Advanced Perovskite Solar Cells

Yuqin Tian, Can Zhu, Kun Hong, Kai Qiu, and Renhui Zhang

Abstract The energy crisis is a huge challenge facing the world today. Natural resources such as coal and oil are consumed in large quantities and their reserves are gradually decreasing. It is imperative to advocate energy conservation. Meantime, it is very important to develop green and clean energy. Solar energy has become one of the most promising green energy sources in recent years because of its sustainable and safe advantages. Solar energy can be converted into effective energy such as heat energy through photoelectric conversion because it doesn't produce harmful gases, solid waste, and other pollutants in the conversion process. In addition, the new solar cells have the advantages of low cost, cleanliness, and they are efficient. Since 2009, a new type of perovskite solar cell has developed rapidly. In order to further improve the photoelectric conversion efficiency of batteries, more researchers in recent years have tried to apply new ceramic materials (perovskite materials) to batteries, and have achieved remarkable results. Relevant research reports show exponential growth. Perovskite solar cells use crystals with perovskite structure as electron transfer materials to improve the light absorption efficiency of the solar cells. Studies show that the structure and performance of the electron transfer layer directly affect the stability and life of the battery, which proves that the appropriate electron transfer layer is very important. New perovskite ceramic materials have been widely used in solar cell devices. This chapter mainly introduces the most common perovskite thin films and their preparation methods, organic–inorganic perovskite solar cells, etc., focusing on their development status, and the main factors affecting their stability. Finally, the current problems and development prospects in the research and application of perovskite solar cells are introduced, which will lay a solid foundation for the deeper understanding of perovskite solar cells and the preparation of new and efficient ones.

Keywords Advanced ceramics · Perovskite solar cell · Electronic transmission material

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1 Introduction

The continuous exploitation of non-renewable resources, such as petroleum and mineral resources, not only brings energy crisis but also causes serious environmental pollution. Therefore, it is an effective solution to solve the soaring world energy demand and global climate crisis to find corresponding alternative energy resources. As the cleanest and most abundant renewable energy, solar energy has attracted the attention of many researchers. Solar energy mainly converts light energy into electric energy and heat energy through photoelectric conversion and photothermal conversion. In recent years, solar cells have experienced the first generation (crystalline silicon solar cells, the most commonly used monocrystalline silicon thin film solar cells, with a maximum conversion efficiency of 24.7%) and the second generation (thin film solar cells, with a maximum conversion rate of 20.3%). Despite their high conversion efficiency, both the first- and second-generation solar cells share a common disadvantage that cannot be ignored, namely, on the one hand, they cause serious harm to the environment, and on the other hand, silicon processing and raw material costs are high. Based on this, the third-generation solar cell–new solar cell, while maintaining high conversion efficiency, has gradually become a research hotspot with the standard of low cost, rich raw materials, non-toxic, and eco-friendly, especially the new perovskite solar cell.

So far, different types of solar cells, such as polysilicon solar cells, single-crystal silicon solar cells, copper indium gallium selenide (CIGS) solar cells, cadmium telluride (CdTe)-based solar cells, quantum dot (QD) solar cells, organic photovoltaic, and perovskite solar cells have been reported. Specifically, energy conversion efficiency and material cost plays a crucial role in the commercialization of solar cells. At present, crystalline silicon solar cells occupy a dominant position in the market. However, expensive manufacturing process requirements and expensive raw materials have forced researchers to try to find an efficient and low-cost new photovoltaic technology. Perovskite materials have attracted much attention because of their excellent photoelectric properties, low material consumption, and considerable production cost [1]. The steady progress of research and development in the perovskite field has increased the possibility that it can replace existing solar cell technology, and it has good prospects [2].

Perovskite is named after the Russian mineralogist L.A. Perovski. The molecular formula of the perovskite structure material is ABX_3, which is generally a cubic or an octahedral structure, and is shown in Fig. 1 [3]. As shown in the structure, the larger A ion occupies an octahedral position shared by 12 X ions, while the smaller B ion is stable in an octahedral position shared by 6 X ions [3, 4]. The crystallographic stability and probable structure of perovskites can be deduced by considering a tolerance factor t and an octahedral factor μ, As shown in formula (1), and (2) [5], Where, t is defined as the ratio of the distance between A–X and B–X in the ideal solid sphere model, which is defined by formula (1), where RA, RB, and RX are the ionic radii of the corresponding ions, μ is defined by formula (2). For halide perovskite (X = F, Cl, Br, and I), and t is usually in the range $0.81 < t < 1.11$.

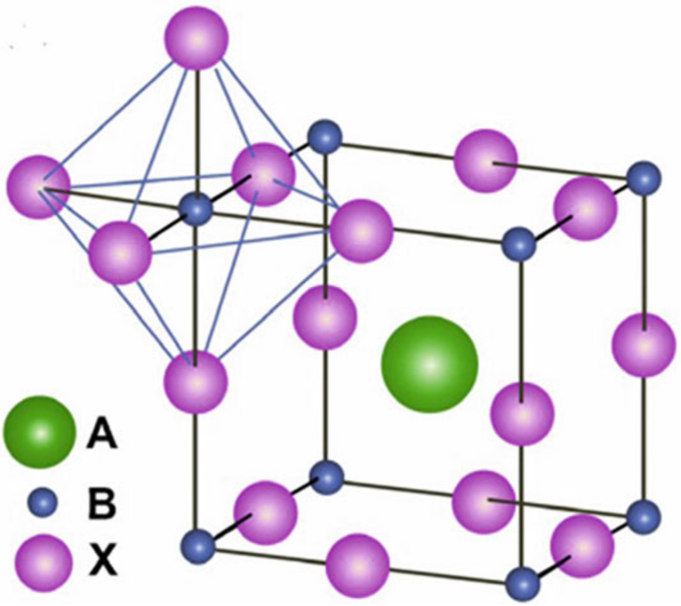

Fig. 1 Perovskite crystal structure. Reprinted from Park [3], with permission from Elsevier. Copyright (2015)

while μ is in the range $0.44 < \mu < 0.90$. For a narrow range of $0.89 < t < 1.0$, the cubic structure in Fig. 1 is likely to exist, and for a lower t value the less symmetrical tetragonal or orthogonal structures will exist [6, 7]. For organic–inorganic halide perovskite, the larger cation A is organic (methylammonium), and the anion X is generally a halogen (I, Br, and Cl), and for high-efficiency solar cells, the B ion is generally Pb [8].

$$t = (r_\mathrm{A} + r_\mathrm{X})/(\sqrt{}2[r_\mathrm{B} + r_\mathrm{X}]) \tag{1}$$

$$\mu = R_\mathrm{B}/R_\mathrm{X} \tag{2}$$

2 Research Status of Perovskite Ceramic Materials

According to the survey, the PCE (power conversion efficiency) of perovskite solar cells increased rapidly from 3.8% to 22.1% in just 7 years from 2009 to 2016 [9]. As a new all solid-state planar solar cell, perovskite solar cell has developed rapidly because of its advantages of simple preparation process, low cost, and high efficiency.

In 2009, Kojima et al. [10] first studied the photovoltaic function of organic–inorganic lead halide perovskite compounds $CH_3NH_3PbBr_3$ and $CH_3NH_3PbI_3$ as visible light sensitizers in photoelectric chemical cells. The nanocentricity self-assembled in the mesoporous titanium dioxide (TiO_2) film exhibits a strong band gap absorption characteristic. Based on $CH_3NH_3PBI_3$, the spectral sensitivity is as high as 800 nm, and the solar conversion efficiency is 3.8%. The battery based on $CH_3NH_3PBB_3$ has a high optical voltage of 0.96 V, and the external sub-conversion efficiency is 65%. In 2011, Im et al. [11] formed (CH_3NH_3) PBI_3 quantum dots on the surface of nano-TiO_2. Kim et al. [12] permeated the spiro-MeOTAD hole transport material into the perovskite-sensitized nanorods' film, and improved the power conversion efficiency (PCE) to 9.4%. Christians et al. [13] used to successfully realize a predecessor's power conversion efficiency of 6.0%, and they had a good photocurrent stability solar cell. Hou et al. [14] reported that the interface between SnO_2 and perovskite was modified by dopamine, the hydrophilicity of SnO_2 increased, the high-quality transformation of the perovskite film was promoted, the prepared absorbing interface has achieved 16.65% high-optoelectronic conversion efficiency, the stability of the interface and has improved. Tavakoli et al. [15] reported modifications to suppress defects in perovskite light harvesting materials. These modifications include engineering of the perovskite interface with the electron and hole selective contact materials. They introduced a planar SnO_2/TiO_2 double layer to ensure fast electron extraction. To passivate surface trap states, they treated the surface of the perovskite facing the hole conductor with iodine dissolved in isopropanol. The modifications resulted in retardation of radiationless carrier recombination, increasing the photoelectric conversion efficiency of the interface to 21.65%, increasing the open circuit voltage to 1.24 V, and decreasing the opening voltage loss to only 0.37 V. At the same time, the device efficiency at 20, 50, and 65 °C remained 96%, 90%, and 85%, respectively, of the initial photoelectric conversion efficiency under continuous light, which is one of the most stable perovskite solar devices reported so far. Qi et al. [16] treated the mixed perovskite with phenylethyl ammonium iodide (PEAI). The existence of PEAI passivated and reduced the surface defects of the film. The prepared perovskite solar cell achieved a high photoelectric conversion efficiency of 23.6%, and the verified steady-state photoelectric conversion efficiency reached 23.32%. Tong et al. [17] reported in 2019 that the addition of thiocyanate (GuaSCN) can significantly improve the structure and photoelectric properties of Sn–Pb mixed low band gap perovskite films. The defect density is reduced by a factor of 10, resulting in a carrier lifetime of more than 1 microsecond and a diffusion length of 2.5 microns. A series of four-terminal solar cells with a high efficiency of 25%, and two-terminal full perovskite polycrystalline thin films of a high efficiency of 23.1% are realized. In 2020, the laminated photovoltaic device achieved a bigger breakthrough. Xiao et al. [18] added the ion-reducing sulfonic acid (FSA) drug, and zwitterionic antioxidant to inhibit Sn^{2+} oxidation, and passivating the mixed lead–tin perovskite film, making the efficiency of monochrome solar cells reach 21.7% (certified 20.7%). The entire perovskite series has a certification efficiency of 24.2% in an area of 1 cm^2. It is currently the highest certified photoelectric conversion efficiency of the global calcium titanium mineral cells.

Because of product instability and human health, researchers are working hard to achieve the Pb-free method. The most direct method to achieve a perovskite material is to use elements with similar properties (such as Sn, Ge) instead of Pb element. Jokar et al. [19] studied organic cation-doped diiodide ($EDAI_2$) and iodide (BAI) as defect passivating surface additives to improve device performance and durability of $FASnI_3$ perovskite solar cells. The addition of BAI significantly changed the orientation of crystal growth, and improved the connectivity of grains. The addition of $EDAI_2$ can cure the pinhole problem, passivate the surface defect states, prevent Sn^{2+}/Sn^{4+} oxidation and induce the slow relaxation of crystal structure. Nishimura et al. [20] prepared tin halide PSCs with an efficiency of more than 13% by adjusting the A-site cation, making its tolerance factor close to 1. The formamidinium cation was partially substituted with an ethylammonium cation, and a more stable tin calcium ore crystal was obtained, with inhibition of order 1 of trap density. Another way to reduce toxicity is to partially replace Pb in perovskite materials. A certain amount of Pb-doped tin perovskite can improve the stability and performance, so that Sn–Pb mixed perovskite has adjustable band gap, which makes it an ideal light absorbing layer for all perovskite laminated batteries [21]. However, the high defect density caused by Sn^{2+} oxidation limits carrier life and charge collection in tin-based titanium mine. Lin et al. [22] recently reported a strategy to reduce Sn vacancies in mixed Pb–Sn narrow-band gap perovskite in which Sn^{4+} (oxidation product of Sn^{2+}) is reduced to Sn^{2+} by proportional reaction with metal tin, A monolithic all perovskite series cell was fabricated, which obtained 24.8% PCCE certification in small area devices (0.049 cm^2) and 22.1% PCCE certification in large area devices (1.05 cm^2). The cell series can still maintain 90% performance after 463 h of operation at the maximum power point under full single solar illumination. From the perspective of the environment, the use of Pb in perovskite solar cells is still a threat. Partial substitution of lead has shown great advantages. In current studies, we must focus on the research of Pb device packages and appropriate recycling mechanisms, the comprehensive transformation of the perovskite solar cell will enable the photovoltaic field to reach a new height.

3 Perovskite Films and Their Preparation Methods

Perovskite is a natural calcium titanate mineral, and its chemical molecular formula is $CaTiO_3$. Typically, a material having the same crystal structure as $CaTiO_3$ is referred to as a perovskite material. In the past few decades, inorganic perovskite oxides and halides have been widely studied because of their wide applications in the fields of optics, magnetism, electronics, and superconductors [23]. Among them, the research of perovskite films in solar cells has attracted great attention. In the early devices, due to the poor quality of perovskite films, there are many defects, which not only reduce the light absorption performance and photocurrent, but also cause the contact between electron and hole transport layer, resulting in the decline of the overall performance. The prepared films used in perovskite batteries also have some problems, such as low viscosity of precursor solution, low crystallinity caused by fast crystallization, and poor contact between substrate and perovskite layer. Therefore, it is crucial for the preparation of high-efficiency solar cells to design a simpler, and more effective preparation method of perovskite films, adjust the coverage and uniformity of the films, and reduce the formation of defect sites [24].

With the advancement of technology, optimizing the preparation technology of perovskite thin films is a necessary condition for the fabrication of high-efficiency devices. Typically, the chemical measurement, crystal phase, and grain structure of fine control materials are needed to prepare an efficient perovskite solar cell. Since these properties can be controlled by the preparation method, a large number of studies have been carried out on the development of thin film deposition techniques. In the evolution process of perovskite solar cells, four main deposition methods are studied, as shown in Fig. 2, which are one-step dissolution process, two-step dissolution process, steam-assisted dissolution process, and thermal evaporation process [25–28]. In addition, in order to improve the quality of perovskite films and improve the efficiency of perovskite batteries, researchers have put forward more advanced film preparation technologies after continuous exploration.

3.1 One-Step Deposition Method

One-step deposition is the earliest technique for preparing perovskite solar cells. Because of its simplicity and low cost, it is the most widely used technique. Chien Huang Chiang et al. [30] developed a one-step hot solution spin coating method to prepare high-quality mixed halide perovskite films. High-quality thin films can be prepared by a simple one-step method with an efficiency of over 16% [31]. However, due to the low surface coverage of perovskite film prepared by one-step solution deposition method, and micropores will occur in the film during the preparation process, which will affect the quality of the film, this method has been gradually replaced by subsequent preparation technologies.

3.2 Two-Step Solution Deposition Method

The perovskite film prepared by one-step deposition method has poor surface coverage and inevitable non-uniformity. In order to overcome this problem, Burschka et al. [26] developed a two-step solution deposition method based on Mitzi et al.'s [10] pioneering work to prepare uniform $MAPBI_3$ films on the MP-TiO_2 layer. The gas-assisted solution method was developed into an improved two-step solution deposition method, and methylammonium iodide (MAI) was introduced on the basis of gas-phase deposition. Chen et al. [27] prepared homogeneous conformers methylammonium lead iodide $CH_3NH_3PbI_3$ ($MAPBI_3$) films with high phase purity by the two-step deposition technique. Heping Shen and his team used the improved

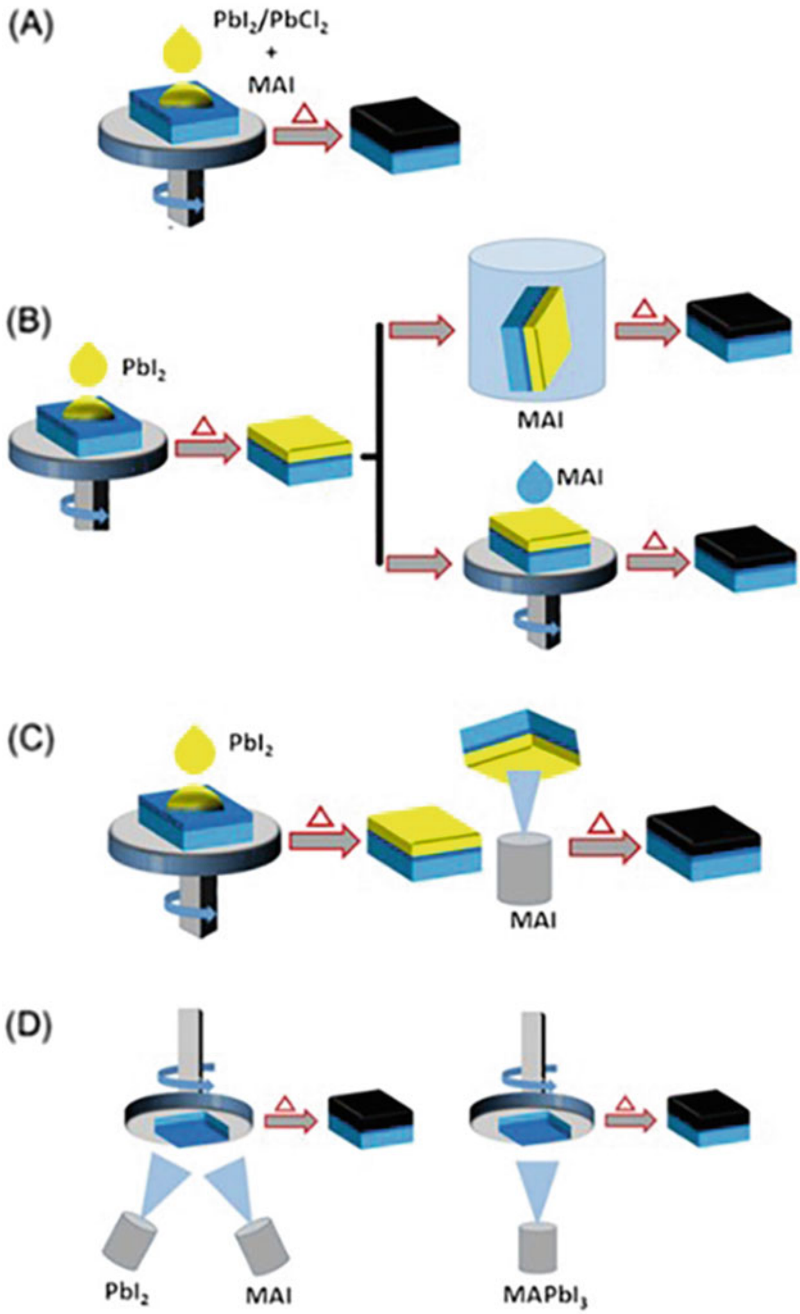

Fig. 2 The four main deposition methods to prepare perovskite (calcium titanium mine) film. **a** One-step dissolution process. **b** Two-step dissolution process. **c** Steam-assisted dissolution process. **d** Thermal evaporation process. Reprinted from Watthage et al. [29], with permission from Elsevier. Copyright (2018)

two-step deposition method to obtain a large area of high-quality perovskite film. The prepared film has less pinholes and good uniformity. The residue of PbI_2 in perovskite films prepared by this method is the minimum [32]. Julian Burschka et al. prepared perovskite thin films by two-step deposition method and achieved about 15% power conversion efficiency (measured under standard AM1.5G test conditions on solar energy). This two-step approach will provide new opportunities for the manufacture of processed photovoltaic cells. It has unprecedented energy conversion efficiency and high stability, which is equivalent to or even higher than the best thin-film photovoltaic devices today [26].

3.3 Vapor-Assisted Solution Deposition

In an improvement of the two-step solution deposition method, the successful preparation of perovskite films can be realized by vapor deposition technology. This method can better control the morphology and grain size through gas–solid crystallization, and effectively avoid the possible film delamination in the process of liquid–solid interaction. In addition, the perovskite films prepared by this preparation technology have the characteristics of uniform surface coverage, large grain size, and high conversion efficiency. Chen Qi et al. demonstrated the preparation of perovskite films and planar geometry photovoltaic devices by vapor-assisted solution method (VASP). The formed perovskite films have the characteristics of full surface coverage on the substrate and have a significant grain size on the microscale [27]. However, due to the long gas–solid reaction time and the low efficiency (only 10–12%) of devices prepared by this method, the use of this method is limited.

3.4 Low Pressure-Assisted Solution Method

Low pressure-assisted solution method is a recently developed method for the preparation of perovskite films. A high-quality double perovskite film $Cs_2AgBiBr_6$ can be prepared using this technology, which has excellent thermal stability and environmental stability. The optimum PCE of the $Cs_2AgBiBr_6$ film is 1.44% under AM1.5 (100 mW cm^{-2}) light, which is the highest efficiency of Pb-free high-stable perovskite solar cell at that time.

The preparation process of the perovskite film determines the use of the perovskite film. Different requirements for battery equipment require different preparation processes. With the advancement of commercialization and technology, a large number of more efficient preparation processes will emerge.

4 Factors Affecting the Stability of Perovskite Solar Cell

The efficiency of perovskite solar cells is increasing because of their excellent photoelectric performance, but the stability is the key factor for the commercialization of perovskite solar cells. The current research results show that the main factors affecting their stability include electron transport materials in solar devices; hole transport layer materials in devices; environment, such as humidity, temperature, etc.

4.1 Electronic Transmission Layer Material

Electronic transport layer (ETL) materials refer to materials that accept and transmit negatively charged electronic carriers, a semiconductor material (i.e., N-type semiconductor) has a higher electron affinity and is commonly used as ETL.

The ideal ETL should have the following factors: (1) The frontier orbits of molecules in ETL should match the conduction and valence bands of perovskite layers; (2) High Electron Mobility; (3) The solubility of ETL in the orthogonal solvent is much higher than the perovskite layer to prevent damage of the Perovskite layer after spinning; (4) Full film coverage; and (5) Ability to passivate defects in perovskite layers.

Electrons in the perovskite layer are transferred to ETL, which is usually collected by metal electrodes [33]. An important feature of ETL is that it can be aligned with the perovskite. For the perovskite layer to be able to absorb light effectively, its UV–visible transmittance must be high [34].

4.1.1 Metal Oxide Electron Transport Layer Material

The electron transport layer of perovskite solar cells studied at present mostly refers to the commonly used TiO_2 in dye-sensitized solar cells [26, 28]. In order to increase the growth reaction speed of perovskite materials, TiO_2 thin films with high specific surface area are usually prepared using nano-sized TiO_2 particles. Dense titanium dioxide can transfer electrons, which is beneficial to both enhancing the light absorption inside the solar cell and improving the conductivity of electrons. However, the porous TiO_2 is easily affected by ultraviolet light, which directly causes a decrease in solar cell efficiency [35]. To avoid the inherent defect of porous titanium dioxide, porous titanium dioxide can be replaced or modified with other materials [36].

Snaith et al. [37] Replaced porous TiO_2 with porous Al_2O_3, thus avoiding its inherent instability. There is still a stable photocurrent when the solar cell is irradiated with the whole solar spectrum for more than 1000 h. The prepared solar cell has good stability and can maintain the original efficiency of nearly 90% after being exposed to air for 500 h without packaging [38].

SnO_2 is another well-known ETL material because of its high transparency, electron mobility, and wide band gap [39]. As a perovskite ETL material, SnO_2 can be stable in ambient air for 30 days after low-temperature treatment [40]. Low-temperature atomic layer based on SnO_2 is relatively better than TiO_2 [41]. WO_3 has good stability and can resist acid corrosion, which has higher mobility than TiO_2 [42].

4.1.2 Organic Electron Transport Layer Materials

Organic electron transport layer materials can be divided into two categories: The first one is n-type organic small molecules, including naphthalene diimide (NDI) and fullerene derivatives: [6]-phenyl-C61-butyric acid methyl ester (PCBM), etc., and the second category is n-type conjugated polymers. Due to the advantages of easy synthesis and purification, adjustable frontier molecular orbital, high electron mobility, low cost, and good chemical/thermal stability, these materials have become a promising ETL material in inverted perovskite solar cells (p–i–n PSC) [43]. Due to the reliability of PCBM as ETL material, Snaith and colleagues demonstrated the experiment of PCBM as ETL material to explore the charge diffusion length in $MAPbI_3$ for the first time [37]. In addition, Lam and colleagues used PCBM as ETL to understand the origin of high performance of $MAPbI_3$ solar cells, and found that PCBM plays a decisive role in inhibiting the migration of halide ions from perovskite layer through halide-π non-covalent interaction [44].

4.1.3 Composite Electron Transport Layer Material

The material using more than one different material as an electron transport layer is classified as a composite material.

Taking TiO_2(compact)/Al_2O_3(mesoporous) electron transport layer as an example, trihalide permeates into the Al_2O_3 nanoparticles skeleton with a thickness of about 400 nm, which can improve the uniformity of perovskite film and inhibit the leakage caused by the appearance of pinholes [45]. Electrons are transported to the dense TiO_2 electron transport layer in the calcium titanium ore body along the surface of the Al_2O_3 nanoframework [25], Using Al_2O_3 instead of TiO_2 as nanoframework can reduce the sintering temperature to 150 °C and obtain devices with an efficiency of 10.9%.

Abrusci et al. [42] introduced benzoic acid substituted C_{60} self-assembled monolayer as a modified layer on the surface of TiO_2 framework, which significantly improved the electron collection efficiency, and increased the photoelectric conversion efficiency of the solar cell to 11.7%. The coating layer can act as a barrier to prevent or reduce exciton recombination, thus reducing the degradation of perovskite material on porous TiO_2 surface and making the device more stable. Ito et al. [46] prepared a layer of Sb_2S_3 on TiO_2 layer by chemical deposition method, which improved the efficiency and photostability of the solar cell in the form of interfacial barrier layer. Under the same conditions, the device covered with Sb_2S_3 barrier retained 65% of the original efficiency after 12 h [47].

Wang et al. [48] used graphene/TiO_2 nanoparticle composites as the electron transport layer. Because graphene has the characteristics of high conductivity and its work function (WF) lies between those of Fluorine-doped Tin Oxide) (FTO) and Titanium dioxide (TiO_2), the electron transport performance is greatly improved, and the photoelectric conversion efficiency of the solar cell is as high as 15.6%. In addition, Zhou et al. [49] doped yttrium in TiO_2 to obtain 19.3% planar heterojunction perovskite solar cells. In addition to the above-mentioned improvement of the performance of perovskite solar cells by doping to obtain composite electron transport layer materials, Table 1 lists some examples of metal ion doped ETL materials.

Table 1 Metal ion-doped ETL materials for perovskite solar cells. Reprinted from Rao et al. [50], with permission from Elsevier. Copyright (2021).

Metal doped ETL	Deposition technique	J_{K} (mA cm)	V_{c} (V)	PCE (%)
Mg–TiO, natiorod	Microwave-assisted hydrothermal method	10.4	0.S02	4.17
Nb–TiOj	Spray pyrolysis	18.08	0.880	10.26
Nb–TiOs nanoiod	Hydrothermal	16.50	0.87	7.5
Al–Ti0$_2$	Spin coating	20.0	1.07	13.8
Mg–Ti0$_2$	Spin coating	18.34	1.08	12.23
Zr–TlO	Spin coating	14.4	0.93	9.1
Mg–TiO$_{\mathrm{a}}$	Dip coaling	21.48	1.09	17.65
Cs–TlO	Spin coating	14.4	0.64	5.26
Nd–TiOa	Spin coating	22.3	1.133	18.2
Li–Ti0$_2$	Spin coating	23	1.114	19.3
Ta–TlO	Spray pyrolysis	19.01	1.01	13.76
Nb–TiO$_a$		19.10	1.00	13.81
Sm–TiO$_2$	Spin coating	18.07	1.04	13.1
Nb–TiO$_2$	Spin coating	22.25	1.08	16.96
Z$_a$–TiO$_j$	Spin coating	22.3	0.95	14
Nb–TiO$_z$	Chemical bath deposition	22.86	1.10	19.23
P.ii–Ti0$_2$	Spray pyrolysis	21.91	0 994	15.7
Sn–TlO	Co-electrodeposition	20.84	1.072	16.83
Ce–TiO$_2$	Hot-bach deposition	21.95	1.07	15
Co–TlO	Spin coating	24.078	1.027	15.75

4.2 Hole Transport Layer Materials

Hole transport layer (HTL) is another important component of PSC. As a hole transport layer, HTL must meet the following conditions: (1) Highest occupied molecular orbital (HOMO) energy level should be higher than the maximum value of valence band of perovskite material, so as to transport holes from perovskite layer to metal electrode; (2) It has high conductivity, which can reduce the series resistance and improve the Fill factor (FF); (3) The HTL layer and the calcium–titanium layer need to be in close contact [51]. The selection of HTL has a significant impact on the stability of PSCs. It is reported that the presence of HTL promotes better coverage of perovskite compared with transparent conducting oxide (TCO)/perovskite/metal [52, 53]. The use of hole transport layer (HTL) material has preliminarily solved the problems of instability and difficult packaging of liquid electrolyte perovskite solar cell.

4.2.1 Organic Hole Transport Layer Materials

Generally speaking, organic materials not only have good film-forming properties, but also have excellent hydrophobic properties, which can effectively block the erosion of water vapor to perovskite materials. Therefore, in order to solve the stability problem of perovskite solar cells, Kim et al. introduced a solid-state hole transport layer material (spiro-OMeTAD [2,2′,7,7′-tetrakis(*N*,*N*-di-*p*-methoxyphenyl-amine)9,9′-spirobifluorene]) into perovskite solar cells and prepa.red the first all-solid-state perovskite solar cells in 2012, with an efficiency of 9.7%. The results show that even without packaging, the efficiency attenuation of the solar cell is still very small after 500 h of operation [54]. In addition, the development of hole transport system with non-lithium salt additives will help to improve the stability of devices [35]. As a hole transport layer, poly[2,5-bis(2-decyldodecyl)pyrrolo[3,4-*c*]pyrrole-1,4(2*H*,5*H*)-dione-(*E*)-1,2-di(2,2′-bithiophen-5-yl) ethene] (PDPPDBTE) polymer can almost maintain the initial efficiency after being exposed to air for 1000 h without packaging [55]. Organic materials such as PTAA (poly[bis(4-phenyl) (2,4,6-trimethylphenyl) amine]) and PEDOT: PSS (poly(3,4-ethylene dioxythiophene): polystyrene sulfonate) polymer can also effectively isolate water vapor infiltration and improve system stability and other properties.

Poly[bis(4-phenyl)(2,4,6-trimethylphenyl)amine (PTAA) is a widely used amorphous polymer. It has been reported that by using PTAA as HTL to manufacture PSC, the PCE of solar cell is more than 20%. Zhao et al. [56] used undoped PTAA as HTL, tried PTAA with different molecular weights, analyzed its influence on perovskite layer deposition, and obtained 18.11% PCE. Their work shows that undoped low molecular weight PTAA can easily develop efficient PSC, which will contribute to the development of commercial PSC. PEDOT:PSS is a water-soluble polymer HTL material with a HOMO energy level of −5.5 eV [50]. PEDOT: PSS as a hole conductor has been widely used in inverted PSCs to obtain high PCE. However, poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS) will degrade PSCs due to its low electron blocking ability and poor chemical stability caused by moisture absorption [57]. Wang et al. [48] modified the surface of PEDOT: PSS with sodium citrate ($Na_3C_6H_5O_7$), which removed part of polystyrene sulfonate (PSS) on the surface of PEDOT: PSS and helped perovskite crystallize in a larger area. In order to reduce the acidity of its PSS group, PEDOT: PSS was treated with ammonia, which not only improved the performance of the corresponding PSC but also enhanced the crystal structure of perovskite films. The lower hole transport efficiency of PEDOT: PSS will lead to the imbalance of charge transport in PSC and eventually lead to hysteresis. The surface treatment of PEDOT: PSS with ethylene glycol and methanol will help to reduce the influence of PSS weak ion conductor, enhance the size of perovskite crystal domain.

4.2.2 Inorganic Hole Transport Layer Materials

Inorganic hole transport layer (HTL) materials have become popular selective contact materials because of their inherent chemical stability and low cost. Inorganic HTL materials have the characteristics of appropriate energy levels and high carrier mobility. They can not only assist charge transport but also improve the stability of PSCs under environmental conditions [57].

The application of copper (I) oxide (Cu_2O) as inorganic HTL in PSCs has been proved to improve the stability of devices stored in glove boxes [58, 59]. In 2016, it was reported that n–i–p heterojunction solar cells composed of FTO/TiO_2/$CH_3NH_3PbI_{3-x}Cl_x$/Cu_2O/Au existed stably at room temperature for 2 months, and the initial efficiency of the device was 90% [60], Fig. 3 shows the structure of an n–i–p solar cell.

Copper(I) iodide (CuI) has high hole mobility and good solubility in organic solvents. It is a good candidate material for inorganic HTL materials [62]. The device made of inorganic compound CuI as the hole transport layer is exposed to continuous light in the air for 2 h without package, and the current basically remained unchanged [63]. Moreover, when CuI is used as the hole transport layer, its efficiency is reduced and it can be recovered. After being placed in the dark for a period of time, its efficiency can basically return to the initial value. Nickel oxide (NiO_x) HTL, which is widely used in PSCs, also shows good stability. In 2015, Nejand et al. [64] found that the long-term durability of NiO_x storage in the environment was proved to be 2 months when applied to equipment as HTLs.

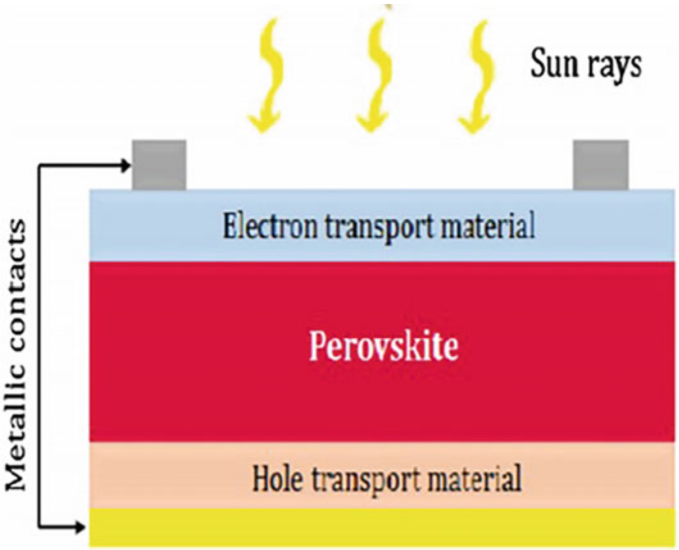

Fig. 3 The n–i–p perovskite Solar cell structure. Reprinted from Azri et al. [61], with permission from Elsevier. Copyright (2019)

4.3 Environmental Factors

In addition to the influence of electron transport layer (ETL) and hole transport layer (HTL) on the stability of PSCs, there are also environmental factors, such as humidity and temperature, which can not be ignored [65].

4.3.1 Humidity

The degradation of perovskite layer is a chemical process, because perovskite material is very sensitive to water and has polarity, so it is easy to cause hydrolysis in humid environment. Studies have shown that when $CH_3NH_3PbI_3$ is exposed to humid environment, PbI_2, $CH_3NH_2I_3$, and HI will be formed. In the process of perovskite degradation, Walsh et al. [66] combined water molecules (as Lewis base) with perovskite chemistry, received an H^+ from ammonium and degraded it through an intermediate step. The intermediate is degraded to CH_3NH_2, HI, and finally, PbI_2, in which HI and solid PbI_2 are dissolved in water. The decomposition of PbI_2 leads to further toxicity. CH_3NH_2 is a polar organic compound, so it is highly volatile and soluble in water [26, 66, 67].

The greater the air humidity, the worse the stability of perovskite solar cells. As shown in Fig. 4, an experimental device designed by researchers is used to control and measure the relative humidity of the film. The facility is considered essential for effective degradation studies.

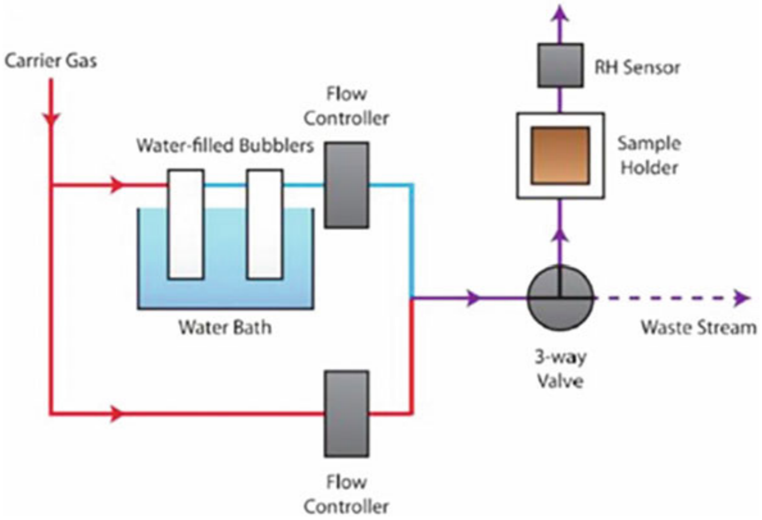

Fig. 4 Schematic representation of the system used to control relative humidity. Reprinted from Wang et al. [68], with permission from Elsevier. Copyright (2016)

4.3.2 Temperature

According to international standards, PSCs need to be stable at 85 °C for a long time in order to compete with silicon solar cells. Temperature is one of the important parameters that determine the overall efficiency of PSCs. In general, solar cells must be able to work in hot climate [50].

Leong et al. [69] studied the performance of m-TiO_2 based PSCs in the temperature range of 80–360 K to study the relationship between temperature and device performance. When $T < 330$ K, the efficiency shows an increasing trend, while when $T > 330$ K, the efficiency decreases significantly.

Under light and high temperature, the degradation of PSCs strongly depends on the composition of halide perovskite. For example, $MAPbI_3$ decomposed significantly when exposed to sample temperatures of 45–55 °C, while $MAPbBr_3$ did not. The stability of PSCs can be further improved by introducing transparent contact agents and strong perovskite components. After 160 h of continuous operation under nitrogen environment and light conditions at 85 °C, PSCs showed an initial efficiency of more than 90% [70].

Perovskite solar cells have developed rapidly in the past decade, and their efficiency has increased from 3.8% to 23.3%. In the practical application process of perovskite solar cells, in addition to improving the photoelectric conversion efficiency of the cells, the stability of the cells can be continuously improved by finding suitable materials and optimizing the preparation method of perovskite film. In addition, the preparation of large-area perovskite thin films is still an important factor restricting the large-scale and commercialization of perovskite solar cells. Therefore, the research and preparation of reliable, stable, and efficient perovskite solar cells are still a systematic, multidisciplinary, and complex project, which requires constant investment of funds and unswervant determination of researchers.

4.3.3 Other Factors

For perovskite solar cell devices, it is very necessary to understand device attenuation mechanism and optimize its stability under the premise of high efficiency. The influencing factors of perovskite stability are complex, In addition to temperature and humidity, light irradiation [71] (ultraviolet light), oxygen and the presence of electric field can cause degradation of perovskite solar cells, thus reducing their stability. In addition, the structure and performance of perovskite solar cells also affect their stability. Therefore, the selection of appropriate devices and reasonable packaging of perovskite solar cells are very critical to improve their stability [71, 72].

5 Industrialization and Challenges of Perovskite Solar Cells

In recent years, the energy conversion efficiency of perovskite solar cells has become comparable to that of conventional commercial solar cells. Due to their light weight and low production cost, perovskite solar cells have gradually replaced traditional solar cells, and many improvements are expected to break through higher efficiency. However, despite the promotion of further development of photovoltaic technology, the realization of large-scale production and commercialization of the perovskite solar cell, there are still some problems to be solved.

On the one hand, solar cells must have the ability to operate in harsh environments such as high humidity, high temperature, strong light, and changeable weather to adapt to different conditions. In order to successfully realize the commercialization of perovskite solar cell technology, scientists are committed to solving several fundamental problems: (1) Controllable growth and deposition of thin film materials; (2) High stability and long life; (3) Scale and repeatability; (4) Low toxicity; and (5) Low cost.

With low surface roughness and low density of structural defects, large-scale perovskite films are critical for industrial applications. A large number of researchers are committed to developing various film deposition technologies, including spin coating, dip coating, and spray deposition. The most widely used method for low-cost deposition of high-quality perovskite films is spin coating. However, this method could not achieve large-scale production because it is difficult to obtain large-area uniform films. The chemical vapor deposition can not only achieve a large area of film deposition but also eliminate the need for toxic organic solvents, which is a very developing film preparation process [73]. Wang et al. [74] introduced butylammonium cation into the mixed cation Pb mixed halide $FA_{0.83}CS_{0.17}Pb\ (Br_{1-y})_3$ to obtain a three-dimensional perovskite structure, which resulted in a considerable power conversion efficiency in the produced perovskite solar cell. It can maintain high stability in an analog illumination environment. Grancini [75] designed a 2D/3D perovskite structure, which not only achieved a high power conversion rate of 22% but also made an outstanding contribution to the improvement of the stability and conversion efficiency of perovskite solar cells.

The environment such as temperature, humidity, oxygen, etc., will cause the rapid aging of solar cells. Attempts have been made to adapt perovskite processing and solar cells to ambient humidity for mass production. Doping perovskite with halide ions, increasing hydrophobicity and interface modification are the main methods to make perovskite solar cells compatible with environmental humidity. In addition, proper packaging of the cell can effectively reduce the aging speed of the solar cell and improve its service life. Packaging technology can not only avoid the harm caused by Pb toxicity but also effectively improve the stability of the solar cell. Hashmi et al. [76] sealed the solar cell with epoxy glue and confirmed that the encapsulated solar cell still maintained good stability after the 1002 h aging test. This study shows that the commercial market prospect of perovskite solar cells is very considerable.

All customers, manufacturers, and investors are very concerned about the toxicity and safety of Pb-containing photovoltaic equipment. The toxicity of Pb has brought great limitations to the development of perovskite solar cells. In recent years, researchers have developed lead-free alternatives after extensive research. slavney et al. [77] combined non-toxic Bi^{3+} into the perovskite lattice of $CsAgBiBr_6$ using a double perovskite structure, which improved the thermal and wet stability of the material. However, we must admit that Pb-free materials are not perfect in the field of perovskite solar cells because the performance of these devices is still not enough to meet the needs of industrial commercialization.

In addition, since the perovskite layer plays a decisive role in the solar cell structure, measurements often were taken to improve the efficiency and stability of the solar cell include (1) Optimization of the preparation method of perovskite films; (2) Changing the chemical composition of the solar cell; and (3) Improving the electron transport layer and hole transport layer to better separate electrons and holes and smoothly reach the two poles to form current, so as to improve the stability of the solar cell and improve its service efficiency. 2D perovskite thin films have strong light absorption in the visible region and strong photoluminescence properties at room temperature. As a perovskite solar cell material, it can maintain its performance even under long-term exposure in high humidity environment [78]. Since the solar cell can be dismantled, timely recycling of the main components of the solar cell can not only save raw materials and production time, but also reduce the pollution of solid waste to the environment [79], According to the investigation, the Pb concentration in perovskite solar cells is as high as 0.55%, and the recovery of Pb solved the great problem caused by the toxicity of Pb components [80]. As shown in Fig. 5, Lichong et al. [81] recovered Pb from lead acid batteries for the preparation of perovskite films, which reduced lead pollution to the environment and greatly reduced resource waste.

The life cycle of a solar cell consists of four parts: raw materials, module manufacturing, module operation, and processing, which together constitute the entire

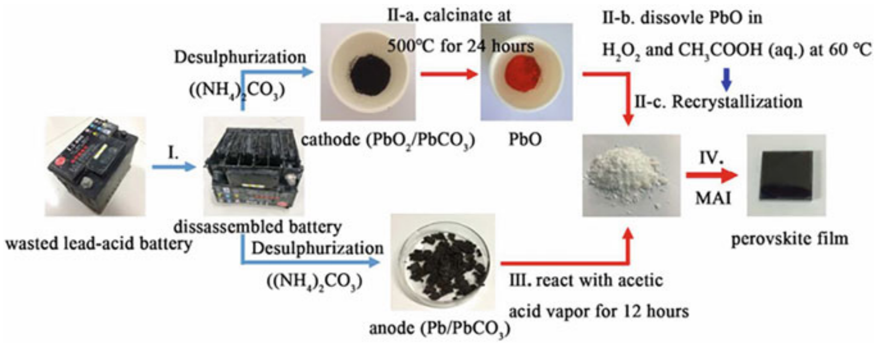

Fig. 5 Schematic diagram of lead recovery from a lead-acid battery. Reprinted from Li et al. [81], with permission from Elsevier. Copyright (2019)

life cycle of electronic equipment. When discussing the application and commercialization of equipment in reality, it is particularly important to comprehensively consider each step. Therefore, in addition to finding suitable materials as raw materials for batteries and solar cells, many factors such as battery/solar cell manufacturing cost, preparation process, and the impact of residues on the environment should be considered comprehensively.

6 Conclusion and Outlook

As a new generation of green cells, perovskite solar cells have gradually replaced crystalline silicon solar cells and occupy a certain position in the solar cell market. Since 2009, perovskite solar cells have developed rapidly because of their simple preparation process, rich raw materials, low cost, and environment-friendly. Organic–inorganic perovskite type light absorbers have made great progress in the past decade. The power conversion efficiency of single-junction solar cells using perovskite light absorbers has reached more than 25%. Some researchers even reported how to adjust the subsequent film thickness and performance to achieve an efficiency of more than 30%. During the development of perovskite solar cells, it has been found that the metal Pb in the solar cells is toxic and pollutes the environment, and the existence of Pb composition will also affect the stability of the cells. Therefore, improving the stability of solar cells and promoting the green and sustainable development of the solar cells are very important for the large-scale production and commercialization of the cells. Based on this, a large number of studies are devoted to the modification and interface modification of perovskite and the search for alternative materials.

Compared with other photovoltaic technologies, PSCs have a huge impact on global energy production. With their simple production process and low material cost, PSCs have made significant progress in conversion efficiency, achieved high efficiency in the laboratory, and will continue to mature with increased stability.

While achieving high efficiency, solving the problem of solar cell stability is very important to realize the practical application of high-efficiency PSCs. Although there are some limitations in the development of perovskite solar cells. Perovskite cells are still a kind of rapid development and great potential solar cells in the present and near future for a long time. We believe that with the continuous innovation of technology, perovskite solar cells will break through a series of problems currently facing, solve the energy crisis and environmental pollution problems, and achieve full coverage of new energy solar cells in the future.

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