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A Short Progress Report on High-Efficiency Perovskite Solar Cells

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Abstract

Faced with the increasingly serious energy and environmental crisis in the world nowadays, the development of renewable energy has attracted increasingly more attention of all countries. Solar energy as an abundant and cheap energy is one of the most promising renewable energy sources. While high-performance solar cells have been well developed in the last couple of decades, the high module cost largely hinders wide deployment of photovoltaic devices. In the last 10 years, this urgent demand for cost-effective solar cells greatly facilitates the research of solar cells. This paper reviews the recent development of cost-effective and high-efficient solar cell technologies. This report paper covers low-cost and high-efficiency perovskite solar cells. The development and the state-of-the-art results of perovskite solar cell technologies are also introduced.

Keywords: Solar cells, Perovskites, Stability, Renewable energy

Introduction

About 85% of the world's energy requirements are currently satisfied by exhaustible fossil fuels that have detrimental consequences on human health and the environment. Moreover, the global energy demand is predicted to double by 2050 [1].

Therefore, the development of renewable energy, such as wind energy, water energy, and solar energy, becomes an imminent requirement. Renewable energy-based power generation capacity is estimated to be 128 GW in 2014, of which 37% is wind power, almost one third solar power, and more than a quarter from hydropower (Fig. 1a). This amounted to more than 45% of world power generation capacity additions in 2014, consistent with the general upward trend in recent years.

Due to abundance, low cost, and environmental friendliness, solar energy attracts increasingly more attention from all over the world, which makes the rapid development of solar cell research in recent years.

In general, a commonly used classification divides the various PV technologies (in commercial as well as in R&D stage) into three generations [2]: first generation, G1: wafer-based; mainly mono c-Si and mc-Si ; second generation, G2: thin film; a-Si, CdTe, CIGS, CuGaSe; third generation, G3: multi-junction and organic photovoltaics (OPV), dye-sensitized solar cells (DSSCs), and solar cells based on quantum dots as well as other nano-materials.

The development of the three-generation solar cells produced a rich variety of solar cells, such as Si solar cells, III–V solar cells, perovskite solar cells (PSCs), thin film solar cells, dye-sensitized solar cells, and organic solar cells. However, practical, low-cost, and high-efficiency third-generation solar cells are yet to be demonstrated. Si solar cells are well developed and mature, but there is little room for further improvement [3–6]. III–V solar cells have a very high efficiency; however, its weakness is the high cost, which limits its applications [7–9]. Quantum dot solar cells have been receiving significant attention because of their low cost and high efficiency, but most efficient devices have been prepared with toxic heavy metals of Cd or Pb [10–12]. Halide perovskites have recently emerged as promising materials for low-cost, high-efficiency solar cells. As the perovskite solar cell technology becomes more and more mature, the efficiency of perovskite-based solar cells has increased rapidly, from 3.8% in 2009 to 22.1% in 2016 [13–16]. However, the stability issues still require further studies.

To give an update of the field, this paper reviews the recent development of high-efficiency PSCs. This report

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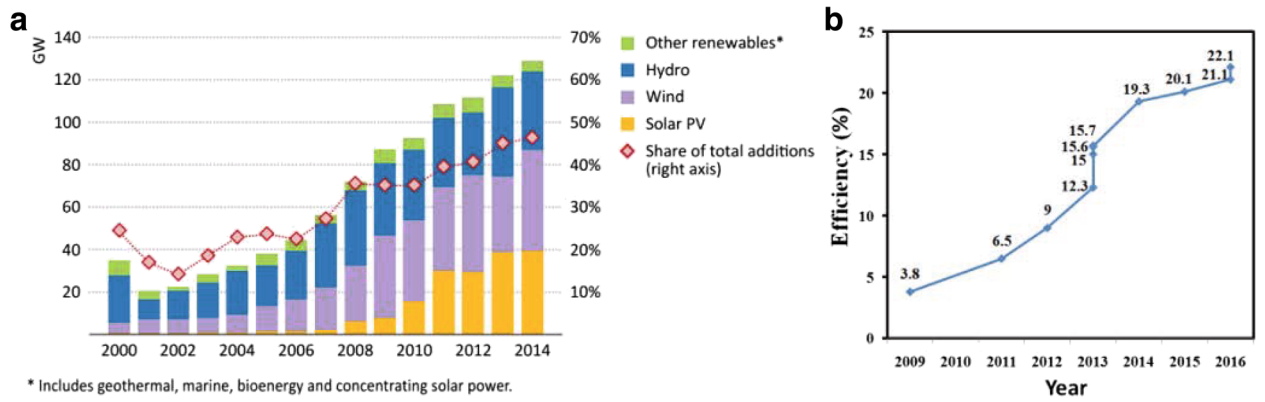

Fig. 1 a Global renewable-based power capacity additions by type and share of total capacity additions [60]. **b** Rapid PCE evolution of perovskite solar cells from 2009 to 2016

briefly introduces the history of PSCs and then focuses on the key progress made in high-efficiency perovskite solar cells. Recent efforts on the stability of perovskite solar cells will also be discussed. At the end of the report,we also give a brief introduction to the interface engineering.

Principle and History of Perovskite SCs

PSCs have recently become one of the hot spots owing to its low preparation cost and high-conversion efficiency in the fields of solar cell research. And it is regarded as a great potential material for its superiority (compared with other materials) that may assist perovskite with ultimate usurping of the reigning cell material.

In 1991, inspired by the principle of photosynthesis, O'Regan and Gratzel reported a landmark construction of solar cell called dye-sensitized solar cell, which can cover the sun light energy into electricity energy with an efficiency about 7% [17]. Presenting numerous advantages such as abundant raw materials, facile processing, and low cost compared with conventional solar cells, these novel solar cells made itself investigated popularly rapidly after its arising. And it is this work that inspired the emergence of PSCs, a DSSC with perovskite compounds.

Perovskite originally refers to a kind of ceramic oxides with general molecular formula ABY_3 discovered by the German mineralogist Gustav Rose in 1839. It was named "perovskite" because it is a calcium titanate($CaTiO_3$) compounds exists in calcium titanium ore [18]. The crystal structure of a perovskite is showed in Fig. 2a. In 2009, perovskite structured materials were first utilized in solar cells by Miyasaka and his colleagues. They creatively replaced the dye pigment in DSSCs with two organic-inorganic hybrid halide-based perovskites, $CH_3NH_3PbBr_3$ and $CH_3NH_3PbI_3$. And, eventually, they gained relatively not considerable power conversion efficiency (PCE) of 3.13 and 3.81%, respectively [13].

However, the work did not gain much attention due to low efficiency and poor stability, which resulted from a hole transport layer (HTL) with liquid electrolyte.

An evolutionary jump then happened in 2012 when Kim, Gratzel and Park et al. [14] used perovskite absorbers as the primary photoactive layer to fabricate solid-state meso-superstructured PSCs. Spiro-MeOTAD and mp-TiO_2 were used as the hole transport and electron transport materials (HTM/ETM), respectively, in their work and resulted in a relatively high efficiency of 9.7% for the first reported perovskite-based solid-state mesoscopic heterojunction solar cell.

After this breakthrough, the investigation of PSCs became hot gradually in photovoltaic (PV) research in the following years. Eventually, the efficiency of PSCs was promoted to 22.1% in early 2016 [1]. Since the maximum theoretical PCE of the PSCs employing $CH_3NH_3PbI_{3-x}Cl_x$ is 31.4%, there is still enough space for development [19].

Figure 2b shows the general configuration of PSCs, which usually comprises a tin-doped indium oxide (ITO)/fluorine-doped tin oxide (FTO) substrate, metal electrode, a perovskite photoactive layer, together with necessary charge transport layers (i.e., a hole transport layer (HTL) [20] and an electron transport layer (ETL) [21]) [22, 23]. Figure 2c, d shows two main device architectures: meso-superstructured perovskite solar cells (MPSCs) [24], which incorporate a mesoporous layer, and planar perovskite solar cells (PPSCs) in which all layers are planar [25].

The working principle of these PSCs can be briefly summarized in the following ways: perovskite layer absorbs the incident light, generating electron and hole, which are extracted and transported by ETMs and HTMs, respectively. These charge carriers are finally collected by electrodes forming PSCs [23].

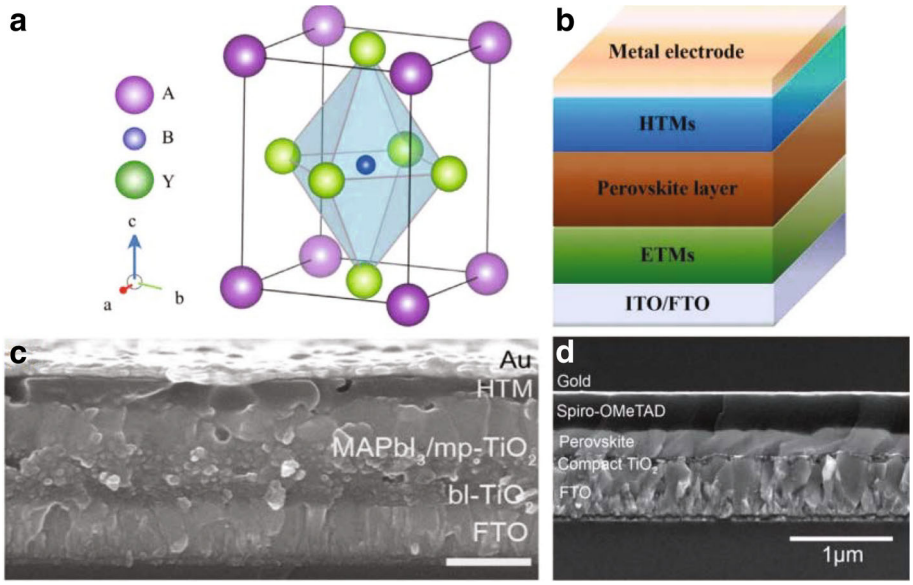

Fig. 2 a Crystal structure of a perovskite [22]. **b** Schematic diagram of general device [23]. **c** Cross-section scanning electron microscopy (SEM) images of a meso-superstructured perovskite solar cell (*scale bar* is 500 nm) [22]. **d** Cross-section SEM images of a normal planar perovskite solar cells with the presence of an HTL and an ETL [22]

High-Efficiency Perovskite Solar Cells

Intramolecular Exchange

In June 2015, Woon Seok Yang and his colleagues report an approach for depositing high-quality $FAPbI_3$ films with which they fabricated $FAPbI_3$ PSCs with a PCE of 20.1% under AM 1.5 G full-sun illuminations [26].

On the road to enhance the efficiency of solar cells, the deposition of dense and uniform films is critical for optoelectronic properties of perovskite films and is an important research topic of highly efficient PSCs. Woon Seok Yang and his colleagues report an approach for depositing high-quality $FAPbI_3$ films, involving $FAPbI_3$ crystallization by the direct intramolecular exchange of dimethyl sulfoxide (DMSO) molecules intercalated in PbI_2 with formamidinium iodide (Fig. 3) . This process produces $FAPbI_3$ films with (111)-preferred crystallographic orientation, large-grained dense microstructures, and flat surfaces without residual PbI_2. Using films prepared by this technique, they fabricated $FAPbI_3$-based PSCs with maximum power conversion efficiency greater than 20%.

Cesium-Containing Triple-Cation Perovskite Solar Cells

Adding inorganic cesium to triple-cation perovskite compositions, Michael Saliba and his colleagues demonstrated a perovskite solar cell which not only possesses higher PCEs of 21.1% but also is more stable, contains less phase impurities, and is less sensitive to processing conditions [27, 28].

They investigated triple-cation perovskites of the generic form "$Cs_x(MA_{0.17}FA_{0.83})_{(100-x)}Pb(I_{0.83}Br_{0.17})_3$," demonstrating that the use of all three cations, Cs, MA, and FA, provides additional versatility in fine-tuning high-quality perovskite films (Fig. 4). They yielded stabilized PCEs exceeding 21 and 18% after 250 h under operational conditions. Even more, the triple-cation perovskite films are thermally more stable and less affected by fluctuating surrounding variables such as temperature, solvent vapors, or heating protocols. This robustness is important for reproducibility, which is one of the key requirements for cost-efficient large-scale manufacturing of PSCs.

Graded Bandgap Perovskite Solar Cells

On November 7, 2016, scientists from University of California, Berkeley, and Lawrence Berkeley National Laboratory reported a new design that already achieved an average steady state efficiency of 18.4%, with a height of 21.7% and a peak efficiency of 26% [29–31]. They use a single-atom thick layer of hexagonal boron nitride to combine two materials into a tandem solar cell and, eventually, obtained high efficiency. The compositions of the perovskite materials are both the organic molecules methyl and ammonia, whereas one contains the metals tin and iodine, while the other contains lead and iodine doped with bromine. The former is tuned to preferentially absorb light with an energy of 1 eV—infrared or heat energy—while the latter absorbs photons of energy 2 eV, or an amber color. Prior to this attempt, the merging of two perovskite materials has failed because the materials degrade one another's electronic performance. This new way to combine two perovskite solar cell materials into one

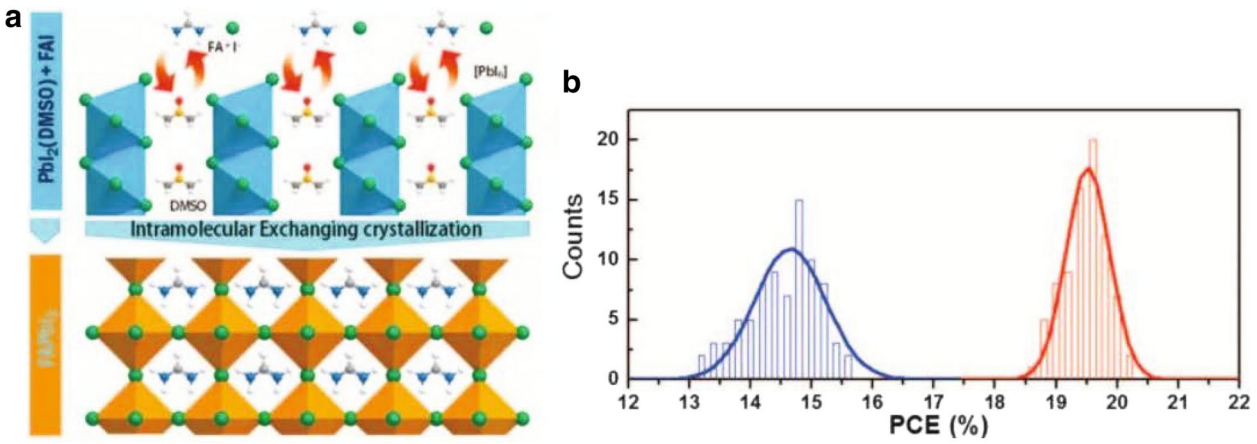

Fig. 3 PbI_2 complex formation and X-ray diffraction. **a** Schematics of $FAPbI_3$ perovskite crystallization involving the direct intramolecular exchange of DMSO molecules intercalated in PbI_2 with formamidinium iodide (FAI). The DMSO molecules are intercalated between edge-sharing $[PbI_6]$ octahedral layers. **b** Histogram of solar cell efficiencies for each 66 $FAPbI_3$-based cells fabricated by IEP and conventional process [26]

"graded bandgap" solar cell demonstrated exciting results. The solar cell absorbs nearly the entire spectrum of visible light. This is very beneficial to improve efficiency. The structure is shown in Fig. 5. They found that freshly illuminated cells tend to have higher PCE than cells that have been illuminated for more than a few minutes. For example, for a given graded bandgap perovskite cell, the PCE is between 25 and 26% in the first 2 min of illumination while the cell reaches a "steady state" with a stable PCE of 20.8% after approximately 5 min. This result indicates that perovskite-based solar cells have time-dependent performance characteristics. The measurement of 40 graded bandgap perovskite cells demonstrated that the average steady state PCE over all devices is 18.4% while the best graded bandgap cell in the steady state exhibited a PCE of 21.7%.

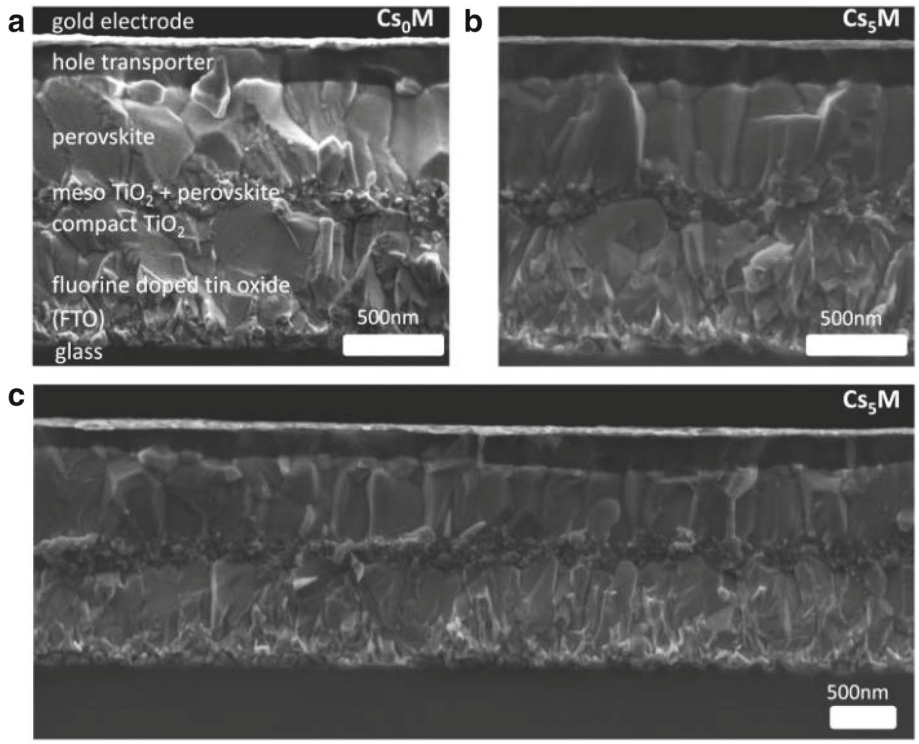

Fig. 4 Cross-sectional SEM images of **a** Cs_0M, **b** Cs_5M, and **c** low-magnification Cs5M devices [27]

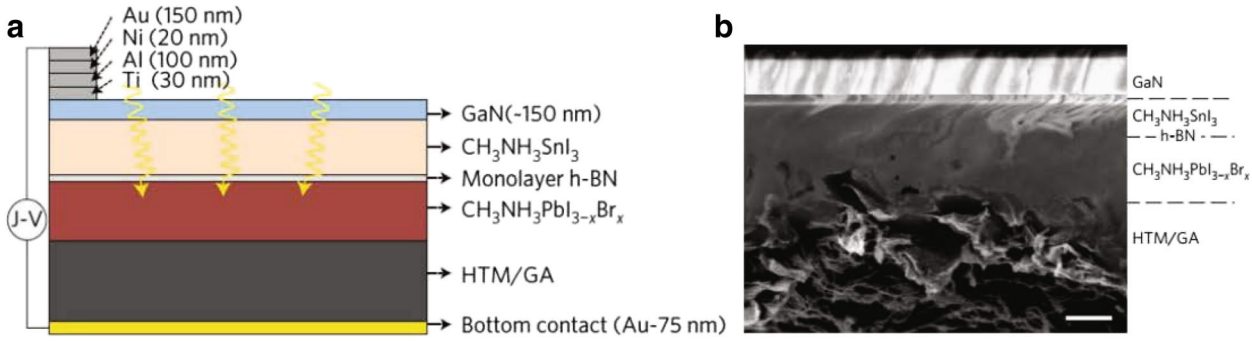

Fig. 5 Cross-sectional schematic and SEM images of perovskite cell with integral monolayer h-BN and graphene aerogel. **a** Schematic of a graded bandgap perovskite solar cell. Gallium nitride (GaN), monolayer hexagonal boron nitride (h-BN), and graphene aerogel (GA) are key components of the high-efficiency cell architecture. **b** Cross-sectional SEM image of a representative perovskite device. The division between perovskite layers and the monolayer h-BN is not visible in this SEM image. The *dashed lines* indicate the approximate location of the perovskite layers and the monolayer h-BN as a guide to the eye. The location of perovskite layers and monolayer h-BN is extracted from the related EDAX analysis. Thickness of the $CH_3NH_3SnI_3$ layer is 150 nm and that of the $CH_3NH\_3PbI_{3-x}Br_x$ is 300 nm. *Scale bar*, 200 nm [29]

Stability of Perovskite Solar Cells

In recent years, the record efficiency of PSCs has been updated from 9.7 to 22.1%. However, the poor long-term device stability of PSCs is still a big remaining challenge for PSCs, which decide whether exciting achievements could be transferred from the laboratory to industry and outdoor applications. Therefore, long-term stability is an issue that needs to be addressed urgently for PSCs. Quite a number of people have shown interest in the issue of stability and given guiding opinions on improving stability [32–44].

Multiple reports have suggested that moisture and oxygen, UV light, solution processing, and thermal stress are four key factors affecting the stability of PSCs. Observed (sometimes rapid) degradation occurs when devices are exposed to those environmental factors [22, 32, 45, 46].

Guangda Niu and his colleagues [32] expressed their views that in order to modulate the stability of PSCs, many factors should be taken into consideration, including the composition and crystal structure design of the perovskite; the preparation of the HTM layer and electrode materials; the thin film fabrication method, interfacial engineering, and encapsulation methods (multilayer encapsulation or helmet encapsulation); and the module technology. Their work verified that oxygen, together with moisture, could lead to the irreversible degradation of $CH_3NH_3PbI_3$ which is always employed as sensitizers in PSCs. They expose $TiO_2/CH_3NH_3PbI_3$ film to air with a humidity of 60% at 35 °C for 18 h, and then, the absorption between 530 and 800 nm greatly decreased (Fig. 6d).

Especially, humidity is an indispensable factor when an experimental investigation on the issue of stability is conducted.

Work lead by Kwon et al. shows that the hygroscopic nature of amine salts results from the origin of moisture instability [47]. Figure 6a shows the likely process of $CH_3NH_3PbI_3$ decomposition which was displayed by Frost et al. [48]. The process indicates that HI and MA are soluble in water, which directly leads to irreversible degradation of the perovskite layer.

Yang et al. investigated this degradation process by performing in situ absorbance and grazing incidence X-ray diffraction (GIXRD) measurements [49]. To make a valid contrast in degradation, they carefully control the relative humidity (RH) in which the films were measured. Figure 6b shows their research result of the influence of RH on the film degradation. The absorption reduced to half of its original value in only 4 h for the 98% RH case while this would take 10,000 h extrapolation of the degradation curve for a low RH of 20%. The result indicates expectedly that higher RH values cause a more rapid reduction in film absorption than a low RH. Moreover, further experiment demonstrates that varied carrier gases, N_2 or air led to no significant change in the degradation of the absorbance, indicating that the main cause of degradation in the perovskite film, under normal atmosphere, is the presence of moisture.

In 2014, De Wolf et al. used another powerful technique, photothermal deflection spectroscopy (PDS), to measure the moisture-induced decomposition of $CH_3NH_3PbI_3$ [50]. They measured the PDS spectra of $CH_3NH_3PbI_3$ layers after exposure to ambient air with 30–40% relative humidity during 1 and 20 h, respectively. Figure 6c shows that the absorptance between photon energies of 1.5 and 2.5 eV drops by two orders of magnitude after exposure to humidity for 20 h. In addition, the absorption edge that occurs at 1.57 eV in its initial state shifts to 2.3 eV, an energy corresponding to the bandgap of PbI_2 [51], which

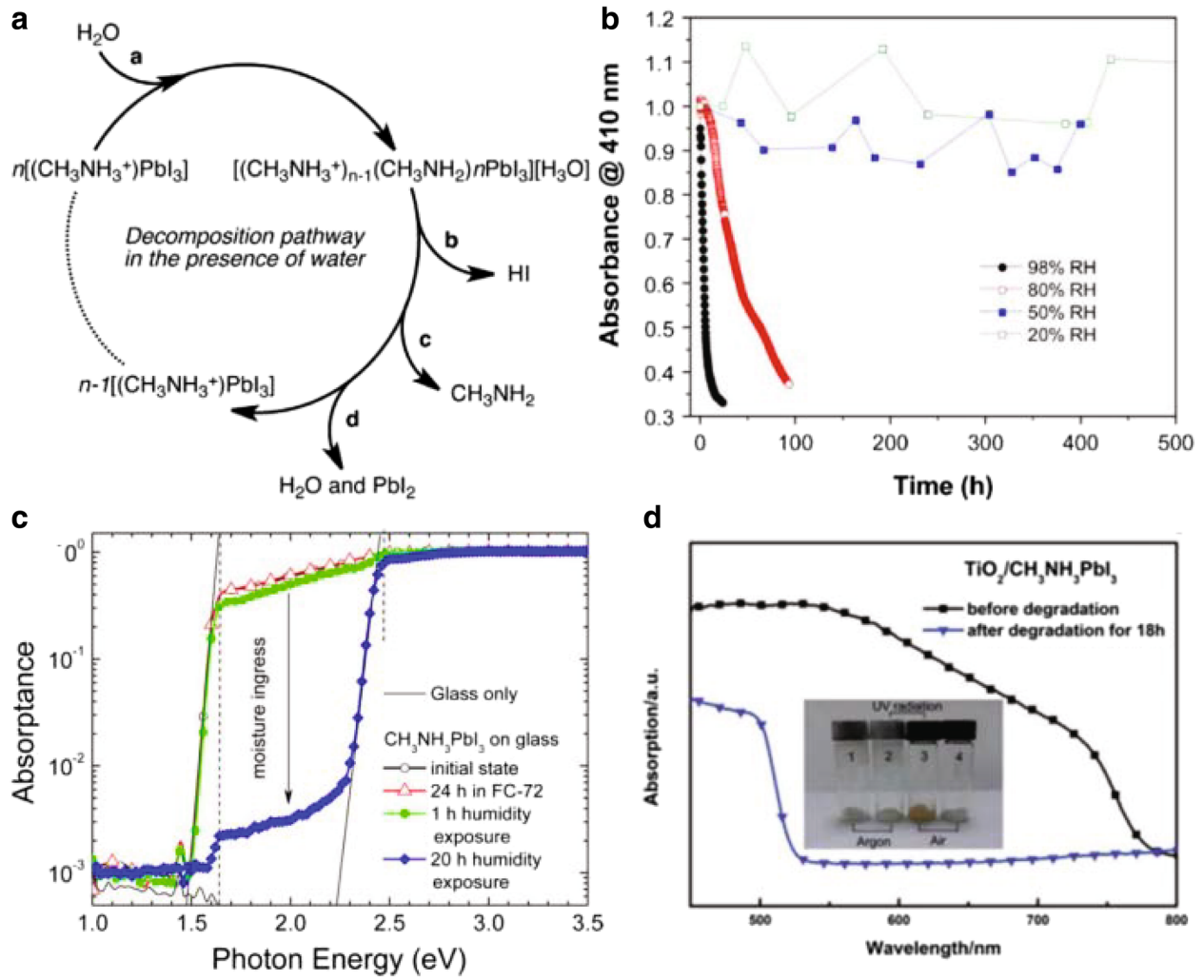

Fig. 6 a Proposed decomposition pathway of $CH_3NH_3PbI_3$ in the presence of a water molecule. The main product of this pathway is PbI_2 [48]. **b** Normalized absorbance measurements (taken at 410 nm) for $CH_3NH_3PbI_3$ films exposed to different relative humidity [49]. **c** PDS spectra for $CH_3NH_3PbI_3$ films before (initial state) and after exposure to a relative humidity in the range of 30–40% for different times. This clearly indicates a significant reduction in absorption in the range of 1.5–2.5 eV after exposure to humidity [1]. **d** Degradation of $CH_3NH_3PbI_3$ in moisture and air atmosphere. UV-vis absorption spectra of $TiO_2/CH_3NH_3PbI_3$ film before and after degradation. The *inset* is a photograph of CH3NH3I exposed to different conditions: (1) CH_3NH_3I exposed to argon and without UV radiation; (2) CH_3NH_3I exposed to argon and with UV radiation; (3) CH_3NH_3I exposed to air and with UV radiation; and (4) CH_3NH_3I exposed to air and without UV radiation [32]

indicate that $CH_3NH_3PbI_3$ can decompose into PbI_2 in a humid ambient due to the dissolution of disordered CH_3NH_3I [35, 52].

Many methods are researched for stability enhancement of PSCs recent years. Xin Wang et al. successfully developed a simple solution-processed CeO_x (x = 1.87) ETL at low temperature. According to their work, CeO_x-based devices exhibit superior stability under light soaking compared to TiO_2-based PSCs [53]. Zhiping Wang et al. presented the first long-term stability study of the new "mixed-cation mixed-halide" perovskite composition $FA_{0.83}Cs_{0.17}Pb(I_{0.6}Br_{0.4})_3$(FA = $(HC(NH_2)_2)$) and discover that the cells are remarkably stable when exposed to full-spectrum simulated sun light in ambient conditions without encapsulation [54]. Han et al. adopted thick carbon as the electrode and the device's own hole transport layer; the cell was stable for > 1000 h in ambient air under full sunlight while it achieved a PCE of 12.8% [55].

Interface Engineering

The interface is vital to the performance of the devices, since it is not only critical to the exciton formation, dissociation, and recombination but also influences the degradation of devices [56]. As a result, the interface engineering for reduced recombination is extremely important to achieve high-performance and high-stability PSCs.

Tan et al. reported a contact-passivation strategy using chlorine-capped TiO_2 colloidal nanocrystal film that mitigates interfacial recombination and improves interface binding in low-temperature planar solar cells. The PSCs achieved certified efficiencies of 20.1 and 19.5% for active areas of 0.049 and 1.1 cm^2, respectively. Moreover, PSCs with efficiency greater than 20% retained 90% of their initial performance after 500 h of continuous room-temperature operation at their maximum power point under 1-sun illumination [57]. Wang and coworkers inserted an insulating tunneling layer between the perovskite and the electron transport layer. The thin insulating layer allowed the transport of photo-generated electrons from perovskite to C_{60} cathode through tunneling and blocked the photo-generated holes back into the perovskite. Devices with these insulating materials exhibited an increased PCE of 20.3% under 1-sun illumination [58]. Correa-Baena et al. provided some theoretical guidance by investigating in depth the recombination at the different interfaces in a PSC, including the charge-selective contacts and the effect of grain boundaries [59].

Conclusions

The development of PSCs in the last few years makes it a promising alternative for the next-generation, low-cost, and high-efficiency solar cell technology. Driven by the urgent need of cost-effective, high-efficient solar cells, PSCs have been intensively investigated in the recent years. Various kinds of methods are used to improve the performance. We summarize the recent development of high-efficiency PSCs. The recorded efficacy of single-junction PSCs has been increased by a few folds to over 22% in the last few years, approaching the best single crystalline silicon solar cells. Undoubtedly, halide perovskite materials have emerged as an attractive alternative to conventional silicon solar cells. However, the stability issue is still urgent to be solved. The recent progress made in the device architectures and new materials open new opportunities for highly stable PSCs.

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Authors' contributions
HT provided the ideas and structure of the whole article and drafted the "Principle and History of Perovskite SCs" and "Intramolecular Exchange" sections. SSH mainly completed the "Cesium-Containing Triple-Cation Perovskite Solar Cells" and "Graded Bandgap Perovskite Solar Cells" sections. CWP wrote "Stability of perovskite solar cells" section. All authors read and approved the final manuscript.

Competing interests
The authors declare that they have no competing interests.

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