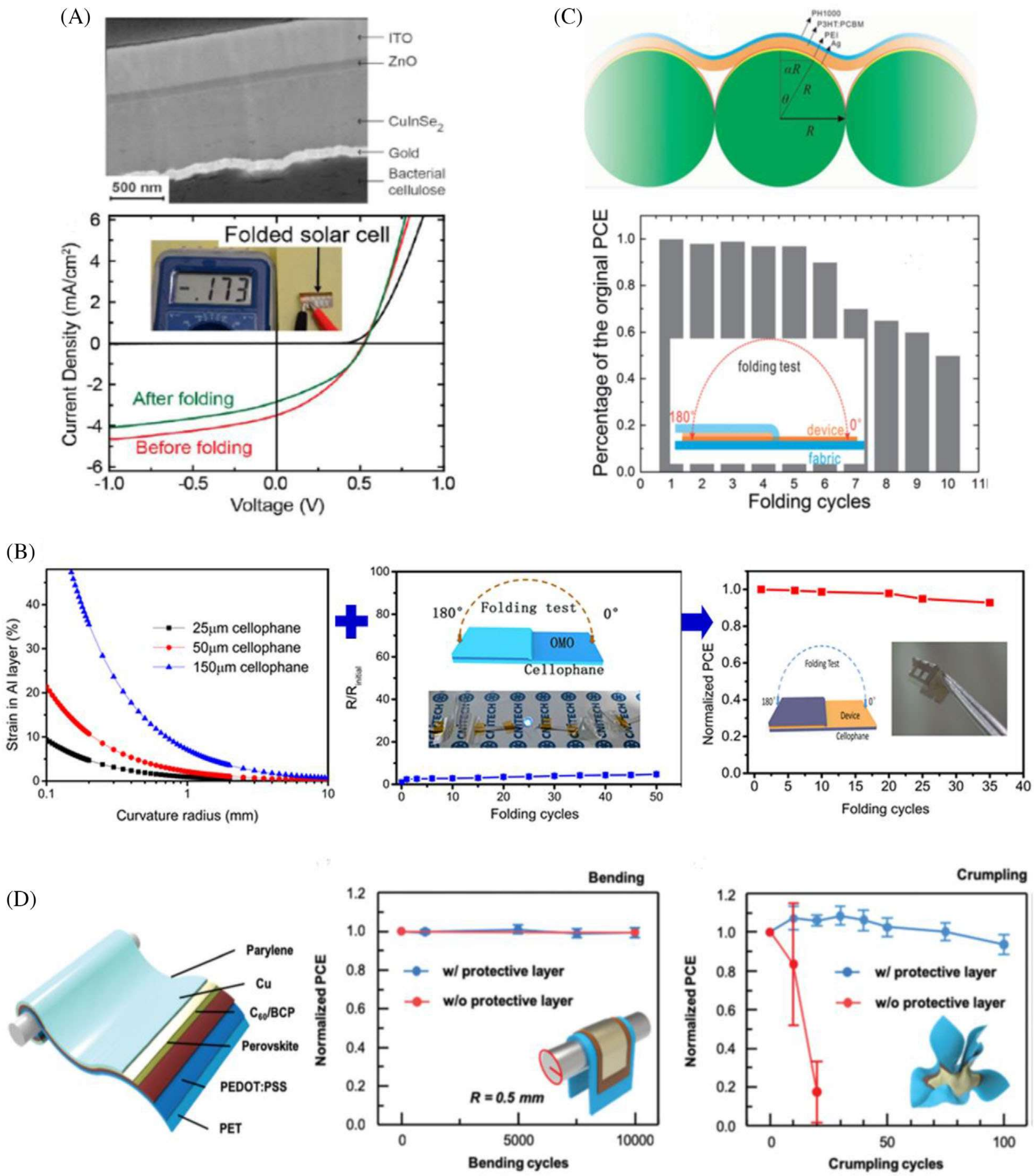

FIGURE 4 Foldable solar cells with crease in the active layers. A, Foldable CuInSe$_2$ solar cells on bacterial cellulose paper.[13] Copyright 2017 American Chemical Society. B, Foldable polymer solar cells using 25 μm cellophane paper substrates and OMO electrodes.[14] Copyright 2018 John Wiley and Sons. C, Foldable polymer solar cells on woven fabric electrodes.[15] Copyright 2017 Royal Society of Chemistry. D, Foldable perovskite solar cells on 2.5 μm PET substrates.[11] Copyright 2019 Royal Society of Chemistry

PCE after bending with a radius of 1mm for 1000 cycles. They could further preserve >80% and >55% of initial value after 50 single folding and 10 dual folding cycles. The difference in performance degradation between single and dual folding was tentatively explained by the different crack morphologies.[23,19] Woven fabric is another popular substrate for foldable solar cells in wearable applications. Zhen et al. prepared foldable polymer solar cells on woven fabric using a free-standing and wet transfer method. The fabric-based solar cells exhibited PCE of 2.90%. Moreover, they

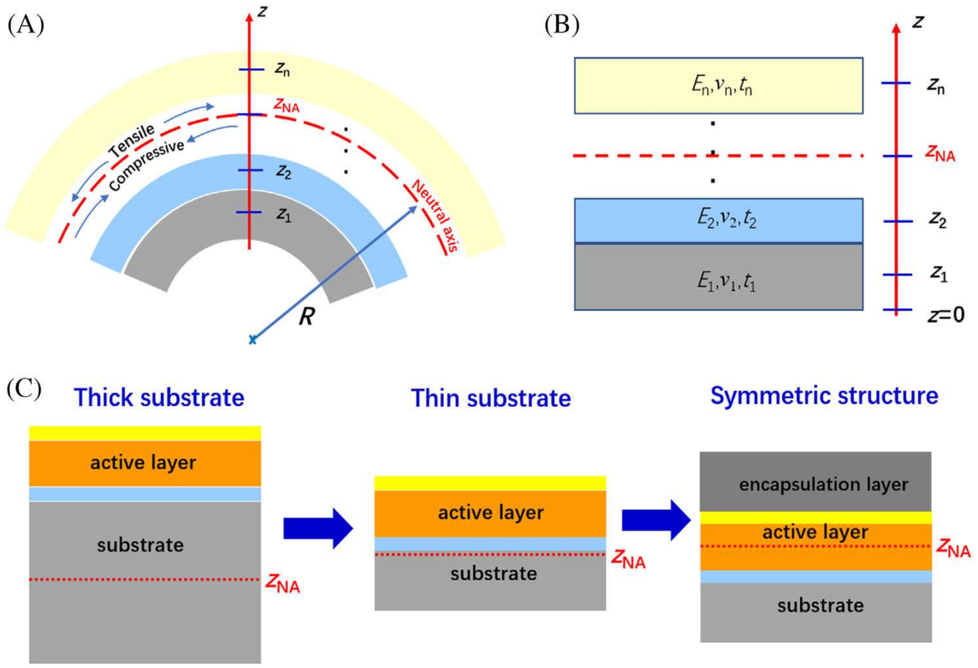

FIGURE 5 Schematic illustration of (A) multilayers flexion with a curvature radius R, (B) structure consisting of n layers, and (C) dependence of z_{NA} position on the device structure

maintained 50% of initial value after folding in half for 10 cycles, supposed to be due to the low Young's modulus of woven fabrics, as shown in Figure 4C.[15] Besides paper and woven fabric, the normally used polymer substrates can also be applied as the substrates for foldable solar cells. Kaltenbrunner et al. demonstrated ultrathin perovskite solar cells on 1.4 μm PET substrates, which exhibited stabilized efficiency of 12% and a power-per-weight as high as 23 W g^{-1}. Furthermore, they can endure multiple bends into curvature radii down to 10 μm. Herein, the flexion radius was determined from the three-dimensional map of the wrinkle morphology.[21] Lee et al. demonstrated ultra-flexible perovskite solar cells on thin PET substrates. The solar cells on 2.5 μm PET substrate exhibited PCE of 17.03%. Moreover, they underwent negligible performance degradation after bending with radius of 0.5 mm for 10,000 cycles. Further applying the protective layer on top of device can improve the crumpling endurance. The solar cells on 2.5 μm PET with protective layer maintained their initial performance as much as 88% after 100 cycles of crumpling, as shown in Figure 4D.[11] Park et al. prepared flexible perovskite solar cells using shape recoverable polymer NOA 63 substrates and PEDOT:PSS electrodes with initial PCE of 10.75%. The devices maintained ~90% of initial performance after bending with radius of 1 mm for 1000 cycles. Furthermore, they still worked after crumpling, with the PCE decreasing from 10.75% to 6.07%.[22]

The PCE and foldability of reported solar cells are summarized in Table 1. It is apparent that foldable solar cells can be realized through constructing appropriate device structure combined with using flexible alternatives, which will be intensively introduced in the following paragraphs. While, the PCE of foldable solar cells is low compared to that of the rigid ones, mainly ascribed to that the properties of absorbers on ultrathin conductive substrates have not been optimized. Considering the scientific and industrial importance of realizing the foldable solar cells with crease directly in the active layers, we will focus on constructing this type of foldable solar cells in the following.

3 | STRUCTURE DESIGN FOR FOLDABLE SOLAR CELLS

The challenge for realizing the foldable solar cell is mainly ascribed to the large stress in the devices under folding, thus the strategies of adjusting the strain and stress in device is an effective way. For the solar cells consisting of multilayers, the induced strain in devices under flexion with curvature radius of R can be expressed as Equation (1). Fortunately, the strain in device is not only influenced by flexion radius, but also by the position of neutral axis (z_{NA}) plane where no strain existed, as shown in Figure 5A. When $z > z_{NA}$, the materials experience the tensile strain; and when $z < z_{NA}$, the materials experience the compressive strain. Thus, tuning the z_{NA} plane is a useful solution to decrease the strain/stress in device during folding. The position of z_{NA} plane in the multilayers can be determined

TABLE 1 Summarize for the performance of representative foldable solar cells

Solar cells/modules	Conductive substrates	Initial PCE	Folding condition	Remaining PCE of initial value
Folding with crease in the predesigned place				
Organic solar cells [16]	AgNWs/ paper	3.2%	Folding with crease in conductive paper	-
Origami-based c-Si solar modules [17]	Metal/polymer as interconnects	1.41%	Folding for 40 cycles	~60%
Folding with crease in the active layers				
$CuInSe_2$ solar cells [13]	ITO/paper	2.25%	Folding for 5 cycles	>80%
Organic solar cells [12]	PEDOT:PSS /PET	14.20%	Folding with radius of less than 1 mm for 1000 cycles	68%
Polymer solar cells [14]	OMO/25 μm paper	5.94%	Folding for 35 cycles with 180° angle	92%
Polymer solar cells [15]	PH1000/woven fabric	2.90%	Folding for 10 cycles with 180° angle	50%
Perovskite solar cells [11]	PEDOT:PSS/2.5 μm PET	17.03%	Flexion with radius of 0.5 mm for 10000 cycles/Crumpling for 100 cycles	>90%
Perovskite solar cells [19]	OMO/25 μm paper	13.19%	Folding for 50 cycles with 180° angle	>80%
Perovskite solar cells [21]	PEDOT:PSS/1.4 μm PET	12%	Flexion with curvature radius of 10 μm	-
Perovskite solar cells [22]	PEDOT:PSS /NOA 63	10.75%	Crumpling	~60%

by Equation (2).[24,25]

$$\varepsilon(z, R) = \frac{z - z_{\text{NA}}}{R} \tag{1}$$

$$z_{\text{NA}} = \frac{\sum_{k=1}^{n} E_k^* t_k z_k}{\sum_{k=1}^{n} E_k^* t_k} \tag{2}$$

where $E_k^* = \frac{E_k}{1-\nu_k^2}$, E_k, ν_k and t_k are Young's modulus, poisson's ratio, thickness of each layer k, z_k is the middle position of each layer on z axis, as shown in Figure 5B.

The dependence of z_{NA} position on the device structure is schematically demonstrated in Figure 5C. When the solar cells are prepared on thick substrate with thickness more than 100 µm, considering the thickness of total active layer is usually lower than 1 µm and the nearly the similar magnitude of Young's modulus value for substrate and active layer, the z_{NA} plane roughly locates in the middle of substrates, as determined from Equation (2). If decreasing the substrate thickness by an order of magnitude or even more, the z_{NA} plane will shift towards the active layers of solar cells. Further applying symmetric device structure, such as coating encapsulation layer on the top of devices, the z_{NA} plane will even enter into the active layers of solar cells. As a result, the strain and stress in devices during folding will be significantly decreased by controlling the z_{NA} plane, leading to the realization of robustly foldable solar cells.

A few investigations preliminarily demonstrated that the strategies of using ultrathin substrate and/or symmetric structure could mitigate strain/stress in device, which could be favorable to improve foldability of various optoelectronic devices. Lee et al. observed that organic flash memories on 6 µm ultrathin substrates maintained initial capacities after folding with a curvature radius of 300 µm for 1200 cycles.[26] Sekitani et al. demonstrated that organic transistors could continue to operate while being folded into a radius of 100 µm, enabled by thin substrate thickness of 12.5 µm and parylene/TFT/PI structure.[27] Kim et al. revealed that silicon integrated circuits exhibited good electrical properties when folded with a radius of 50 µm. This extreme flexibility was ascribed to 1.2 µm ultrathin substrate and PI/CMOS/PI structure.[28] Recently, the above-mentioned strategies of using ultrathin substrate and/or symmetric structure to tune neutral plane have been also applied to realize robustly foldable solar cells. Choi et al. investigated the effect of substrate thickness on the flexibility of perovskite solar cells. When decreasing the substrate thickness from 100 to 2.5 µm, the strain in device significantly decreased, inhabiting the formation of the transgranular and intergranular cracks in perovskite thin films (Figure 6A). As a result, perovskite solar cells on 2.5 µm substrate could sustain the initial performance after 10,000 cycles of bending with a radius of 0.5 mm (Figure 6B). Furthermore, when introducing protection layer at the top of the devices, the neutral plane shifted to the perovskite absorber layer. As a result, the devices sustained the initial performance of as much as 88% after 100 cycles of crumpling (Figure 6C), enabling to power wearable devices.[11] Similarly, Kaltenbrunner et al. fabricated perovskite solar cells on 1.4 µm PET substrate, the devices could sustain initial performance during flexion with curvature radius to 10 µm and less.[21] Song et al. theoretically investigated the effect of substrate thickness on the strain in active layers of polymer solar cells. It was observed that reducing substrate thickness from 125 to 25 µm could effectively decrease strain in polymer solar cells, especially when flexion with the curvature radius lower than 1 mm. At curvature radius of 0.2 mm (similar to folding radius), strains in polymer solar cells on 150, 50, and 25 µm substrates were 35, 10, and 5%, respectively. Considering that the strain of 5% was lower than the crack onset strains for metal or organic layers which were in the range of 10–30%, foldable polymer solar cells on 25 µm substrate were experimentally realized.[14]

4 | FLEXIBLE MATERIALS FOR FOLDABLE SOLAR CELLS

Besides structure design, applying foldable materials to replace brittle ones would provide the room to improve device foldability. It is well known that when the strain in active layers exceeds their crack onset strain, cracks will be formed and extend to the adjacent layers after cyclic flexion, leading to the degradation even failure of solar cells. Therefore, adopting ductile layers with large crack onset strain instead of brittle ones is valuable. For the solar cells, the flexible alternatives mainly include substrates, transparent electrodes, and absorber layers.

4.1 | Flexible substrates

The most distinct and important difference between foldable and rigid solar cells is the folding endurance, which is predetermined by the substrates. Thus, the key requirements of substrates for foldable solar cells should be firstly considered. As far as we known, the following properties must be concerned to achieve both high efficiency and robust foldability of solar cells: (1) surface roughness, (2) thermal endurance, (3) coefficient of thermal expansion, (4) optical transparency, (5) resistance to solvent and moisture, (6) mechanical properties, such as the Young's modulus, toughness. In general, the substrates used in foldable

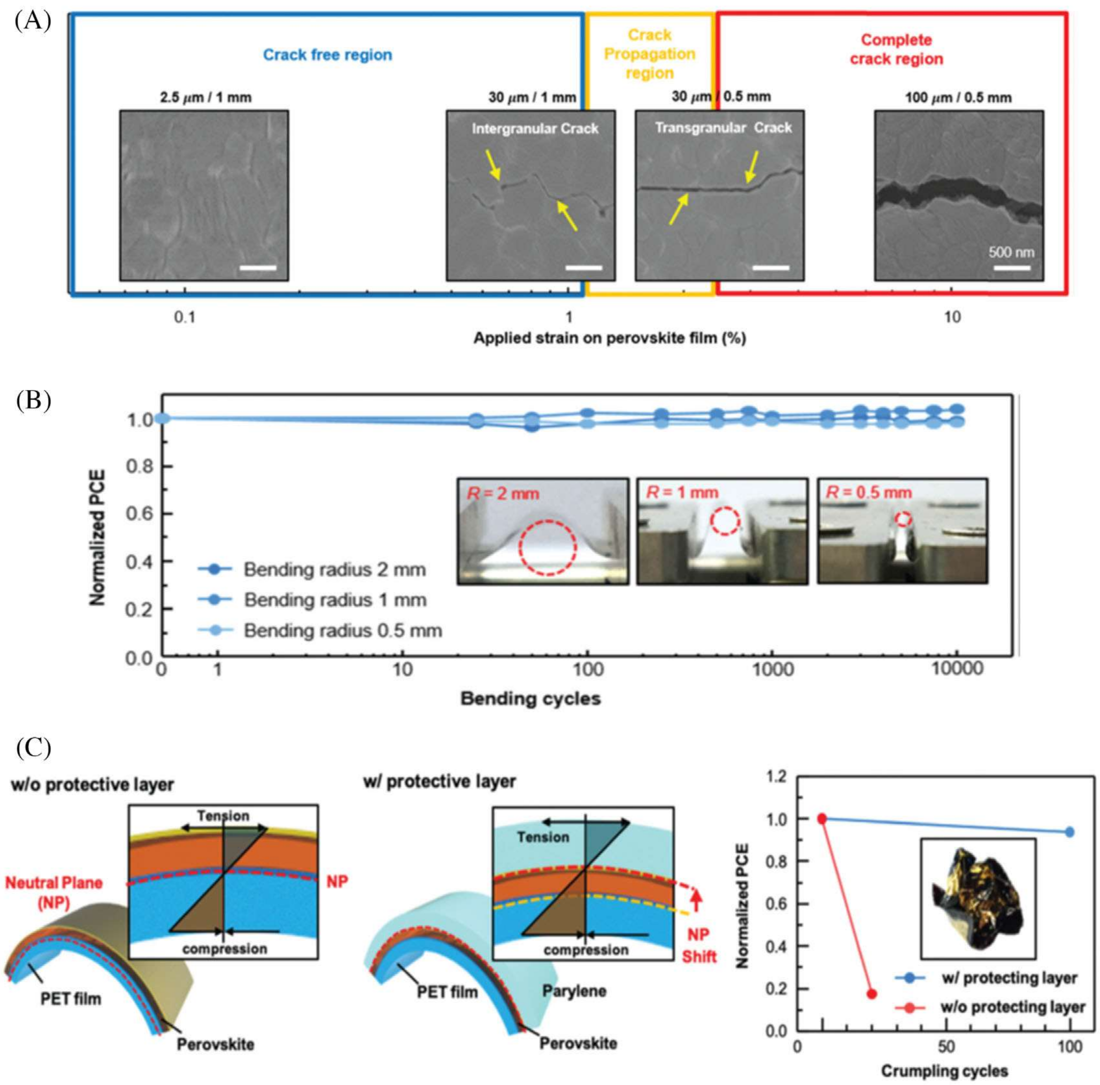

FIGURE 6 A, The applied strain and crack formed in perovskite films for solar cells on substrates with different thicknesses ($d = 2.5$, 30, 100 μm) and subjected to different bending radii ($R = 0.5$, 1 mm), B, Normalized PCE of perovskite solar cells on 2.5 μm substrate as a function of bending cycles, C, Illustrations of neutral plane shift in perovskite solar cells without and with a protective layer, and normalized PCE of the devices as a function of crumpling cycles without and with the protective layer.[11] Copyright 2019 Royal Society of Chemistry

solar cells mainly include the polymer films, metal foils, cellulose paper, fiber, woven textile, etc.

Polymer films play an important role in foldable solar cells due to their low cost, light weight, high optical transmittance, bendability, and resistance to solvent. The widely used flexible polymer substrates are PET, PEN, and PI, which have been applied in foldable solar cells as well as other optoelectronics. White et al. demonstrated highly flexible polymer light-emitting diodes on 1.4 μm PET, which were operable under extreme flexion condition with curvature radius under 10 μm.[29] Lee et al. demonstrated organic flash memory on 6 μm-thick Mylar substrate, which maintained their programming and erasing capability in the folded state as well as after 1200 folding cycles at a curvature radius of 300 μm.[26] Kaltenbrunner et al. demonstrated perovskite solar cells using 1.4 μm PET substrates and PEDOT:PSS electrodes, which can endure flexion into curvature radii down to 10 μm and less. In addition, the solar cells were stretchable.[21] However, PET and PEN, with the glass transition temperature of 105°C and 125°C, respectively,[30] are not sufficiently thermally stable, limiting the high temperature process for high-performance solar cells, especially for the mesoporous perovskite solar cells and CIGS solar cells. PI which has a high glass transition temperature >300°C, is a promising substitution for traditional PEN and PET substrates. Especially the development of colorless PI (CPI) enables its wide applications in solar cells without limitation in the direction of incident light. Rogers et al. demonstrated CMOS circuits that exploiting silicon nanoribbons on 1.2 μm PI

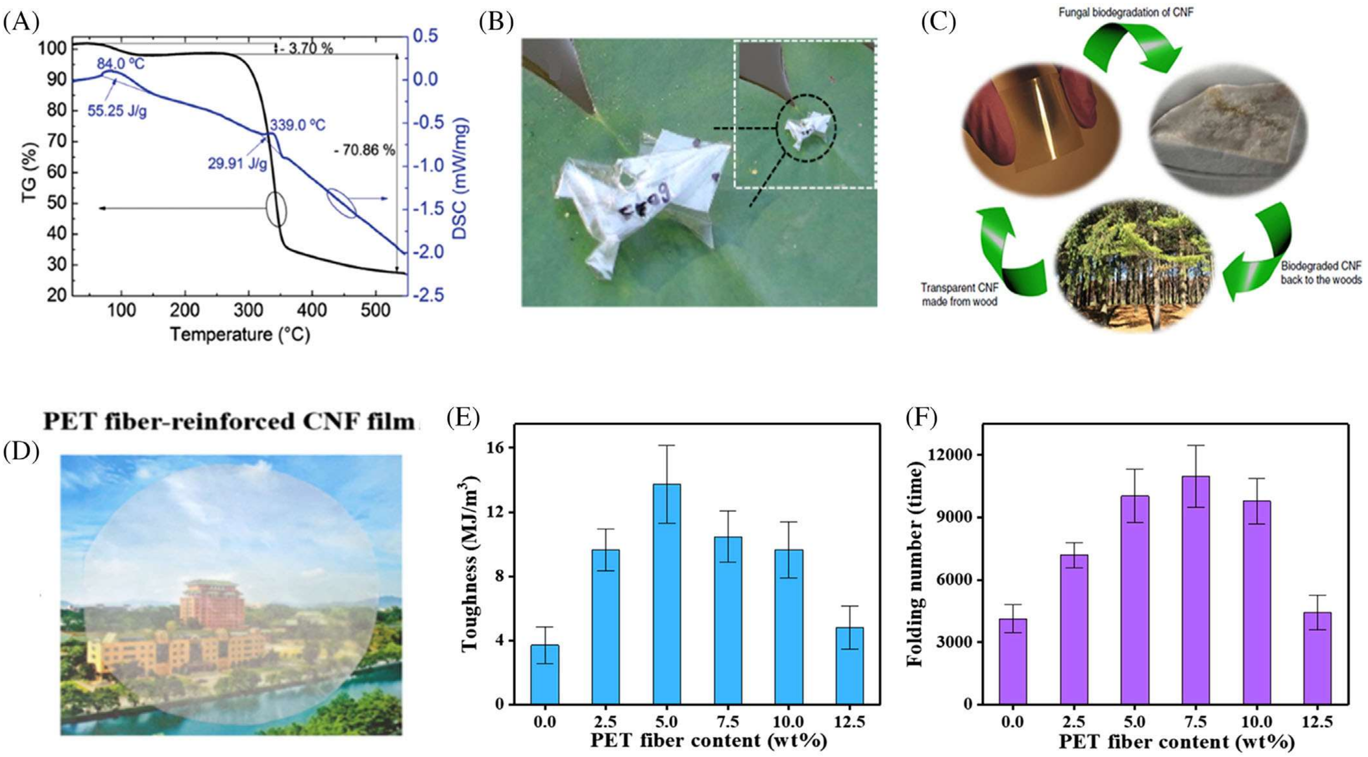

FIGURE 7 Advantages of paper substrates.[39–41] A, High thermal stability of ~280°C derived from TG/DSC results, Copyright 2015 John Wiley and Sons. B, foldability, Copyright 2020 American Chemical Society. C, recyclability. Copyright 2015 Nature Publishing. Improvement in mechanical properties of paper by polymer addition.[49] D, photograph of PET fiber-reinforced cellulose nanofiber films; comparison of E, Toughness and F, Folding endurance of CNF films with different PET fiber contents. Copyright 2020 Elsevier

substrate followed by PI encapsulating layer, composing PI/Si-CMOS/PI structure. As a result, Si-CMOS were extremely foldable, which were operable and exhibited good electrical properties after wrapped over the edge of a microscope over slip with thickness of ~100 µm.[28] Park et al. utilized 60 µm CPI as the substrates. ITO on CPI achieved a low sheet resistance of 57.8 Ω sq^{-1} and a high transmittance of 83.6% after annealing at 300 °C. As a result, the fabricated CPI-based perovskite solar cells exhibited a high PCE of 15.5%. More importantly, the solar cells on CPI exhibited better mechanical flexibility than those devices on PET did in the whole bending radius of 2–12 mm, mainly due to the lower thickness of CPI substrate.[31] Thanks to the mature product of ultrathin polymer, highly foldable solar cells on polymer substrates have been realized through using the strategies of ultrathin substrates or symmetric structure. However, PCE of solar cells on ultrathin polymer substrates should be further improved through process optimization.

Metal foils intrinsically have the high thermal endurance, high conductivity, low water vapor transmission, and excellent mechanical property, making them very suitable for the substrates of flexible solar cells. However, though normally bendable thin film solar cells on metal foils, for example, Ti, Cu foils, have been reported,[32–35] no foldable solar cells has been realized. It is considered to be due to the following reasons: firstly, the Young's module of metal foil is as high as several tens of GPa and even higher than 100 GPa; Secondly, it is not easy to realize ultrathin metal foils. As a result, the neutral axis plane locates far from the active layers, leading to the high strain in functional layers of devices under folding deformation.

Some other novel flexible substrates also have potential application in foldable solar cells, such as textile, cellulose paper, fiber.[14,15,19,36–38] Paper, with the advantages of low cost, light weight, foldability, degradability, and recyclability (Figure 7A–C), is promising substrate for foldable solar cells as well as other optoelectronics.[39–42] Moreover, paper possesses conformal foldability, meaning no external force is needed during folding. Thus, complex three-dimensional structure transformation as well as downsizing can be easily realized. Recently, with the development of smooth, transparent nanocellulose paper, high-performance thin film solar cells can be prepared on paper.[43–47] While the highest folding cycle for reported paper-based solar cells was only 50 times, far behind the practical requirements, which is considered to be limited by the intrinsic foldability of pure paper substrates. It is reported that the foldability of paper can be improved by optimization of paper morphology, or addition of other materials. Hu et al. reported that compared with roll paper, the plastic–paper showed 10 times increasement in folding endurance capacity both in the longitudinal and transverse directions, due to its densely packed structure. As a result, the paper with the addition of epoxy resin can endure more than 100 folds under 14.7 N pulling force.[48] Similarly, Fang et al. demonstrated that the PET fiber-reinforced cellulose nanofibril (CNF) films exhibited not only a maximum average toughness of 13.7 ± 2.4 MJ m^{-3} that was nearly four times stronger than that of pure CNF film (3.7 ± 1.1 MJ m^{-3}), bust also superior folding endurance of over 10^4 times that was an order of magnitude higher than that of pure CNF film (~4×10^3 times) under a tension of 9.8 N, as shown in Figure 7D–F. The favorable combination of foldability and toughness of CNF film was primarily ascribed to the uniform distribution and close packing of small-diameter PET fiber in the CNF network.[49] Furthermore, W. Cao *et al*. presented an effective method to improve the folding resistance of CNF films by adding MXene. The d-$Ti_3C_2T_x$/CNF composite paper exhibited up to 14,260 folding times under 4.9 N pulling load.[50]

Based on our understanding, polymer and paper substrates possess the advantages of smooth surface, high transparency, foldability, and low Young's module. In addition, the fabrication of ultrathin polymer and paper is gradually mature. Therefore, they are believed as promising substrates for foldable solar cells.

4.2 | Flexible transparent electrodes

To date, ITO still maintains its predominance as transparent electrodes for high-performance flexible thin film solar cells. While it is apparent that the intrinsic brittleness of ITO causes crack problem, which negatively influences the mechanical stability of the flexible devices. In addition, the low fabrication temperature leads to the inferior conductivity of ITO films, thus thicker ITO films are fabricated on flexible substrates, leading to the poorer mechanical robustness and low transmittance. It was usually observed in reported papers that when flexible solar cells were prepared on ITO electrodes, they degraded after bending with radius of 4 mm or even lower,[51,52] not to mention the folding process. Thus, flexible transparent electrodes as alternatives is important for realizing foldable solar cells. For application in foldable solar cells, the flexible electrodes should satisfy the following requirements in order to achieve high PCE as well as high foldability: (1) high conductivity, (2) high transparency especially in the visible wavelength range, (3) mechanical robustness, (4) spatially homogeneous, and (5) preparation technology compatible with flexible substrates. By now, carbon nanotube, graphene, ultrathin metal, metal nanowire, metal grids, conductive polymer, and their complex, have been widely applied in the robust bendable and foldable solar cells. The review for flexible transparent electrodes has been reported in other papers.[53–57] Herein, we focus on the application of flexible transparent electrodes for mechanical robust and highly efficient foldable solar cells.

Ge et al. developed the strategies to prepare high-conductivity and highly foldable PEDOT:PSS electrodes. Two constructive approaches were used to enhance PEDOT:PSS properties: Xylitol was used as the polyhydroxy compound dopant in the PEDOT:PSS aqueous solution and gentle methanesulfonic acid was employed for surface post-treatment at room temperature. As a result, PEDOT:PSS with both xylitol doping and MSA treatment (mentioned as D-PEDOT:PSS) exhibited an overall transmittance of ~80% across 350–850 nm wavelength region, low square resistance of 82 Ω sq^{-1}, high mechanical stability with deviation in resistance on the range of 20% after 1000 folding cycles with radius smaller than 1 mm, as well as uniform phase separation. Flexible solar cells using PBDB-T-2F:Y6 photoactive layer and D-PEDOT:PSS electrodes showed a high PCE of 14.20%. Moreover, these flexible solar cells also displayed remarkable mechanical stability, maintaining 68% of the original PCE after 1000 folding cycles with extremely small radius of less than 1 mm,[12] as shown in Figure 8A–C. Song et al. intensively investigated the optoelectrical properties of ultrathin silver electrodes prepared by magnetron sputtering at room temperature. Through using the strategies of seed layer or dopants, continuous and ultra-smooth Ag films with thickness ~10 nm or even lower were realized, providing it with low sheet resistance ~10 Ω sq^{-1} and semi-transparency. Preserving the conductivity, improvement of transparency was further achieved by sandwiching ultrathin Ag between two oxide layers forming the structure of oxide/ultrathin Ag/oxide (OMO). Thus, OMO electrodes possessed average transmittance >80% in wavelength range of 400–800 nm.[58–61] Furthermore, OMO exhibited good mechanical flexibility: the resistance of OMO on paper was almost constant under continuous bending with radius of 1 mm for 1000 cycles, and the resistance of OMO on paper only increased five times compared to the initial value after folding for 50 cycles. Using OMO electrodes instead of ITO, both foldable polymer solar cells and perovskite solar cells were demonstrated, which can endure folding for 35 and 50 cycles at 180^o angle, respectively,[14,19] as shown in Figure 8D–F. Yoon et al. applied the strategy of MoO_3 doping for monolayer graphene films to significantly decrease sheet resistance by over 50% to 552.0 Ω sq^{-1}. In addition, the single-layer graphene presented high transmittance of ~97% over the visible wavelength range. As a result, flexible perovskite solar cells utilizing MoO_3 doped monolayer

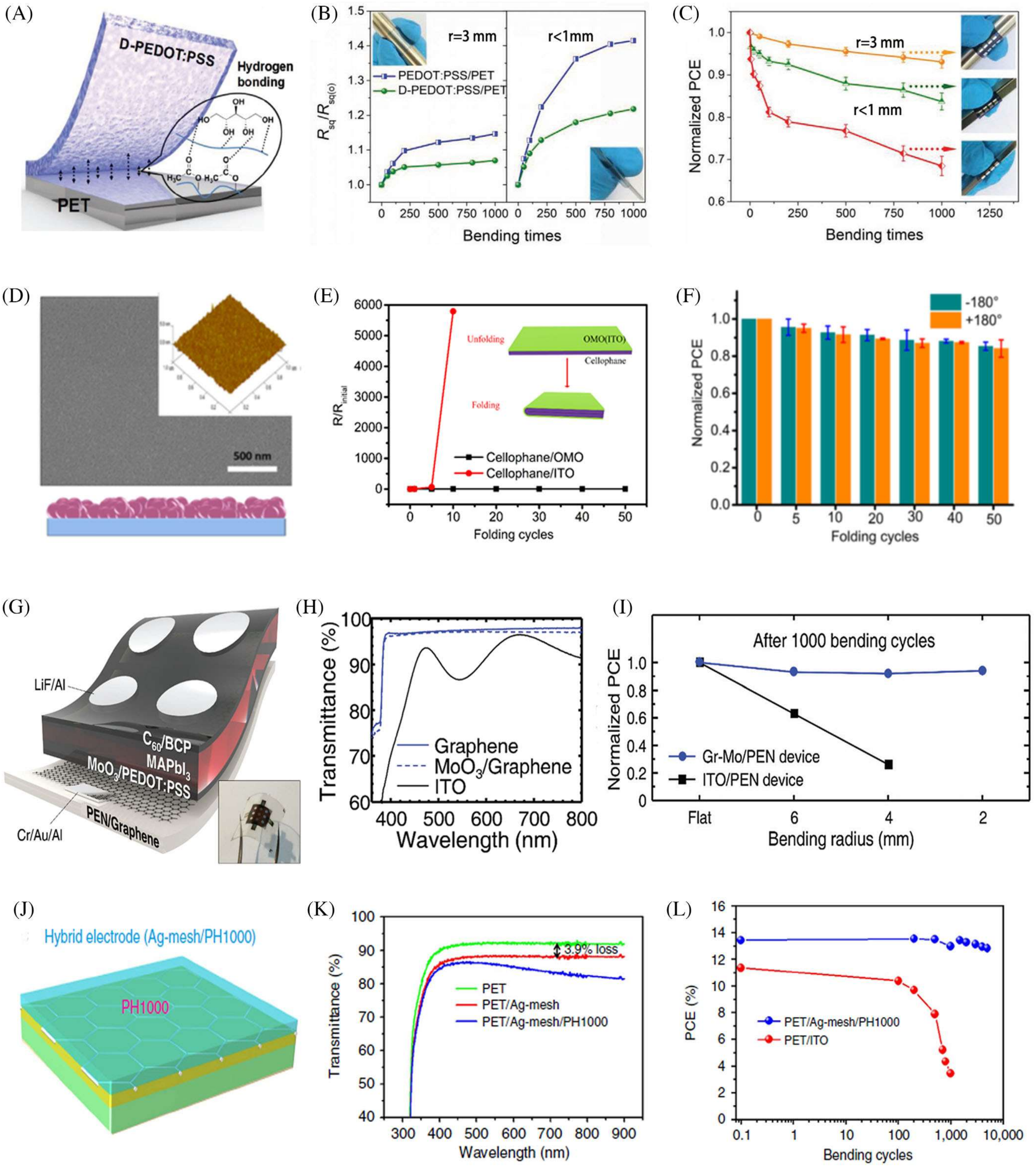

FIGURE 8 Mechanical robust solar cells using flexible transparent electrodes. A, schematic diagram of D-PEDOT:PSS/PET interface, (B) Sheet resistance change of D-PEDOT:PSS and PEDOT:PSS, and (C,) Normalized PCE of organic solar cells after bending (orange), folding between Ag electrodes (green), and folding on top of the Ag electrodes (red).[12] Copyright 2020 John Wiley and Sons. D, Surface morphology of 6 nm Ag films, E, Resistance change of OMO and ITO with folding cycles, and F, Normalized PCE of perovskite solar cells after folding with different cycles.[19,59] Copyright 2019 John Wiley and Sons. Copyright 2018 Elsevier. G, Device structure of graphene-based perovskite solar cells, H Transmittance of graphene, MoO_3-doped graphene, and ITO on PET, and I Normalized PCEs of solar cells using MoO_3-doped graphene and ITO electrodes after bending with various radii.[52] Copyright 2017 Royal Society of Chemistry. J, Diagram of Ag-mesh/PH1000 hybrid electrode, K, Transmission spectra of bare PET, PET/Ag-mesh, and PET/Ag-mesh/PH1000, and L, PCEs of perovskite solar cells as a function of bending cycles at a radius of 5 mm.[62] Copyright 2016 Nature Publishing

graphene as transparent electrodes reached PCE of 16.8% with no hysteresis. More importantly, the flexible devices also demonstrated superb stability against bending deformation, maintaining 85% of initial PCE after bending with curvature radius of 2 mm for 5000 cycles, paving the way for foldable photovoltaic applications,[52] as seen from Figure 8G–I. Li et al. fabricated hybrid electrodes consisting of embedded Ag-mesh and conductive polymer (PH1000) on PET, which showed high transmittance >80% in the visible region, low sheet resistance of ~3 Ω sq^{-1}, and low surface roughness of 2 nm, suggesting potential application in flexible solar cell.[62,63] Flexible perovskite solar cells using hybrid electrodes exhibited PCE of 14% and excellent bending stability retaining over 95.4% of its initial PCE value even after 5000 fully bending cycles with bending radius of 5 mm,[62] as exhibited in Figure 8J–L.

Though many flexible transparent electrodes with excellent optoelectrical properties demonstrate their potential application in foldable solar cells, many questions should be further answered, such as which flexible electrodes are better for the application in highly efficient and foldable solar cells. As far as we known, considering the high photovoltaic performance, high foldability, large area uniformity for foldable solar cells, conductive polymer and OMO electrodes are more promising.

4.3 | Ductile absorber for foldable solar cells

Besides the substrates and transparent electrodes, the mechanical properties of absorber layer also significantly influence the foldability of solar cells. Firstly, the mechanical properties, especially the Young's module and hardness of different kinds of absorber are various. For example, it was reported that the elastic module of polymer was ~1 GPa,[64] while the elastic module of perovskite was ~10 GPa,[22,65–68] and the elastic module of inorganic (CIGS or a-Si) increased to several tens of GPa or >100 GPa.[25,69,70] Thus, the brittleness of polymer, organic-inorganic perovskite, inorganic Si or CIGS films increased in turn, leading to the difference in bendability or foldability of thin film solar cells. This can be used to explain why ultraflexible or foldable solar cells are more easily realized in polymer or perovskite solar cells rather than inorganic solar cells. Song et al. applied the same strategies of using the 25 μm ultrathin cellophane substrates combined with OMO flexible transparent electrodes to construct flexible polymer/perovskite/silicon thin film solar cells. The formed polymer and perovskite solar cells can endure folding for dozens of cycles, while thin film silicon solar cells were only bendable with radius of 1 mm for 50 cycles.[14,19,71] Besides the type of absorber, the microstructure of absorber, for example, composition, grain size, bonding strength, also affects the mechanical properties of thin film solar cells. Kim et al. demonstrated highly efficient and mechanically robust all polymer solar cells using PBDTTTPD polymer donor and the P(NDI2HD-T) polymer acceptor. It was reported that compared to PBDTTTPD:PCBM (1:1.5 w/w) with elastic modulus and the elongation at break of 1.76 GPa and 0.12%, respectively, the PBDTTTPD:P(NDI2HD-T) blend films possessed lower tensile modulus and higher elongation at break of 0.43 GPa and 7.16%, respectively. This was due to the greater intrinsic flexibility of polymer than fullerenes, as well as the strengthened donor/acceptor interfaces by the entanglements between the polymer chains. As a result, the PBDTTTPD:P(NDI2HD-T) bend films maintained original conductance and morphology after bending with small radius of 1 mm, while the conductance degraded and the crack formed in PBDTTTPD:PCBM after bending with the same condition.[64] Also, the mechanical properties of perovskite absorber will be influenced by its structural characteristics based on simulative and experimental results. Rakita et al. investigated elastic modulus and nanohardness values of $APbX_3$ (A = Cs, CH_3NH_3; X = I, Br) single crystals from nano-indentation experiments. It was observed that elastic properties were mainly dominated by the type and strength of B-X bond. Br-based perovskite possessed higher Young's modulus than that of the I-based one [$E(CH_3NH_3PbI_3) = 14.2 \pm 1.9$ GPa, $E(CH_3NH_3PbBr_3) = 19.6 \pm 0.3$ GPa]. In addition, it was shown that the organic group made the entire structure stiffer (higher elastic modulus) through comparison the Young's modulus between $CH_3NH_3PbBr_3$ and $CsPbBr_3$.[65] Park et al. investigated the mechanical properties of perovskite polycrystalline films using the instrumented nanoindentation technique. The elastic modulus of $CH_3NH_3PbI_3$ was determined as 12.3 ± 1.2 GPa from the strain-stress curve in the elastic regime.[22] Lee et al. considered that the mechanical properties determined by the nanoindentation could be inaccurate compared to the uniaxial tensile testing. The $MAPbBr_3$ layer exhibited elastic modulus of 4.38 (±0.23) GPa and fracture strength of 45.4 (±7.5) MPa corresponding to fracture strain of 1.1 (±0.14)%, based on uniaxial tensile testing.[66] Padture et al. investigated the effect of grain size on the mechanical stability of perovskite thin films. It was exhibited that the fine-grained $MAPbI_3$ films with grain size of 290 nm tended to fracture intergranularly with the corresponding toughness of 0.41 J m^{-2}, which was related to the grain-boundary toughness. In contrast, the coarse-grained $MAPbI_3$ films with larger grain size of 730 nm tended to facture at the $MAPbI_3$/ETL interface with the corresponding toughness of 1.14 ± 0.24 J m^{-2}, which was related to the toughness of the $MAPbI_3$/ETL interface.[67] However, the investigation on the mechanical properties of absorber is still in the initial state. More work should be done to know which kind of absorber is beneficial for the foldable solar cells.

4.4 | Strong interface adhesion for foldable solar cells

Besides the flexible active layers, the interface adhesion will also affect the mechanical flexibility of solar cells, which has not aroused enough attention by now. When the fracture strength of each functional layer is large enough, the fracture may firstly occur at the relative weak path of the interface, especially when the interface binding is based on the low van der Waals force. Thus, strong interface adhesion is required to avoid the interface delamination. Im et al. introduced APTES (3-aminopropyl triethoxysilane) adhesion promoter between PET substrate and $AuCl_3$-doped graphene transparent electrodes. The $AuCl_3$-GR/APTES/PET did not show significant degradation of the sheet resistance with respect to the bending radius and the bending cycles, due to the formation of covalent bonds by the APTES inter-layer. Whereas the $AuCl_3$-GR/PET substrate had significant degradation of the sheet resistance after a bending test due to the break-off or delamination of $AuCl_3$-GR from the PET substrate. Accordingly, the perovskite solar cells constructed on the $AuCl_3$-GR/APTES/PET substrate exhibited excellent bending stability which maintained their PCE at over 90% of the initial value after 100 bending cycles at $R \geq 4$ mm.[72]

5 | CONCLUSION AND OUTLOOK

By now, the foldable solar cells generally exhibit low PCE and inferior folding stability compared with that of normal bendable solar cells. However, due to the advantages of size compactness and shape transformation, they have unique applications such as portable and wearable electricity supplier, as well as building, transportation and agriculture integrated photovoltaics. The development of highly foldable and efficient solar cells is of great importance. Here, we have provided a concise overview on the recent progresses in foldable solar cells, and discuss the critical requirements to realize robust foldable solar cells including the structure design and flexible alternatives. In order to push foldable solar cells development, we will provide our personal perspectives on the challenges and future directions for foldable solar cells as follows:

Firstly, there is a lack of standards for the folding procedure, especially how to produce crease. Nowadays, some fold solar cells around cylinder with radius smaller than 1 mm; while others directly fold solar cells with or without force. In our opinion, the latter procedure is more reasonable. However, the curvature radius of direct folding is often difficult to characterize. As far as we known, the folding radius depends on the device total thickness and imposed force during folding, which is various with the range from several micrometers to several sub-millimeters. Thus, it is better to mention the detailed information of device and folding procedure in the experiments. Further giving the direct characterization of devices during folding, such as SEM, is beneficial to exactly know the folding radius. It is believed that the specific definition of folding procedure can promote the development of foldable solar cells.

Secondly, considering the practical application of foldable solar cells, the performance of foldable solar cells needs to be improved, including the photovoltaic performance and folding stability. As seen from Table 1, the PCE of foldable solar cells is lower than that of the rigid solar cells, which is supposed to be attributed to the following two reasons: on one hand, ultrathin substrates used to construct foldable solar cells are easily curved due to the induced stress, leading to the nonuniformity of the temperature distribution as well as thin films properties. On the other hand, the growth behavior and the morphologies of absorber on flexible electrodes have not be optimized. Higher PCE of foldable solar cells can be achieved if the above two processes are optimized. Besides photovoltaic performance, the folding stability should be improved. Take the application of portable power supplier for example, supposing it works for 5 years and is used three times a day, more than 5000 cyclic folding stability should be allowed. It is considered that the intensive investigation on failure mechanism of solar cells under repeated folding and unfolding is an effective way to improve the mechanical stability. For the solar cells with multilayers, the folding induced crack and delamination may firstly occur in active layer or interface, depending on the stress distribution in the device during folding, the crack onset stress of each functional layers, as well as the bonding at the interface. Thus, some critical data should be offered, such as crack onset stress of flexible substrate, electrodes, absorber and interface, as well as stress distribution in devices. Therefore, more theoretical and experimental work should be conducted to reveal the failure mechanism of foldable solar cells as well as to improve the folding endurance of solar cells.

Finally, scalability and environmental stability of foldable solar cells are the fundamental challenges to overcome before large-scale application. Nowadays, all the results are based on the small-sized foldable solar cells, the performance of large-sized solar cells is unknown, not to mention the solar modules. The dependence of photovoltaic performance and foldability of solar cells on size need to be intensively investigated. In addition, encapsulation for foldable solar cells especially on the ultrathin substrates is another challenging topic.

It is believed that with efforts from both academia and industry, foldable solar cells will add more colors to our daily life in the future.

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