REVIEWS

Progress in flexible perovskite solar cells with improved efficiency

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Abstract: Perovskite solar cell has emerged as a promising candidate in flexible electronics due to its high mechanical flexibility, excellent optoelectronic properties, light weight and low cost. With the rapid development of the device structure and materials processing, the flexible perovskite solar cells (FPSCs) deliver 21.1% power conversion efficiency. This review introduces the latest developments in the efficiency and stability of FPSCs, including flexible substrates, carrier transport layers, perovskite films and electrodes. Some suggestions on how to further improve the efficiency, environmental and mechanical stability of FPSCs are provided. Specifically, we considered that to elevate the performance of FPSCs, it is crucial to substantially improve film quality of each functional layer, develop more boost encapsulation approach and explore flexible transparent electrodes with high conductivity, transmittance, low cost and expandable processability.

Key words: perovskite solar cells; flexible electronics; thin film deposition; carrier transport

1. Introduction

The development of photovoltaics that convert sunlight into electricity is a promising strategy to meet the rapidly growing energy needs. In the past few decades, silicon-based photovoltaic technology has made significant progress with respect to the cost and efficiency, which promoted clean energy production. Along with the advance of silicon solar cells, a variety of thin-film photovoltaics have been studied in order to further meet the application requirements of lightweight and low-cost. Especially in recent years, great interest in flexible and wearable electronic products has led to growing research activities on flexible and stretchable thin-film photovoltaic. Integrating these thin and soft solar collectors into walls, windows and portable objects will change the current way of energy production, reduce pollution, and greatly expand the usage scenarios and ways of obtaining energy.

Recently, the organic–inorganic hybrid perovskites have become the ideal candidate to develop flexible solar cells due to their excellent optoelectronic properties. In particular, due to its excellent light harvesting ability, only about 300 nm thick film is sufficient to absorb essentially all visible light above its band gap. Thus, ultra-thin and ultra-light solar cells can be prepared[1–8]. The structure and physical properties of organic–inorganic halide perovskites were first reported in 1978[9, 10]. They have a cubic structure with the general formula of ABX_3, where A is typically $CH_3NH_3^+$ (MA^+) or $NH_2CHNH_2^+$ (FA^+), B is Pb^{2+} or Sn^{2+}, and X is halogen anion (Cl^-, Br^-, or I^-). By adjusting the composition of A, B and X in the crystal, the bandgap of the metal halide perovskite can
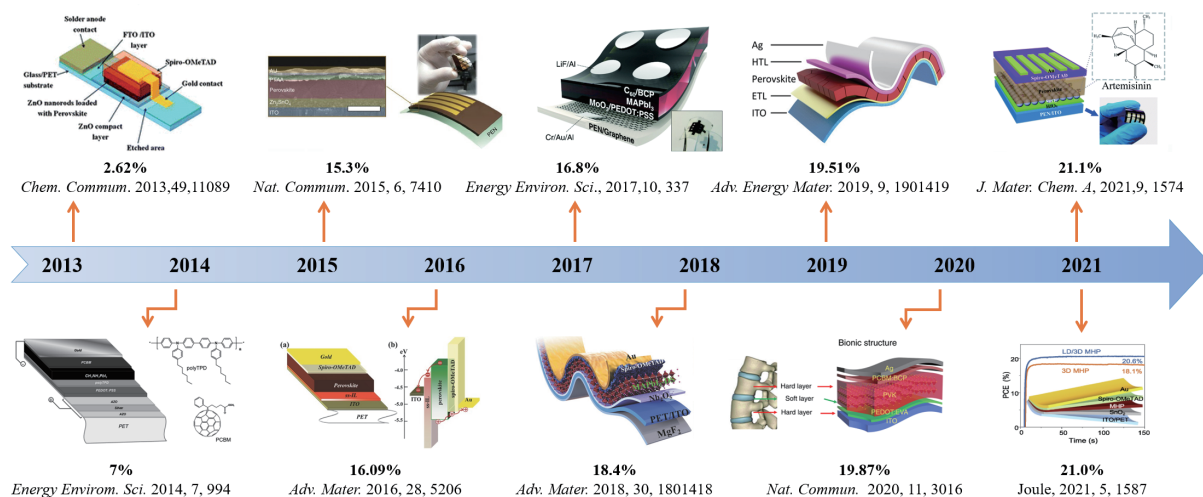
be continuously tuned in the range of 1.15 to 3.06 eV. $CH_3NH_3PbI_3$ and $CH_3NH_3PbI_{3-x}Cl_x$ are the most commonly studied absorbers in the early perovskite solar cells (PSCs) research. Since the $CH_3NH_3PbI_3$ perovskite material was first used as absorber in solar cells in 2009, the power conversion efficiency (PCE) of PSCs has rapidly increased from 3.81% to a certified 25.5% within ten years development[5, 7, 8, 11–16]. Similar to rigid PSCs, flexible perovskite solar cells (FPSCs) have also made great progress with respect to device efficiency. Recently, the PCE of FPSCs has reached 21.1%[17], Fig. 1 shows the PCE improvement of FPSCs from 2013 to 2021 under various optimization approaches. However, their poor long-term stability and relatively lower PCE compared with rigid counterparts are the main challenges that limit its further commercialization.

FPSCs are mainly composed of substrate (PEN, PET, etc.), bottom electrode (FTO, ITO, etc.), electron transport layer (ETL), perovskite active layer, hole transport layer (HTL), and the counter electrode (Au, Ag, etc.). At present, the typical FPSCs structure are sandwich structure, in which the perovskite active layer is in the middle, with electron (or hole) blocking layers and electrodes on both sides. When the perovskite material is excited by light, it will produce photo-generated electron–hole pairs. Since the valence band maximum (VBM) of ETL is much lower than the VBM of perovskite materials, and their conduction band minimum (CBM) is at the same position, ETL plays the role of blocking holes and transporting electrons. The CBM of the HTL on the other side is much higher than the CBM of the perovskite material, and the positions of the VBM of them are the same, so the HTL plays the role of blocking electrons and transporting holes. The ETL and HTL are connected to the external circuit through the electrodes on both sides, which can realize the directional transfer of charges to form current.

2.62%
Chem. Commum. 2013,49,11089

15.3%
Nat. Commun. 2015, 6, 7410

16.8%
Energy Environ. Sci., 2017,10, 337

19.51%
Adv. Energy Mater. 2019, 9, 1901419

21.1%
J. Mater. Chem. A, 2021,9, 1574

2013   2014   2015   2016   2017   2018   2019   2020   2021

7%
Energy Envirom. Sci. 2014, 7, 994

16.09%
Adv. Mater. 2016, 28, 5206

18.4%
Adv. Mater. 2018, 30, 1801418

19.87%
Nat. Commun. 2020, 11, 3016

21.0%
Joule, 2021, 5, 1587

Fig. 1. (Color online) The PCE evolution of FPSCs from 2013 to 2021[17–26].

Table 1. Performance parameters of polymer substrate[28].

Substrate	PEN	PET	PI	PC
T_g (°C)	120–155	70–110	155–270	145
T_m (°C)	269	115–258	250–452	115–160
Density (g/cm³)	1.36	1.39	1.35–1.43	1.20–1.22
Modulus (MPa)	$(0.1–0.5) \times 10^3$	$(2–4.1) \times 10^3$	2.5×10^3	$(2.0–2.6) \times 10^3$
Work temp (°C)	–	−50 to 150	<400	−40 to 130
CTE (ppm/°C)	20	15–33	8–20	75
Water absorption (%)	0.3–0.4	0.4–0.6	1.3–3.0	0.16–0.35
Solvent resistance	Good	Good	Good	Poor
Dimensional stability	Good	Good	Fair	Fair

T_g: glass transition temperature, T_m: melting temperature, CET: coefficient of thermal expansion.

Here we analyze the factors that affect the performance of FPSCs, mainly lies in 1) the roughness of the flexible substrate affects the quality of the perovskite film, 2) the high resistance and low light transmission of the flexible substrate lead to the low short-circuit current of the solar cell devices, 3) the permeability of the flexible substrate to water and oxygen results in long-term instability, 4) the mechanical stress occurred during the device fabrication brings into the fracture of perovskite layers. Then we summarized the development of flexible substrates, carrier transport layers, light absorption layers and electrodes to address the above concerns to improve the performance of flexible perovskite solar cells. Finally, we discussed the packaging process of FPSCs to further prolong the device's lifetime.

2. Performance improvement of flexible substrates

The most obvious difference between the flexible substrate and rigid substrate is whether the resulting devices can be bent and stretched. Its bending performance is mainly determined by the physical properties of the flexible substrate. An excellent flexible substrate is a basis for building a high-performance flexible perovskite solar cell. It should have the following characteristic. 1) Excellent mechanical properties, 2) Good optical properties: A suitable substrate should be transparent to the light absorption range of perovskite, particularly in the visible light region. 3) Good chemical properties: Be-

cause a variety of chemical compounds are used in the preparation of FPSC, the substrate should have chemical stability. 4) Blocking the permeation of oxygen and water: The permeation of oxygen and water is the main factor causing the performance degradation of perovskite solar cells. At present, the commonly used substrates for flexible perovskite solar cells include polymer substrates, metal substrates, ultra-thin flexible glass substrates and other special materials, etc.

2.1. Polymer substrates

Polymer substrates have been most widely used in FPSC due to their advantages of high light transmittance, high flexibility, low cost, and good chemical stability[1, 23, 25, 27]. The parameters of commonly used polymer substrates, such as polyethylene terephthalate (PET), polyethylene naphthalate (PEN), polyimide (PI) and polydimethylsiloxane (PDMS), are shown in Table 1[28]. Among them, PET and PEN are the most commonly used flexible substrates, which are further coated with transparent conductive indium tin oxide (ITO) to serve as the electrode. So far, the efficiency of single-junction FPSCs using PEN and PET substrates has reached 21.0%[17, 26]. Yang *et al.* prepared an FPSC with a PCE of 21.1% in 2020, and the device structure is PEN/ITO/HfO$_x$/SnO$_2$/perovskite: artemisinin/Spiro-OMeTAD/Au. The composition of the perovskite absorber is Rb$_{0.05}$Cs$_{0.05}$(FA$_{0.83}$MA$_{0.17}$)$_{0.9}$Pb(I$_{0.95}$Br$_{0.05}$)$_3$, doping with artemisinin molecules. Artemisinin molecules with carbonyl groups can easily interact with exposed surface Pb^{2+} ions through the Lewis acid-base interaction, which
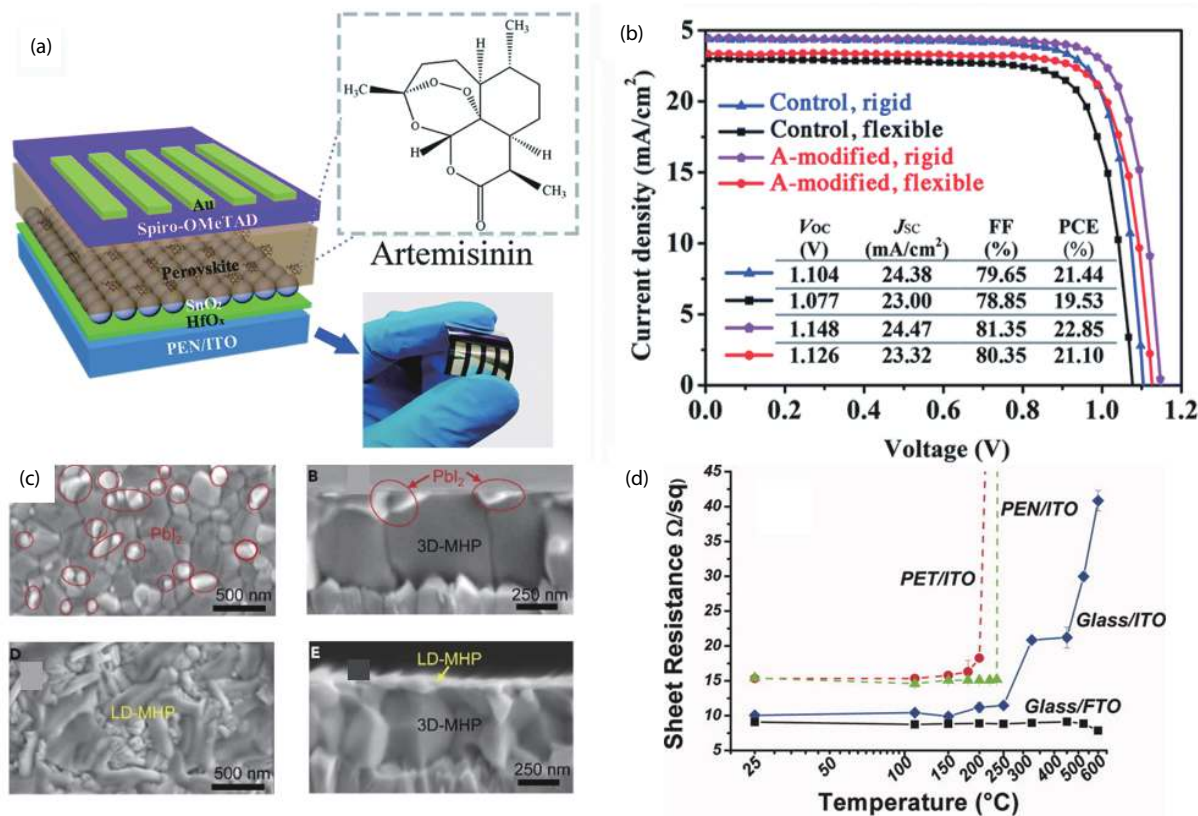
Fig. 2. (Color online) High-efficiency FPSC based on PEN and PET substrates. (a) Schematic diagram of the FPSC structure based on a perovskite layer doped with artemisinin. (b) J–V curves on rigid and flexible substrates with and without artemisinin doping[17]. (c) Scanning electron microscope characterization of thin film deposited on glass/fluorine-doped tin oxide (FTO) substrate[26]. (d) The room temperature sheet resistance of conductive PET/ITO, PEN/ITO, glass/ITO, and glass/FTO substrates after heat treatment at different temperatures for 30 min[29].

effectively passivates grain boundaries and surfaces (as shown in Figs. 2(a) and 2(b))[17]. Shi et al. prepared FPSC based on PET/ITO/SnO$_2$/(CsPbI$_3$)$_{0.04}$[(FAPbI$_3$)$_{0.9}$(MAPbBr$_3$)$_{0.1}$]$_{0.96}$/ low dimension perovskite/Spiro-OMeTAD/Au structure[26]. In this structure, the 2D perovskite is formed by in situ spin-coating 3-CBAI solution on the three-dimensional perovskite, which can effectively "seal" surface defects as shown in Fig. 2(c), obtain a PCE of 21.0%. However, there are some urgent issues in polymer substrate that need to be solved, such as its limited processing temperature tolerance, and the subsequent processing should be performed at less than 150 °C. When elevating the processing temperatures to 150 °C, the sheet resistance of plastic substrate will increase accompanying with substrate deformation, as shown in Fig. 2(d)[29]. Another key challenge in plastic substrates is their poor isolation from oxygen and moisture. This will further lead to severe device degradation, since perovskite materials are unstable in humid environments. Therefore, further packaging is required to improve the device stability.

2.2. Metal substrates

Compared with polymer substrates, metal foil has better thermal stability, flexibility and conductivity, which makes them promising flexible substrates for FPSCs. The metal foil is generally used as both the substrate and the electrode, thus simplifying the preparation process. Flexible perovskite solar cells need to have light transmission on at least one side, but the opacity of the metal foil requires the top electrode on the other side to have light transmission.

Ti foils is the most commonly used metal foil substrate in FPSCs due to the compatibility of Ti foils with the subsequent growth of TiO$_2$ electron transport layer. In 2015, Lee et al. used Ti foil as a flexible substrate and fabricated a flexible perovskite solar cell device with a Ti/compact TiO$_2$/mesoporous TiO$_2$/perovskite/Spiro-OMeTAD/Ag structure for the first time, with an initial efficiency of 6.15%[30]. After that, they further used Ag nanowire grids and Ag embedded indium tin oxide as the top transparent electrode to increase the PCE of the device to 7.45% and 11.01%[31, 32]. Watson et al. reported that the Ti foil-based mesoporous FPSC using Al$_2$O$_3$ mesoporous film as the scaffold[33], using ex-situ prepared and laminated transparent electrodes, they obtained a PCE as high as 10.3%. They also predicted the potential application of "self-generating" TiO$_2$ through the thermally induced in situ oxidation of titanium foil. Lee et al. proved this concept, and they used thermal oxidation on titanium foil to obtain TiO$_2$, which become the electron transport layer of FPSCs[34]. By controlling the annealing temperature, the concentration of oxygen vacancies in TiO$_2$ is modulated, which significantly affected the electron collection efficiency of the device. The TiO$_2$ obtained in the two-step annealing process of the Ti foil at 400 °C enabled the device to deliver a PCE of 14.9%. During this period, the second annealing step in an O$_2$ atmosphere is very important for removing oxygen vacancies without dramatically increasing the thickness of TiO$_2$. FPSC based on Ti foil exhibits excellent flexibility. After 1000 cycles under 15 and 4 mm bending conditions, the device maintains almost 100% of the initial PCE, and 77% of the initial
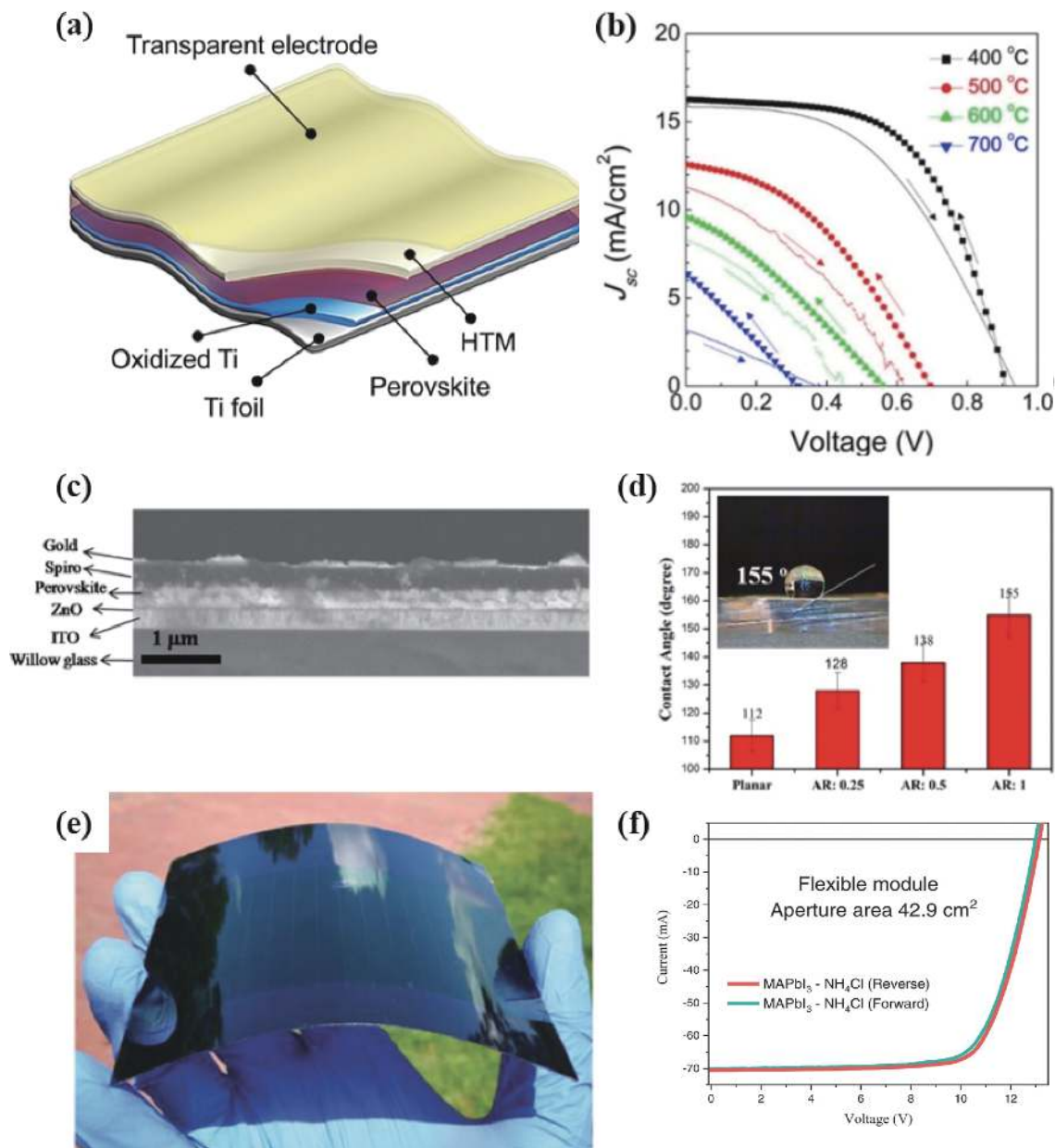
Fig. 3. (Color online) (a) Schematic diagram of FPSC structure based on Ti foil. (b) $J-V$ curves of Au/Cu/HTM/CH$_3$NH$_3$PbI$_3$/TiO$_2$/Ti cells under 100 mW/cm^2 AM 1.5G solar light with the same oxidized thickness of TiO$_2$ layer (~50 nm) based on the same ambience, air, with different annealing temperatures[35]. (c) FPSC cross-section SEM based on ultra-thin Willow Glass substrate[37]. (d) Static contact angle of deionized water on PDMS layers with different aspect ratios. (e) Photograph of a flexible perovskite module. (f) $J-V$ curve of the champion flexible perovskite modules[38].

PCE is maintained even after 1000 cycles under 1 mm bend radius. They further found that the resistivity of the TiO$_2$ layer on the Ti foil did not change under the conditions of $R \approx 15$ and 4 mm, but it decreased significantly under the conditions of $R \approx 1$ mm. Cracks are proven to be generated in the surface-oxidized Ti foil due to bending at a low radius, and leakage current emerged on the inner surface of the cracks, which leads to a deterioration of device performance. In addition, TiO$_2$ nanotubes and nanowire arrays grown on Ti foil were used to fabricate FPSC, and 13.07% of PCE was obtained[35].

In addition to Ti foil, Cu foil is also used to prepare FPSCs. Ahmadi et al. prepared cuprous iodide on the copper foil by exposing the pre-cleaned Cu foil to iodine vapor, and then used it as the hole transport layer of FPSC. Using zinc oxide as the electron transport layer and sprayed silver nanowires as the top electrode, the maximum PCE of the device is 12.80%[36].

2.3. Ultra-thin flexible glass substrate

Ultra-thin flexible glass substrates retain most of the advantages of rigid glass substrates, such as resisting high temperatures, blocking moisture and oxygen, and high light transmittance. Tavakoli et al. used 50 μm thick willow glass as a flexible substrate for the first time to prepare a flexible perovskite solar cell with a PCE of 12.06%[37]. Adding a PDMS anti-reflective nanostructure film on the front surface of the device can further boost the efficiency to 13.14%. At the same time, the prepared FPSC exhibits excellent stability be-
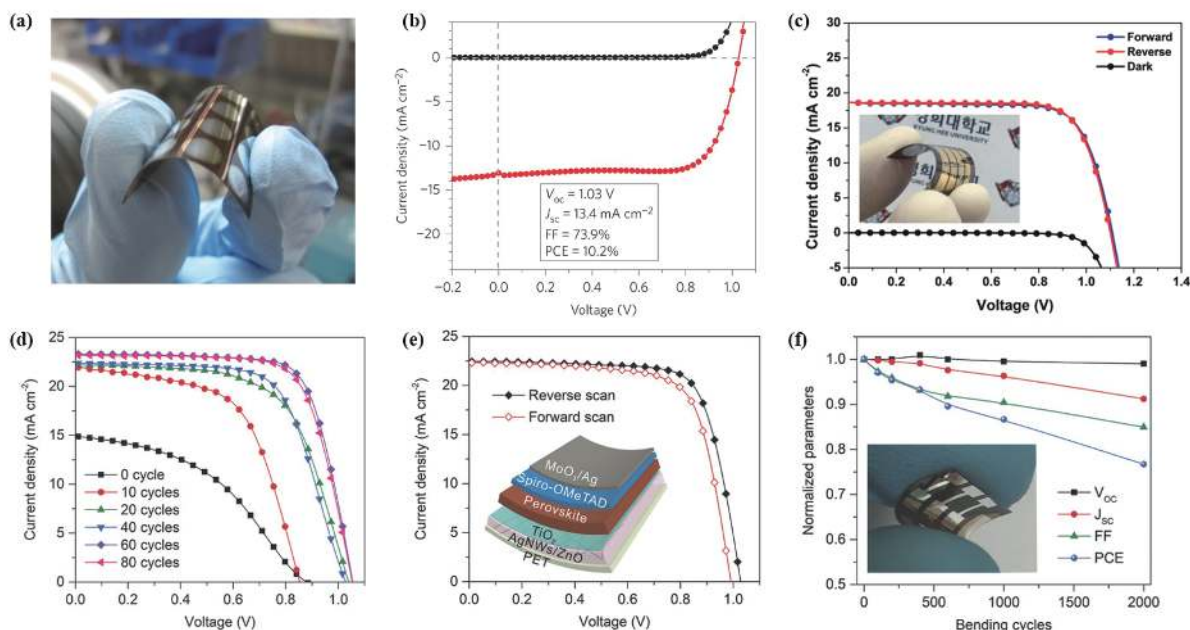
Fig. 4. (Color online) (a) Flexible perovskite device diagram[18]. (b) *J*–*V* curve of FPSC based on ZnO prepared at low temperature. (c) Light and dark *J*–*V* curves of FPSC[41]. (d) *J*–*V* curve under different ALD cycles. (e) Optimized FPSC structure and its *J*–*V* curve. (f) Variation of VOC, JSC, FF and PCE with bending times[42].

cause of the poor permeability of the flexible glass to oxygen and moisture, as shown in Fig. 3(d). Learned from this strategy, Dou *et al*. produced an FPSC efficiency of 18.1% by using MgF_2 as the anti-reflection layer and replacing ITO with indium zinc oxide (IZO) on ultra-thin flexible glass[39].

Compared with widely used polymer substrates, the more prominent advantage of ultra-thin flexible glass is its smooth and non-deformable surface at high temperatures. Based on this advantage, a large-area module (42.9 cm^2) was prepared with an efficiency of 15.86%[38], facilitated by the high-quality perovskite film obtained by doctor blade coating with an NH_4Cl additive. The flexible perovskite module is shown in Fig. 3(e). However, compared with other flexible substrates, fragility, high weight and relatively high cost may become the big concerns, which also limits its wide application.

3. Carrier transport layer

The carrier transport layer in the perovskite solar cell plays an important role, which is responsible for extracting electrons and holes from the interface and transporting them to the corresponding electrodes[40]. Therefore, the requirements for the carrier transport layer are band structure matched with perovskite, high light transmittance and excellent carrier mobility. For FPSCs, low-temperature processing requirements are posed. Therefore, research on flexible perovskite solar cells mainly focuses on low-temperature preparation of carrier transport layers.

3.1. Electron transport layer

The excellent electron transport layer (ETL) should have high electron mobility and high light transmittance. At the same time, the conduction band maximum of ETL is essential to match the energy level of the perovskite, which can effectively transport electrons and block holes.

Because of its high electron mobility and low-temperature processability, ZnO was first used as an electron transport layer to prepare FPSCs[18]. But the PCE of this device is

only 2.62%, due to the low-temperature (<100 °C) processed ZnO dense layer exhibits low mobility and high defect density. Liu *et al*. improved the preparation method of ZnO[41]. They spin-coated the mixed solution of butanol/chloroform containing ZnO nanoparticle dispersion directly on ITO substrate without calcination. The PCE of corresponding FPSC was over 10% as shown in Fig. 4(b). Heo *et al*. increased the heat treatment temperature of the electron transport layer to 150 °C, resulting in a more compact ZnO layer. Fig. 4(c) shows that the PCE is increased to 15.6%.

Although the ZnO has excellent electronic properties, there are hydroxyl or residual organic acetate ligands on its surface, which leads to charge recombination at the interface and poor quality of perovskite deposits. In addition, there is proton transfer reaction between ZnO and perovskite, which leads to decomposition reaction of interface[43–48]. Therefore, Jin *et al*. used low-temperature atomic layer deposition (ALD) on the surface of ZnO to obtain TiO_2. This ultra-thin TiO_2 protective layer can effectively passivate surface defects and prevent decomposition reactions. Figs. 4(d)–4(f) show that as the number of ALD cycles (*x*) increases, the thickness of the TiO_2 layer increases, and the PCE based on a rigid substrate reaches a maximum PCE of 18.26% at *x* = 60. The PCE of FPSC based on this technology has increased to 17.11%[42]. In addition to ZnO electron transport materials, researchers also pay great attention to the development of low-temperature treatment of TiO_2 electron transport layer for FPSC.

Compared with ZnO, TiO_2 has better environmental stability and better perovskite compatibility. However, the most efficient TiO_2 electron transport layer needs to be obtained through an annealing process at 450 °C. In 2013, Doampo *et al*. first tried to use TiO_x for p–i–n structured FPSC[49]. The TiO_x precursor solution is composed of titanium isopropoxide and isopropanol solution, and the TiO_x flat film is annealed at 130 °C. The corresponding PCE of FPSC is 6.4% (as shown in Figs. 5(a) and 5(b)). So far, a variety of low-temperat-
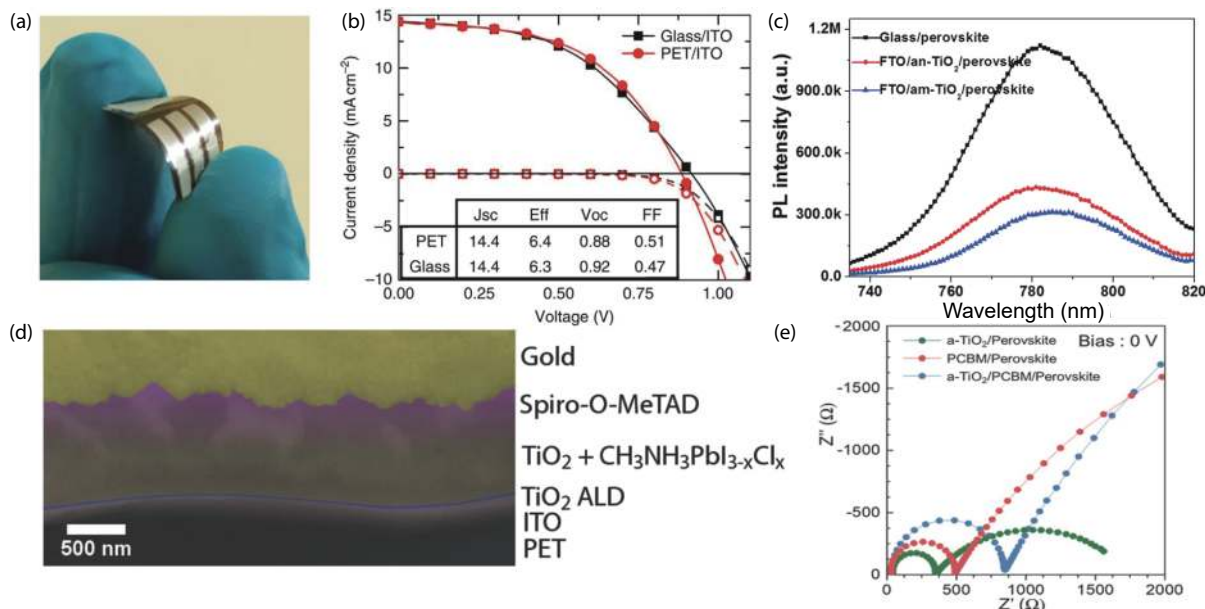
Fig. 5. (Color online) (a) The first FPSC based on the TiO$_2$ electron transport layer and (b) its *J–V* curve as the FPSCs performance of the electron transport layer[49]. (c) Steady-state PL spectra of glass/perovskite, FTO/anatase-TiO$_2$/perovskite and FTO/amorphous-TiO$_2$/perovskite film[50]. (d) FPSC cross-section scanning electron microscope with ALD deposited TiO$_2$ dense layer and UV-irradiated mesoporous TiO$_2$[51]. (e) Impedance diagram (*Z''– Z'*)[57].

ure processing technologies for TiO$_2$ have been developed, such as sputtering, atomic layer deposition (ALD), and vacuum evaporation deposition. Yang *et al*. sputtered an amorphous TiO$_2$ film by DC magnetron at room temperature, and this film has good solar spectral transmittance[50]. The steady-state photoluminescence (PL) shows that compared with the anatase-TiO$_2$/perovskite film, the PL intensity of the amorphous-TiO$_2$/perovskite film is weaker, indicating that the charge transfer effectively occurs before the carrier recombination at the interface. And amorphous-TiO$_2$ improves the extraction of electrons from the absorption layer to the electron transport layer (as shown in Fig. 5(c)). Giacomo *et al*. used plasma-enhanced atomic layer deposition (PEALD) to deposit a dense TiO$_2$ film on a flexible PET/ITO substrate at low temperature, and then spin-coated or screen-printed to deposit mesoporous TiO$_2$[51]. The cross-section of the obtained device is shown in Fig. 5(d), and the PCE is 8.4%. The prepared photovoltaic module with an area of 8 cm^2 showed a PCE of 4.3%. In addition, deposition of TiO$_2$ colloidal particles and subsequent low-temperature annealing or ultraviolet treatment are also effective methods for forming the TiO$_2$ electron transport layer[52, 53]. Deep charge traps at the interface between TiO$_2$ and perovskite will cause TiO$_2$-based PSCs to exhibit severe hysteresis. In addition, due to the photocatalytic effect of TiO$_2$, deep traps will be introduced on the surface[54–56], the passivation interface layer is necessary. Kim *et al*. coated PC$_{60}$BM on PEALD-deposited TiO$_2$ to passivate traps and improve electron extraction, and finally achieved the highest efficiency of 17.70%, while significantly reducing the hysteresis[57]. The FPSC was analyzed by electrochemical impedance spectroscopy (EIS) in Fig. 5(e), where the first semicircle in the high frequency range (> 1 kHz) can interact with the dielectric response of the perovskite layer (*C$_g$/R$_2$*) is related, and the second semicircle in the low frequency range (≈ 1 Hz) can be due to the surface charge or ion charge accumulation at the interface causing electrode polarization (*C$_s$/R$_1$*)[58]. The capacit-

ance difference shown in the low frequency range can be seen that TiO$_2$/PCBM/Perovskite has a smaller capacitance.

Compared with TiO$_2$, SnO$_2$ has a deeper conduction band and higher electron mobility. It is easy to process by solution method at low temperature, and it is more stable under illumination[59]. The investigations on SnO$_2$ as electron transport layer are mainly focusing on how to improve the conductivity and mobility and modify the conduction band to enhance the electron extraction and transmission in FPSCs. Park *et al*. used Li-doped SnO$_2$ as the electron transport layer, and the prepared FPSCs had a high PCE of 14.78%. Li-doping could reduce the conduction band and increase the conductivity of SnO$_2$ layers, which will facilitate electron extraction and transport[60]. Wang *et al*. employed PEALD SnO$_2$ treated with water vapor as electron transport layer to improve the photovoltaic performance of FPSC. It was found that the water vapor treatment at 100 °C can improve the electrical conductivity and mobility of SnO$_2$, because annealing with water vapor can promote the complete reaction of organic materials and the formation of pure SnO$_2$. Based on that, PCE up to 18.36% was achieved in the devices[61]. Liu *et al*. reported a similar strategy to improve the quality of SnO$_2$ by hydrothermally annealing the SnO$_2$ layers. Huang *et al*. precisely controlled the deposition of SnO$_2$ layer by adjusting the solution concentration to form a fully covered SnO$_2$ layer. The best PCE of FPSCs based on SnO$_2$ was increased to 19.51%[24].

Zn$_2$SnO$_4$ is a new type of ETL for high-efficiency FPSC applications. Shin has developed a new route to synthesize highly dispersed Zn$_2$SnO$_4$ by the introduction of a Zn–N–H–OH complex derived from hydrazine via simple solution process at low temperature (90 °C)[20]. The refractive index of Zn$_2$SnO$_4$ is low (about 1.37), which shows a transmission increase of about 20% in the entire visible light range, due to the anti-reflection effect. The PCE of FPSC based on Zn$_2$SnO$_4$ with the employment of MAPbI$_3$ as the absorber layer is 15.3%. On this basis, they introduced Zn$_2$SnO$_4$ quantum dots

on top of the Zn$_2$SnO$_4$ nanoparticles layer. Among them, the relatively large-sized nanoparticles (>10 nm) have a higher work function, which is beneficial to the electron transfer at the interface between the electron transport layer and the ITO. The Zn$_2$SnO$_4$ quantum dots are favorable for generating large built-in potential at the Zn$_2$SnO$_4$ quantum dots/perovskite interface, and the combination of the quantum dots and nanoparticle reduces the energy barrier. Accordingly, based on Zn$_2$SnO$_4$, PCE of FPSC reached 16%[62].

In addition to all these well-developed inorganic ETLs, Yoo *et al.* also demonstrated a simple interfacial layer that replaces TiO$_2$ with C$_{60}$ with the aid of polyethyleneimine ethoxylation (PEIE)[31]. In addition to serving as an electron collecting interfacial layer, the PEIE layer also helps to C60 processing and protect them from corrosion during the subsequent solution spin coating process. Through this low-temperature-treated double-layer ETL, FPSC with a PCE of 13.3% was demonstrated. Interestingly, the solid ionic liquid 1-benzyl-3-methyl-imidazolium chloride was also used as ETL and processed at low temperature. The resulting device achieves 16.09% efficiency in FPSC with no hysteresis, and good repeatability[21].

The various electron transport layers described above are all prepared under low temperature processes. Among them, the widely used inorganic electron transport layer materials TiO$_2$, SnO$_2$ and ZnO have been successfully developed with low temperature preparation processes. ZnO, which is the most readily developed FPSCs electron transport layer material, is limited for further application by the decomposition reaction at the interface. To solve this problem, interface passivation, e.g. using PCBM or TiO$_2$, become effective solution[42, 63]. TiO$_2$ is often limited by the relatively poor materials quality obtained from the low-temperature preparation process, and the efficiency of resulting device has only developed to 17.7%[57]. At present, modification on low-temperature preparation methods, including electron beam evaporation, magnetron sputtering, atomic layer deposition and liquid phase method, seems to be meaningful to improve the materials optoelectronic properties. SnO$_2$, which is easily obtained by solution processing at low temperatures, stands out and develops rapidly with respect to the device efficiency, and is currently the most widely used one.

3.2. Hole transport layer

The hole transport layer (HTL) in FPSC is essentially to have hole extraction and hole transport capability, as well as bending durability. In the n–i–p structure, the hole transport layer does not need to be transported because that it does not hinder the light absorption of the perovskite. Spiro-OMeTAD, which is widely used in traditional n–i–p structured PSCs, can provide excellent performance[1, 17, 23, 24, 64, 65]. For p–i–n-structured FPSCs, the hole transport layer requires high optical transparency. The NiO$_x$ with superior hole transport performance, high optical transparency, and low-temperature manufacturing processes is currently the most promising materials. By introducing low-temperature processed (130 °C) NiO$_x$ HTL, better energy alignment can be achieved, and the device efficiency based on the PET/ITO substrate is 13.43%[65]. Interestingly, as shown in Fig. 6(a), the deposited NiO$_x$ nanoparticles (NPs) can also be processed at room temperature without any other post treatment. By introducing a defect-free NiO$_x$ nanostructured films, the PCE of 14.53% can be achieved with negligible hysteresis[66]. Generally, the original NiO$_x$ has low conductivity, and the thickness needs to be carefully optimized during the device fabrication process. Li and his colleagues developed Cu-doped NiO$_x$ NP as HTL to improve its conductivity as shown in Fig. 6(b), thereby enhancing the PCE of FPSC to 17.16%[67].

Docampo *et al.* used PET/ITO/poly(3,4-ethylenedioxythiophene):polystyrene sulfonate (PEDOT:PSS)/MAPbI$_{3-x}$Cl$_x$/PCBM/TiO$_x$/Al device structure to obtain the first FPSC[49]. The device based on this structure has a PCE of 6.4%. The commonly used PEDOT:PSS HTL show some disadvantages, such as strong acidity and high water absorbability. In addition, it cannot provide perfect hole extraction ability when contact with perovskite (energy alignment of hole transfer: PEDOT:PSS: ≈ 5.0 eV; MAPbI$_3$: ≈ 5.4 eV), which will cause voltage loss in the device. Besides, polymer hole transport materials copolymerized by 1,4-bis(4-sulfobutoxy) benzene and thiophene group (PhNa-1T) have been successfully developed to replace PEDOT: PSS as HTL. The structure of PhNa-1T is shown in Fig. 6(c). Because PhNa-1T can effectively reduce the potential energy loss at the HTL/perovskite interface and achieve better contact (as shown in Fig. 6(d)), it enhances the extraction of holes from the perovskite to the HTL and inhibits interfacial recombination. The FPSC with PhNa-1T HTL achieved a PCE of 14.7% with increased V_{oc}[68].

Poly[bis(4-phenyl) (2,4,6-trimethylphenyl) amine] (PTAA) is often used as HTM layer, since the film can be obtained by low-temperature spin coating that is compatible with FPSC preparation. Moreover, PTAA are of high conductivity and reliable durability. Unlike Spiro-OMeTAD, which requires doping to improve performance, undoped PTAA still performs well in high-efficiency PSC. For example, Qiu *et al.* prepared an FPSC based on the PET/ITO/TiO$_2$/MAPbI$_{3-x}$Cl$_x$/PTAA/Au structure in 2015 and achieved a PCE of 13.5%[69]. In 2017, Huang *et al.* used PTAA and double fullerene layers (phenyl-C61-butyric acid methyl ester (PCBM) and C60) as hole transport layer and ETL, respectively. In this architecture, the charge transfer from the perovskite to the PTAA layer is fast, and the efficiency of the prepared FPSC has reached 18.1%[70].

4. High-quality perovskite films

As the light absorption layer, the quality of the perovskite film directly affects the performance of the flexible perovskite solar cells. Compared with the perovskite film prepared on the glass substrate, the quality of the perovskite film on the flexible substrate is different, which is limited by the high roughness and high thermal resistance of the flexible substrate. Because the resistance of the most widely used low-cost flexible polymer substrate will rise sharply when the temperature is higher than 150 °C, the development of flexible perovskite solar cells mainly focuses on the preparation of perovskite thin films at low temperature.

4.1. Perovskite film preparation method

4.1.1. Spin-coating

The quality of the perovskite film on the flexible substrate is generally not satisfactory because of the poor wettability and high roughness of the surface of the flexible substrate. Choosing an appropriate preparation process to control the nucleation and crystallization process of the film is
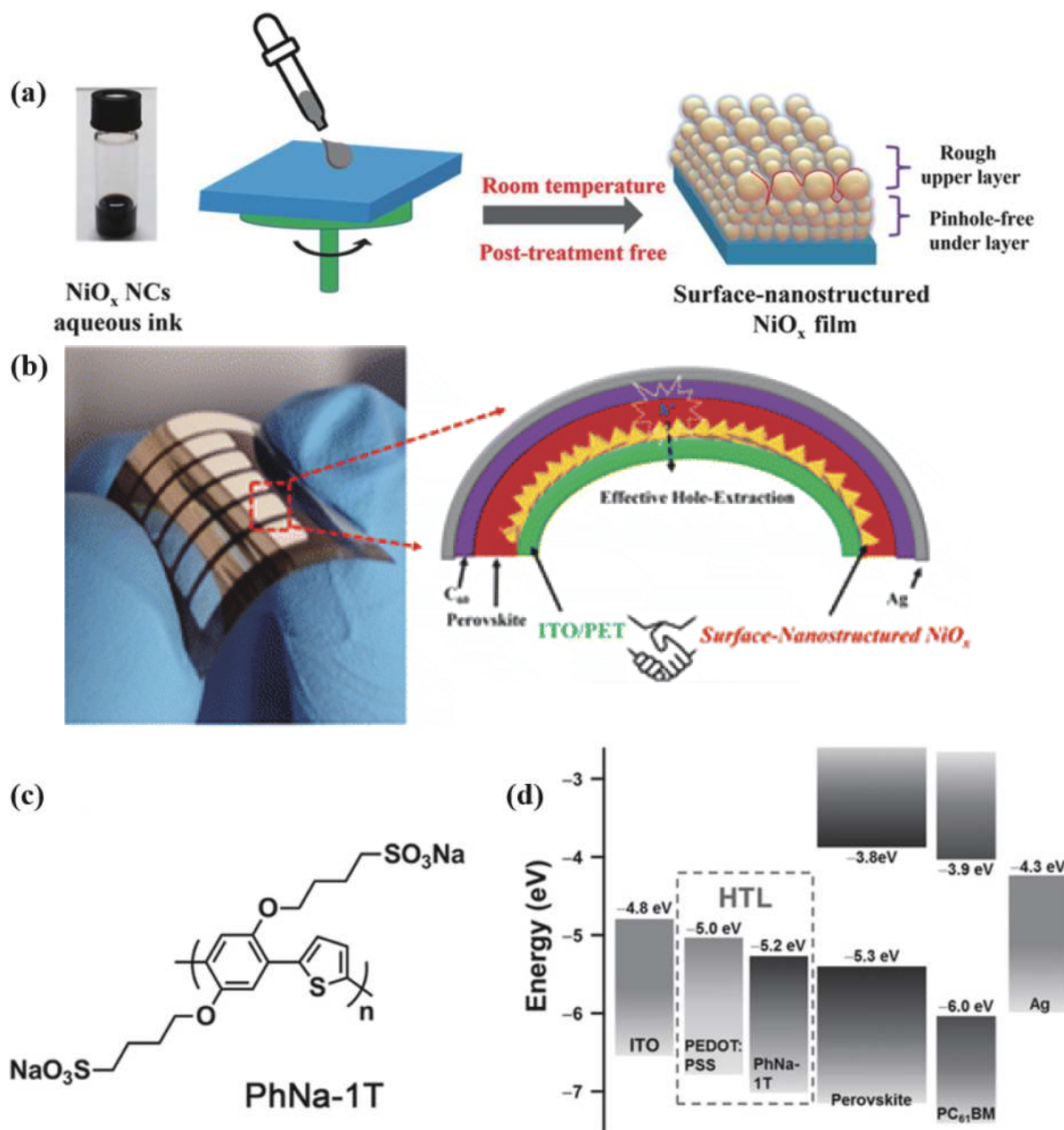
Fig. 6. (Color online) (a) Schematic diagram of the fabrication of nanostructured NiO$_x$ thin films[65]. (b) Cu-doped NiO$_x$ FPSC device structure[67]. (c) PhNa-1T structure diagram (d) Energy band diagram using different hole transport layers[68].

the effective way to obtain a high-quality film. As a simple and effective method, the spin coating process can be adopted to the preparation of small-area devices and are able to obtain perovskite films with a wide range of thickness. In general, there are one-step and two-step spin coating methods.

In the one-step deposition method, the mixed precursor solution containing MA/FA halides and metal halides is directly deposited onto the desired substrates through a one-step spin-coating procedure. The sample is then annealed at low temperature (100–150 °C) to produce a perovskite film. The one-step spin coating process usually involves an anti-solvent. Cheng *et al.* added chlorobenzene (CB) as an effective anti-solvent during the spin-coating of perovskite in N, N-dimethylformamide (DMF) to prepare a highly uniform perovskite layer[71]. Seok's team used toluene (TB) as an anti-solvent to prepare perovskite in a mixed solvent of dimethyl

sulfoxide (DMSO) and gamma-butyrolactone (GBL)[72]. Considering the precise control of the step of adding anti-solvent, this method is more challenging to produce large-area PSCs. On the other hand, due to better morphology and interface control, the two-step method is better than the one-step method. However, some problems in the two-step method, such as the incomplete conversion of PbI$_2$ to perovskite, severely limit its large-scale application in high-efficiency PSCs with good reproducibility. Although this method is an anti-solvent-free process, there will be edge effects, and the larger the area, the more serious the impact.

4.1.2. Scalable deposition method

Spin coating can effectively be used to produce a uniform perovskite layer on a small area, but this method is difficult to obtain a large area of uniform perovskite. In order to fabricate large-scale flexible perovskites, it is particularly im-

portant to use scalable printing technology to deposit perovskite absorbers. Among them, blade coating has become one of the promising laboratory technologies due to its easy handling and less waste of raw materials. In 2015, Jen et al. used a blade coating method to sequentially deposit poly(3,4-ethylenedioxy-thiophene): poly(4-styrenesulfonate) (PEDOT:PSS)/CH$_3$NH$_3$PbIXCl$_{3-x}$/[6,6]-phenyl-C$_{61}$-butyric acid methyl ester (PC$_{61}$BM)/Bis-C$_{60}$, the Ag was used as the top electrode in the device. The prepared FPSC exhibited a PCE of 7.14%[73]. Huang et al. deposited the perovskite film by gas-assisted blade coating in the ambient environment at room temperature[38]. As shown in Fig. 7(b), the air blade moving with the blade blows N$_2$ gas on the coated wet film at the same time to remove the solvent and induce crystallization. The PCE results are shown in Figs. 7(c) and 7(d). They achieved 19.72% PCE on a small area, and at the same time obtained 15.86% PCE on a 42.9 cm^2 large area module. However, their devices are made on corning willow ultrathin glass, which is limited in flexibility and cost. Wang et al. deposited the perovskite which is added with Lewis base thiourea (TU) on the PEN/ITO polymer substrate by blade coating[74]. As shown in Fig. 7(e), they have improved the interface design and adopted a double hole transport layer (PEDOT:PSS and poly[bis(4-phenyl)(2,4,6-trimethylphenyl)amine] (PTAA)). On the one hand, it achieves more effective carrier extraction (as shown in Figs. 7(f) and 7(g)), on the other hand, it enhances the flexibility of the ITO substrate. This interface optimization design increases the device efficiency of FPSC to 19.41%.

Vacuum thermal evaporation is a relatively mature technology that can deposit large-area and uniform thin films, and its relatively low temperature characteristics can also be compatible with the preparation process of FPSCs[7, 75–78]. Wu et al. developed a full-vacuum strategy, using a two-step flash evaporation method to prepare perovskite films. First, PbI$_2$ film and MAI were vapor-deposited sequentially[79]. Meanwhile, the post-annealing process was added to realize the "solid-solid" reaction of the film, and the PCE of 13.15% was realized on the 16 cm^2 flexible perovskite solar module. However, the current vacuum evaporation still has some issues such as difficulty in detecting organic sources and high vacuum requirements.

In perovskite roll-to-roll deposition compatible technology, gravure printing is continuous and stable. Seo et al. obtained a FAPbI$_3$ film by gravure printing and soaked it in tert-butanol (tBuOH) to obtain a more uniform and highly crystalline film. The FPSC prepared entirely using the roll-to-roll process showed the best PCE of 13.8%[80].

The combination of slot-die coating and roll-to-roll process exhibits advantages, including less material waste, good film thickness control, one-dimensional patterning suitable for tandem interconnected cells without the need for a large number of patterning processes, and high manufacturing speed[81, 82]. Gao et al. used a blow-assisted drop-casting method compatible with roll-to-roll to prepare CH$_3$NH$_3$PbI$_3$ films. The formulation was optimized and applied to the flexible substrate, so that 11.16% of PCE was obtained through slot-die coating[83].

4.2. Composition engineering

Composition engineering has been proven to be an effective strategy to improve the quality of perovskite films. In 2017, Huang et al. proposed that the deposition conditions of the perovskite on the rigid substrate need to be optimized on the flexible substrate to improve the film morphology[70]. They proposed that the difference of the perovskite film quality may be caused by the difference in the thickness, surface roughness and thermal conductivity of the rigid glass/ITO relative to the flexible PET/ITO substrate. They adopted a method of regulating the ratio of precursors (PbI$_2$: FAI), using a non-stoichiometric solution of 1 : 0.95. The reduced organic precursor content can form a uniform and pinhole-free perovskite film on the PET/ITO/PTAA substrate, and has a longer carrier lifetime, a smaller trap state density, and a reduced precursor residue. After improving the film morphology and optoelectronic properties of perovskites, the PCE on the flexible ITO/PET substrate is 18.1%.

4.3. Additive engineering

The grain boundaries of perovskite, often as charge carrier recombination centers and ion migration channels, are currently an important factor affecting the efficiency and stability of PSCs[84]. The additives can effectively improve the crystalline quality of the film. Feng et al. used dimethyl sulfide (DS) as the additive to control the growth of perovskite films deposited on flexible substrates[23]. Fourier transform infrared spectroscopy shows that the DS additive can react with Pb^{2+} and form an intermediate phase, which delayed the crystallization process. The prepared perovskite films exhibit large grains and low defect densities, and the PCE is improved to 18.4%. Wu et al. used NMP and MACl as synergistic additives to optimize FAPbI$_3$ perovskites. The introduction of NMP can form FAI·PbI$_2$·NMP intermediate, which delayed the crystallization process. The employment of MACl can facilitate the phase transition of perovskites at low temperature. Based on the collaborative improvements, the FAPbI$_3$ based FPSCs showed a PCE of 19.38%[64].

Meng et al. incorporated a self-healing polyurethane (s-PU) with dynamic oxime-carbamate bonds in the grain boundaries of perovskite films. The s-PU could release the mechanical strain and repair cracks at the grain boundaries, which significantly improved the stretchability and deformability of perovskite films. The prepared FPSCs could recover 88% of initial PCEs after 1000 cycles at 20% stretching. In addition, due to the defect passivation of s-PU at the grain boundaries, the stretchable FPSCs exhibited a best efficiency of 19.15%[85].

5. Electrode

In order to absorb sufficient incident light, one of the two electrodes in the solar cell device should be transparent, while the other is usually opaque. For opaque electrodes, the metal foil conductive substrate has been introduced above. In addition, the evaporated thick metal film and printed carbon layer have also been mature and widely used. They possess the characteristic including good electrical conductivity, easy manufacturing, and appropriate energy levels that aligned with other functional layers of the device. In the transparent electrode, it is necessary to have the characteristics of high transparency, high conductivity, mechanical flexibility and chemical stability. ITO, which is widely used in glass substrates, has also been used in FPSCs. Apart from this, some new transparent electrodes, such as conductive polymers, ultra-thin metal films, Ag nanowires based electrodes, carbon
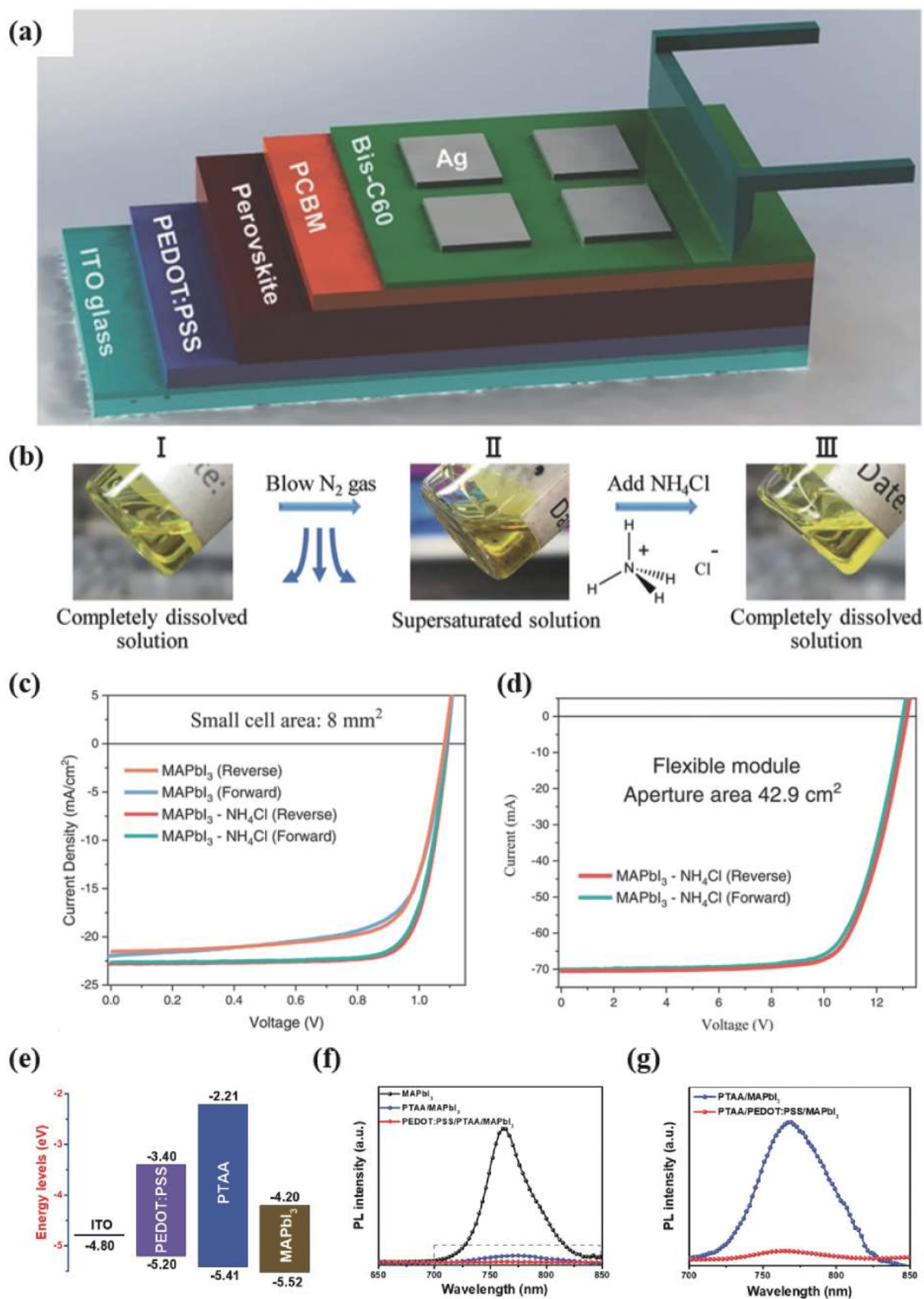
Fig. 7. (Color online) (a) Schematic diagram of the device prepared by blade coating method[73]. (b) Blow N_2 gas and precursor solution with the addition of NH_4Cl[38]. (c) J–V curve under an area of 8 mm². (d) J–V curve under an area of 42.9 cm². (e) Double hole transport Energy band diagram. (f, g) Under the layer $MAPbI_3$, PTAA/$MAPbI_3$ and PEDOT:PSS/$MAPbI_3$ diagram of PL and its partial enlargement[74].

nanotubes and graphene, have also been explored.

5.1. Indium tin oxide (ITO)

In the transparent electrode, ITO is the most commonly used material in FPSC, mainly because it is easy to obtain, and has good transmittance and conductivity. However, the high cost and high mechanical rigidity of ITO still plague the development of FPSCs. According to the published work, FPSCs based on ITO-PEN lose about 50% of the initial photoelectric conversion efficiency after a bending radius of 4 mm for 1000 cycles[86]. In order to explore the origin of the degradation during the period, they compared the resistance of two flexible devices (PEN/ITO/TiO$_x$/perovskite/Spiro-OMeTAD and PEN/TiO$_x$/perovskite/Spiro-OMeTAD) after bending. As shown in Fig. 8(a), on the PEN/ITO substrates, a sharp increase in resistance was observed at the beginning. This is because the Young's modulus of the ITO and the substrate are different, which causes the ITO layer coated on the flexible substrate to form large crack lines perpendicular to the bending direction (as shown in Figs. 8(b) and 8(c)), resulting in a sharp increase in the resistance of the FPSC.

In order to overcome the poor mechanical strength of ITO, some organic molecules or polymer scaffolds are used to improve the interface contact. Inspired by the flexible structure of vertebrae, Meng et al. designed an adhesive polymer interface layer (PEDOT:EVA, Poly(3,4-ethylenedioxythiophene): poly(ethylene-co-vinyl acetate)) (As shown in Fig. 8(d))[25]. The brittle ITO and perovskite are tightly bonded together due to the viscosity of EVA in the PEDOT:EVA film, so the flexibility of the flexible substrate is improved. The PEDOT:EVA ink is synthesized by miniemulsion method, which has good dispersion and stability. Finally, the FPSC prepared by meniscus-coating has a PCE of 19.87%. As shown in Figs. 8(e) and 8(f), when the bending radius is 10, 5, or 3 mm, after 7000 cycles, it can still maintain about 96%, 95%, and 85% of the initial PCE, respectively.

5.2. Conductive polymer

PEDOT:PSS composed of conductive polymer PEDOT and water-dispersible PSS:PSS is by far the most successful and most widely used conductive polymer. PSS has the dual effects of doping charge and improving water solubility[87]. Therefore, the conductivity of PEDOT:PSS can be adjusted by the ratio of PEDOT and PSS[88]. PEDOT:PSS film has excellent visible light transmittance and mechanical properties, making it very suitable for transparent conductive electrodes of flexible solar cells. For example, spin-coated PEDOT:PSS onto a Noland Optical Adhesive 63 substrate or an ultra-thin PET substrate as a transparent conductive electrode to obtain a device with a PCE higher than 10%[89]. The prepared ultra-thin and lightweight devices have excellent flexibility and stretchability, and its power per unit weight is calculated as high as 23 W/g.

5.3. Ag-nanowire-based composites

Ag nanowires and grids have attracted widespread attention as alternative materials to ITO. Ag nanowire networks and Ag grids have good electrical conductivity, light transmittance and mechanical flexibility, which are used as transparent conductive electrodes in many optoelectronic devices. In FPSC, they are used solely or together with other materials as top or bottom transparent conductive electrodes. For ex-

ample, a bare Ag nanowire grid is used as the top transparent conductive electrode of FPSC[30]. Since silver nanowires are used as bottom electrodes, their rough surfaces usually hinder the improvement of device performance, so some other transparent conductive components can be combined to prepare a composite electrode with a smooth surface, such as doped Fluorine-based zinc oxide[90], graphene oxide[91], and PEDOT[92]. Among them, the silver grid/PEDOT:PSS composite electrode can be simply fabricated in an embedded Ag-meshing flexible PET substrate by spin coating[93]. Accordingly, the sheet resistance of this composite is as low as 1.2 Ω per square inch. The obtained FPSCs showed a relatively high PCE, with almost no decrease even at 2 mm bending radius. Based on the excellent flexibility of the device, even after 1500 bending cycles with a bending radius of 5 mm, PCE can remain stable, maintaining 95.4% of its initial value after 5000 cycles. For comparison, PET/ITO-based devices showed a significant performance degradation after 100 bending cycles. At the same time, FPSC also shows relatively good storage stability. After 500 h of storage in an inert atmosphere at room temperature, PCE can maintain an initial value of 91.6%, which is better than a rigid control device. However, Ma et al. also reported conflicting result about stability in another report. They found that the device based on the PH1000/Ag electrode is unstable due to the rapid reaction between Ag and the perovskite precursor solution caused by the redox reaction between Ag and PH1000[94]. While, the ammonia water and PEI modified layer on PH1000 can inhibit this redox reaction and effectively improve device performance.

5.4. Carbon nanomaterials

Carbon nanomaterials have been used in rigid PSCs in different forms (such as carbon black/graphite composites, carbon nanotubes (CNT) and graphene) as back electrodes, partly because of their low cost and good electrical conductivity. Extensive research on carbon nanotubes and graphene electrodes is mainly based on their excellent properties, including good chemical stability, mechanical flexibility, high electrical conductivity and visible transparency. In 2014, the laminated CNTs network has been used as both the top electrode and the hole transport material in rigid PSC[95]. For FPSCs, Wong et al. transferred the CNT film grown by chemical vapor deposition (CVD) to the top of the perovskite film as a transparent conductive electrode and a hole collector, in which a Ti foil substrate is used as bottom contact[96]. In fact, CNT networks can also be directly transferred to transparent plastic substrates, where they are used as bottom transparent conductive electrodes, similar to ITO and FTO. Matsuo et al. reported that the primary PCE of the FPSC device was 5.38%, and this efficiency was further improved to 12.8% by the same group in 2017, partly due to the optimization of the device structure and the quality of the functional layer[97]. Compared with ITO-PEN control sample, CNT-based FPSC also has higher mechanical strength. Carbon nanotubes are also used to fabricate top electrodes with ITO-PEN as the bottom substrates, and their stability is much higher than that of control devices based on silver electrodes[98]. In addition, CNTs are also used as transparent conductive electrodes in irregular stretchable solar cells (such as fiber cells)[99].

Graphene was first used as an interfacial layer in FPSC with an ITO-PET substrate in 2015[100]. Later, Yan et al. used
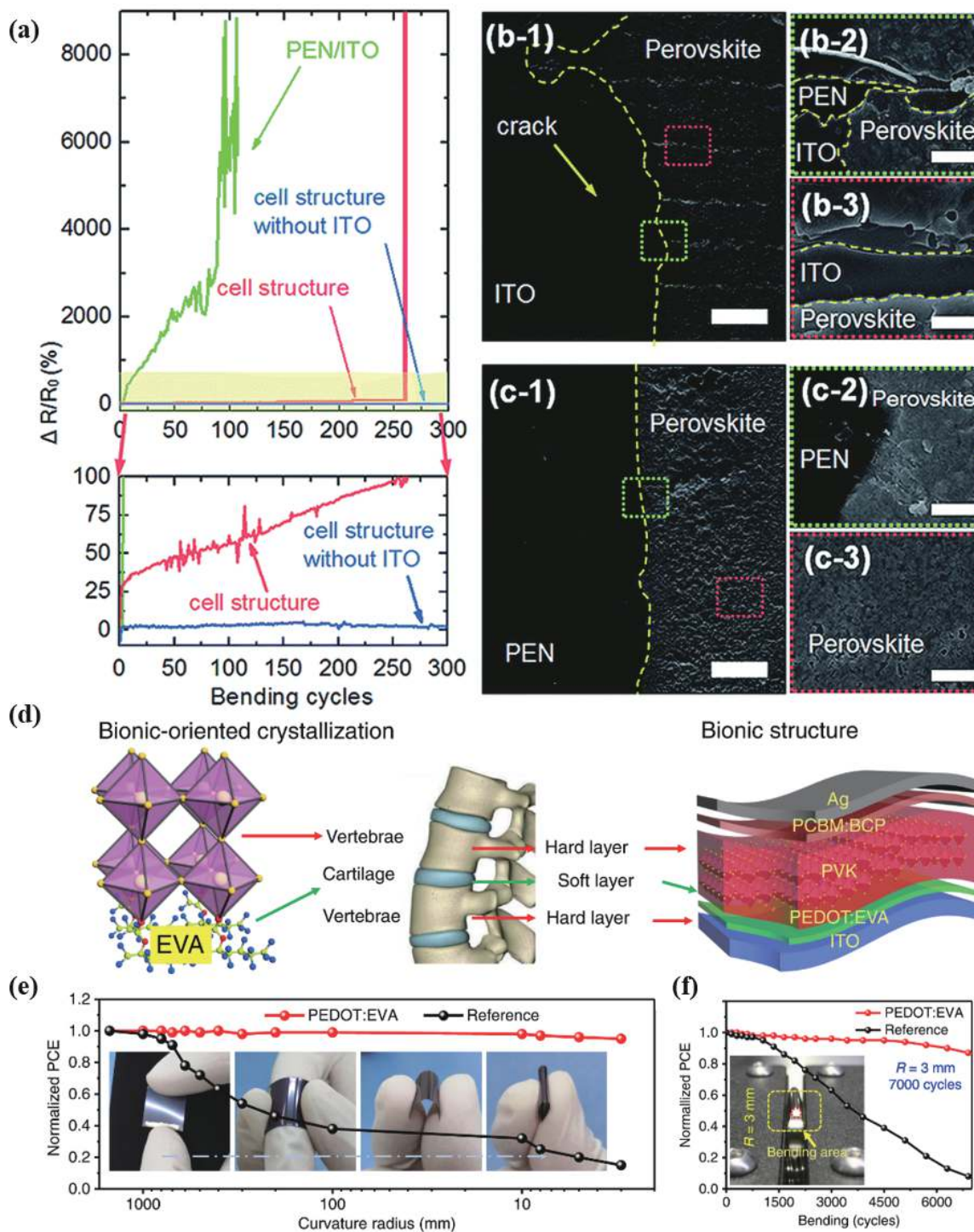
Fig. 8. (Color online) (a) Resistance change of multilayer structure with bending cycle ($\Delta R/R_0$ (%)). (b, c) Low-magnification SEM images of PEN/ITO/TiO$_x$/perovskite and PEN/TiO$_x$/perovskite after 300 bending cycles, scale bar: 100 μm[86]. (d) The bionic mechanism of vertebrae and FPSCs. (e) PCE of FPSC after 500 cycles of bending at different bending radii. (f) The average PCE value of FPSC with a bending radius of 3 mm and bending 7000 cycles[25].

graphene as the transparent conductive electrode of FPSC, in which they grow a single graphene on a copper substrate by CVD and then transfer it to a PET substrate[101]. The PCE of the resulting device is 11.5% with high flexural durability. Doping strategies are often used to improve the conductivity of graphene. Choi evaporated 2 nm MoO$_3$ on graphene to induce hole doping of graphene, and achieved 16.8% PCE in the resulting FPSC device[22]. FPSCs based on all-carbon electrodes were also realized by Guo *et al*. The graphene film

formed on the PET substrate is used as bottom transparent conductive electrode, while the cross-laminated CNT film is used as the top counter electrode[102]. The all-carbon electrode device has excellent flexibility. Even after bending with a radius of curvature of 4 mm for 2000 times, the device retains approximately 84% of its original PCE. After 200 bends at a radius of curvature of 2.2 mm, it still remains 85% of the initial PCE. As a comparison, a device with a PEN/ITO/TiO$_2$/PCBM/CH$_3$NH$_3$PbI$_3$/Spiro-OMeTAD/gold structure loses 87%

of its initial PCE with a radius of curvature of 4 mm, and is almost completely damaged when bent to 2.2 nm. The results show that the performance degradation of the reference device is mainly caused by the conductivity loss in the brittle ITO layer. The slight degradation of the all-carbon electrode-based device is most likely due to fractures in the crystalline perovskite layer and delamination in the device during its deformation. In addition, the all-carbon electrode device exhibits excellent stability, and it retains about 90% of the original efficiency after 1000 h light or 1570 h heat treatment in the dark and air at 60 ℃, respectively.

6. Encapsulation methods

While the PCE is a crucial criterion to evaluate the performance of a PSC, the lifetime is also important standard since perovskite materials are very sensitive to many stressors, e.g. moisture, oxygen, light, heat, etc.[87, 103]. In this context, device encapsulation is necessary. However, some well-established encapsulation techniques for rigid PSCs, e.g., attaching a glass plate onto a device by thermosetting epoxy, cannot be readily used in FPSCs. Therefore, new encapsulation materials and methods are required to be developed.

As an efficient encapsulation material, various factors need to be considered: light transmission, oxygen transmission rate (OTR), water vapor transmission rate (WVTR), resistance to ultraviolet (UV), chemical inertness, mechanical flexibility, and strength, etc.

The combination of polyurethane resin encapsulation and Cr_2O_3-Cr interlayer proves that the air stability of ultra-thin FPSC has been improved. In particular, it was found that the use of a chromium oxide-chromium intermediate layer can effectively protect the metal top contact from the perovskite reaction, which is beneficial to prolong the long-term stability[104].

Nanocone PDMS has anti-reflection and waterproof functions and is attached to the front of the flexible substrate. These nanocone PDMS packaging materials help improve optical transmittance and achieve waterproof effects[37]. The multi-layer encapsulation film composed of a superhydrophobic layer and a relatively hydrophilic layer significantly enhances the stability of the PSC in a very humid environment[109, 110]. The superhydrophobic film containing polymethylmethacrylate (PMMA), polyurethane (PU) and SiO nanoparticles is in contact with the environment, which can repel water in a humid environment and prevent water from penetrating into the fragile perovskite layer. Among them, the moderately hydrophilic PMMA layer acts as a desiccant, extracting residual water from the perovskite layer itself during the operation of the solar cell. When exposed to humidity exceeding 95%, the dual function of the coating film helps the PSCs to keep at 17.3% PCE for 180 minutes[110].

7. Conclusion and outlook

Organic–inorganic halide perovskite has become a promising candidate for flexible solar cells due to its excellent optoelectronic performance, excellent mechanical tolerance and low-temperature solution processability. Facilitated by the endeavor on exploration of flexible substrate, charge transport layer, high-quality perovskite film and transparent top electrode, the highest PCE of flexible PSCs reached 21.1%.

For the flexible substrates, the polymer substrates based FPSCs show the best PCE, but bring to poor environmental stability. Metal substrates, ultra-thin flexible glass substrates and other special materials, etc. have been explored to improve the device stability.

For the charge transport layer in FPSCs, the main focus is developing low-temperature process approach. At present, SnO_2 is the best choice for electron transport layer in n–i–p structure due to its better stability and low-temperature processability.

For perovskite films, efforts have been devoted to the preparation of dense, uniformly covered, and high-quality films on flexible substrates. In terms of thin film growth, the spin coating is suitable for small-area device, but limited for large scale production. Component and additive engineering are indispensable in thin film fabrication, which can effectively passivate defects and improve crystal crystallization. Also, the band structure and mechanical property of perovskite layers can dramatically influence the photovoltaic performance of the resultant PSCs under mechanical strain.

For transparent conductive electrodes, ITO is the most common choice, but silver nanowires and carbon materials also become promising alternatives. Benefiting from its better mechanical flexibility, FPSCs based on silver nanowires and carbon materials show better bending stability.

The further development of FPSCs is discussed in the following aspects. 1) Preparation of large area solar cell modules. So far, the reported FPSCs with excellent PCE are all prepared by spin coating, and the effective area is small. With the increase of device area, more stringent requirements are posed on the quality of the perovskite film, and the density and uniformity of other functional layers (carrier transport layer and perovskite layer) are also crucial. While methods such as doctor blade coating, thermal deposition, and gravure printing can be selected for large-scale production, the roll-to-roll manufacturing technology may be an effective way to develop large-area FPSCs. 2) The environmental instability is also the most challenging issue for the development of FPSCs. In addition to the device degradation under heat, light, and electricity stressors, the polymer substrate of FPSCs accelerates the permeation of water and oxygen, which further brings to water and oxygen instability. It thus requires more robust encapsulation approach to fabricate FPSCs with improved long term stability. 3) In addition to improve the PCE, it is also necessary to pay attention to mechanical stability of FPSCs, since the application scenarios of FPSCs include wearable devices. Exploring flexible transparent electrodes with high conductivity and transmittance, low cost, and scalable processability is still an urgent need to meet the commercialization goal.

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