High-Efficiency Perovskite Solar Cells: Progress and Prospects

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Abstract. The quest for sustainable and efficient energy sources has intensified the exploration of alternative materials and technologies for photovoltaics. As an emerging thin-film photovoltaic technology, chalcogenide solar cells are unanimously at the forefront of research and development due to their excellent light-absorbing capacity, low manufacturing cost, simple structure, and remarkable photovoltaic conversion efficiency. First, this paper reviews the significant progress made in the development of high-efficiency chalcogenide solar cells, including the development of material compositions, device structures, and fabrication techniques that have propelled PSCs to achieve efficiencies over 25%. The paper then highlights innovative approaches in interface engineering, tandem cell configurations, and manufacturing methods that have contributed to the high-efficiency results. While emphasizing efficiency gains, the paper outlines stability challenges, recognizing their role in the practical application of PSC technology. Finally, the paper outlines promising research avenues and technological breakthroughs that could further enhance the efficiency and application of PSCs in the global energy mix, giving them the potential for a wide range of applications in photovoltaic power generation.

1 Introduction

The transition towards renewable energy sources is imperative in the global effort to mitigate climate change and reduce the utilization of fossil fuels. Solar energy, widely available and accessible to all, emerges as a promising alternative energy source, capable of sustainably fulfilling the increasing global energy needs while being environmentally beneficial. Perovskite solar cells play an important role in a variety of photovoltaic technologies, and their remarkable potential for high efficiency and low-cost production makes them an industry disruptor [1,2]. At the center of this transformation lies the unique structure of chalcogenide, which takes the general form ABX_3. where "A" stands for cations such as methylammonium (MA^+), formamidine (FA^+), or cesium (Cs^+); "B" stands for metal ions such as lead (Pb^{2+}) or tin (Sn^{2+}); and "X" is the halogen anion (Cl^-, Br^- or I^-) [3,4]. Since the debut of PSCs in 2009, their power conversion efficiency (PCE) has increased rapidly from an initial 3.8% [5]. In recent years, research teams have managed to push the efficiency of chalcogenide solar cells to new heights through continuous R&D and innovation. For example, Tsinghua University Professor Yi's team through the development of new materials for hole transport and the use of a vacuum evaporation method to manufacture chalcogenide thin film, successfully increased the photoelectric conversion efficiency of chalcogenide solar cells to 26.41%, a figure that breaks through the world's highest publicly published record [6]. This significant increase in efficiency is credited to the perovskite material's outstanding light absorption, high charge carrier mobility, and adjustable bandgap, which allows it to absorb a wide range of sunlight spectrum. [7]. The versatility of perovskite compounds allows for diverse fabrication techniques, ranging from spin-coating to vapor deposition, making PSCs adaptable to flexible substrates and scalable for mass production [8]. Developing mixed-cation and mixed-halide perovskites has further enhanced PSCs' thermal stability and environmental resilience, addressing some of the early concerns related to their durability [9]. However, despite the substantial advancements in PSC efficiency, challenges of long-term material stability and lead toxicity remain. These challenges underscore the necessity for continued innovation in material science and device engineering to realize the full potential of PSCs in commercial applications.

This paper comprehensively reviews the current research progress of high-efficiency perovskite solar cells, elucidating the molecular and structural characteristics underpinning their performance. While the primary focus is on efficiency enhancement through material and device optimization, this paper will briefly touch upon the ongoing efforts to address stability and environmental concerns, paving the way for the next generation of PSCs.

2 Development of PSCs

The journey of PSCs began with a groundbreaking discovery by Kojima et al. in 2009, marking a new era in photovoltaic technology. They introduced organometal halide perovskites as effective light-absorbing materials in solar cells, achieving an initial efficiency of 3.8%, a modest beginning that hinted at the potential of these

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materials [5]. This milestone laid the groundwork for rapid advancements in the field.

Subsequently, a significant leap was made by Kim et al. in 2012, who developed a solid-state perovskite solar cell with markedly improved efficiency, achieving PCE over 9% and stability by incorporating a spiro-MeOTAD hole conductor. This innovation opened new avenues for further development and research of PSCs [10].

The development of the trajectory of PSCs saw various milestones, including the introduction of planar heterojunction structures by Lee et al. in 2012, which simplified the device architecture and further boosted efficiencies [11]. The ensuing years witnessed higher efficiency and stability, achieving efficiencies exceeding 26%. Figure 1 shows the development of the photoelectric conversion efficiency of PSCs in the past decade.

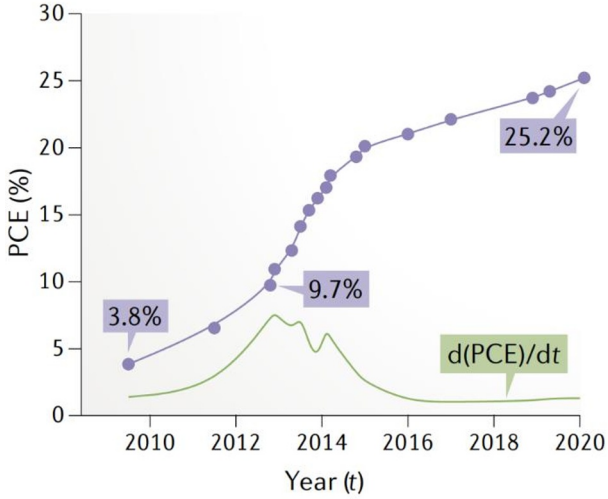

Fig. 1. Schematic diagram of general device [11].

3 Principles of PSCs

The working principle of PSCs can be divided into three main steps: light absorption, charge separation, and current collection. First, the Perovskite material has excellent light absorption capabilities, absorbing energy from visible and near-infrared light. When sunlight hits the surface of the PSCs, photons are absorbed by the material, creating electrons and holes. The absorbed sunlight energy then excites the electrons in the chalcogenide, causing them to jump into the conduction band and create holes at the same time. Due to the special crystal structure of Perovskite, the electrons and holes create a strong coupling effect within the material, driving them apart. The electrons will move along the conduction band, while the holes will move along the valence band. Finally, the separated electrons and holes are transported out by the transport layer material and collected by the electrodes, respectively. In PSCs, conductive glass or conductive polymers are usually used as electrode materials. These electrode materials have good conductivity properties and can effectively collect electrons and holes and direct them to the external circuit.

The operation of PSCs is predicated on the perovskite layer's ability to absorb sunlight, generating electron-hole pairs. The internal electric field separates these charge carriers within the device's architecture, moving toward their respective electrodes to produce an electric current.

A landmark reaction in the synthesis of perovskite materials for PSCs involves the combination of lead iodide (PbI_2) and methylammonium iodide (CH_3NH_3I) to form methylammonium lead iodide ($CH_3NH_3PbI_3$):

$$PbI3 + CH3NH3I = CH3NH3PbI3 \quad (1)$$

This simple yet effective chemical equation underscores the synthetic accessibility of perovskite materials, contributing to the low production costs associated with PSCs.

More precisely, Figure 2 shows the typical PSC assembly consisting of an active layer made of perovskite material and a base layer of either fluorine-doped tin oxide (FTO) or tin-doped indium oxide (ITO), which is a top metal electrode. The electron transport layer (ETL) and the hole transport layer (HTL), which are crucial layers for charge transfer, do this. PSCs work fundamentally by having the perovskite material absorb light, which starts the process of generating electrons and holes. Following their effective separation and channeling through the ETL and HTL, respectively, these particles are finally gathered at the electrodes that make up the PSC device [12].

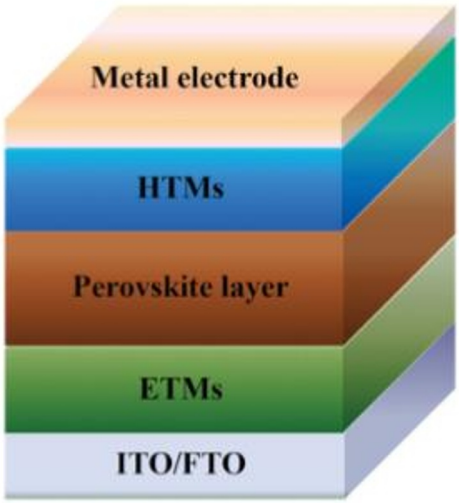

Fig. 2. Schematic diagram of general device [12].

4 High-Efficiency Perovskite Solar Cell

Woon used an intramolecular exchange procedure to create superior-quality ($FAPbI_3$ films, which he then used to build high-performance perovskite solar cells. Through a direct substitution of formamidinium iodide (FAI) for intercalated PbI_2 molecules in dimethyl sulfoxide (DMSO) molecules, $FAPbI_3$ was crystallized. By adding toluene (as a non-solvent) to a solution of 1.0 M dissolved in DMSO, this method produced $FAPbI_3$ films with a (111)-preferred crystallographic orientation, dense microstructures with large grains, and smooth surfaces without residual PbI_2(DMSO) precursor. This precursor was then annealed at 60°C under vacuum to remove one molecule of DMSO, resulting in PbI_2(DMSO). Subsequently, the PbI_2(DMSO) was dissolved in

dimethylformamide (DMF) and this film formed spin-coated onto a substrate. Finally, a solution containing FAI and a small amount of MABr dissolved in isopropyl alcohol was applied to the pre-deposited PbI_2(DMSO) layer and transformed into $FAPbI_3$ through a low-temperature spin-coating process. Solar cells fabricated using the approach demonstrated a maximum power conversion efficiency (PCE), achieving 20.1%, primarily attributed to the optimized film quality. High-efficiency perovskite solar cells were produced by this technique, which successfully regulated the crystallization of $FAPbI_3$ by direct intramolecular exchange between DMSO and FAI. This discovery is interesting because it effectively controls the formation of $FAPbI_3$ crystals by intramolecular exchange between DMSO and FAI, resulting in high-efficiency perovskite solar cells. This method provides a fresh technique to make high-quality perovskite films, which is important for perovskite solar cells' potential commercial use [7].

The use of a triple cation approach—cesium (Cs), formamidinium (FA), and methylammonium (MA)—has become a cutting-edge tactic in the quest to improve the stability and efficiency of PSCs. This new composition tackles the intrinsic structural and thermal instability of single-cation perovskites. In 2016, Saliba demonstrated that adding Cs into the FA/MA matrix significantly improves the material's stability under thermal stress and operational conditions, leading to PSCs with a stabilized power conversion efficiency (PCE) achieving 21.1% [13]. This enhancement is attributed to the optimal bandgap narrowing and the mitigation of phase impurities facilitated by the robust crystal structure afforded by the Cs inclusion. The triple cation perovskite showcases superior performance and remarkable reproducibility and longevity, with notable efficiency retention after extended periods under operational stress. This advancement underscores the potential of multi-cation engineering in pushing the boundaries of perovskite solar cell technology toward commercial viability, thereby marking a significant milestone in the quest for sustainable and efficient photovoltaic solutions.

In 2022, Wang et al. address the issue of iodide invasion in organic HTLs and present a unique method for improving the stability and efficiency of perovskite solar cells. The research team devised a lithium-free organic layer stabilization method through ionic coupling, utilizing an ion exchange process between positive polymer radicals and molecular anions. Through the use of this novel technique, an HTL was created that has hole conductivity eighty times greater than that of conventional lithium-doped layers and retains high conductivity and energy-level alignment even in the face of extreme iodide invasion. They used 1,1,2,2,3,3-hexafluoropropane-1,3-disulfonimide ($HFDF^-$) anions, which were exchanged into poly(triarylamine) (PTAA) films, to produce HFDF-HTL, which had significantly improved characteristics. The development of PSCs with a certified power conversion efficiency (PCE) of 23.9% and remarkable long-term stability—they retained 92% of their efficiency after 1000 hours of operation under normal lighting and 85°C—was made possible by this methodological accomplishment [14]. The study marks a significant departure from conventional PSCs incorporating lithium salt–doped HTLs, known for triggering significant device instability through moisture penetration and the migration of small lithium ions into the perovskite layer, which leads to irreversible degradation.

Wang et al.'s research stands as a testament to the potential of ion-exchange strategies in circumventing the limitations of lithium doping, offering a pathway toward the development of more stable and efficient PSCs. Their findings underscore the importance of innovative material engineering in overcoming the challenges facing perovskite solar cell technology commercialization.

After one year, 2023, scholar So Min Park with his colleagues focused on addressing the critical challenge of interfacial recombination in inverted PSCs by introducing a novel approach to enhance the uniformity and efficiency of the hole-selective contacts on textured substrates. The key to their method lies in the deployment of a conformal self-assembled monolayer (SAM) comprised of phosphonic acid molecules, specifically tailored to serve as the hole-selective layer. This SAM is applied directly onto the textured substrates, which are designed to improve light management within the cell, thereby augmenting its PCE. The researchers identified that the conventional formation of SAMs encountered issues with cluster formation during the adsorption process, leading to incomplete and uneven coverage. To combat this, they utilized a co-adsorbent strategy involving 3-mercaptopropionic acid (3-MPA) to disrupt the formation of these high-order clusters. By effectively disassembling these clusters, the strategy ensured a more homogeneous distribution of the phosphonic acid molecules across the textured substrate, as is shown in Figure 3. This meticulous engineering of the SAM's application significantly minimized interfacial recombination and enhanced the electronic structure at the interface. The exceptional laboratory-measured PCE of 25.3% and the certified quasi-steady-state PCE of 24.8% for the inverted PSCs were the results of their work. These efficiencies represent a noteworthy breakthrough in the area as they approach 95% of the theoretical maximum required by the Shockley–Queisser limit.

Additionally, the study shows remarkable stability under conditions of accelerated aging. For example, an encapsulated device maintained 95% of its maximum performance after 1000 hours of tracking the powerpoint at a temperature of 65°C and 50% relative humidity, simulating operational stress [15]. This achievement not only represents a significant step forward in the development of high-efficiency, stable PSCs but also underscores the potential of innovative materials engineering to overcome longstanding challenges in solar cell technology. The successful integration of light management strategies with advanced interface engineering opens new avenues for further advancements in PSCs and other optoelectronic devices requiring similar enhancements.

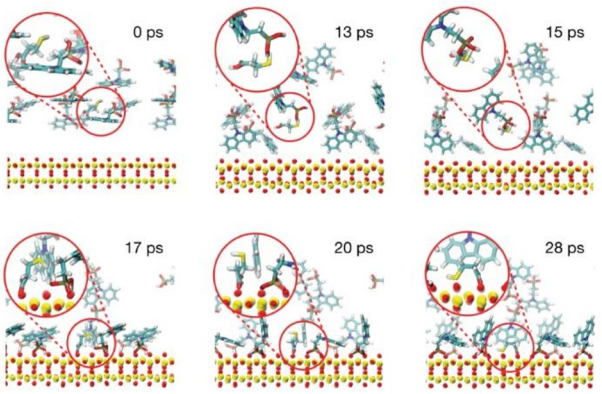

Fig. 3. The role of 3-MPA as a co-absorbent [15].

5 Conclusion

The field of PSCs represents a vibrant frontier in the quest for sustainable energy solutions, marked by rapid advancements and innovative breakthroughs. The journey from initial efficiencies of 3.8% to the current record-breaking efficiencies exceeding 26% underscores not only the inherent potential of perovskite materials but also the relentless pursuit of optimization in material compositions, device architectures, and fabrication techniques. The evolution of PSCs has been characterized by continuous research between scientific curiosity and technological ingenuity, leading to the development of mixed-cation and mixed-halide perovskites, enhanced interface engineering, and the exploration of tandem cell configurations. These efforts have collectively pushed the boundaries of efficiency, while also addressing critical challenges related to stability and environmental sustainability.

As current scholars stand on the cusp of commercializing PSC technology, it is imperative to reflect on the journey thus far and envision the path ahead. The remarkable efficiency gains achieved through meticulous material science and device engineering offer a blueprint for future innovations. However, the journey is far from over. The next chapter in the development of PSCs will undoubtedly involve tackling the twin challenges of long-term stability and lead toxicity, through continued innovation in material science and the exploration of non-toxic alternatives. Furthermore, the scalability of manufacturing processes and the integration of PSCs into the global energy mix present logistical and economic challenges that must be addressed to exert the full potential of this promising technology.

In conclusion, high-efficiency perovskite solar cells are at the forefront of the renewable energy revolution, representing the potential for a sustainable and efficient energy future. The progress made thus far is a testament to the power of scientific inquiry and technological innovation, offering a solid foundation for further advancements. The prospects for PSCs in the renewable energy sector are brighter than ever, promising to unlock new possibilities for clean, sustainable, and accessible energy solutions worldwide. As researchers continue to explore and innovate, the potential for PSCs to contribute significantly to the global energy mix is both promising and profound, heralding a new era of renewable energy technology.

References

1. M.A. Green, A. Ho-Baillie, H.J. Snaith, Nature Photonics, 8, 7 (2014)
2. N.J. Jeon, J.H. Noh, W.S. Yang, et al., Nature, 517 (2015)
3. J.Y. Kim, J.W. Lee, H.S. Jung, et al., Chemical Reviews, 120, 15 (2020)
4. S.D. Stranks, H.J. Snaith, Nature Nanotech. 10, 5 (2015)
5. J.J. Zhou, L.G. Liu, Y. Liu, et al., Joule, (2024)
6. National Renewable Energy Laboratory, 2024 Best Research-Cell Efficiencies https://www.nrel.gov/pv/cell-efficiency.html
7. W.S. Yang, J.H. Noh, N.J. Jeon, et al., Science, 348, 240 (2015)
8. Z. Li, T.R. Klein, D.H. Kim, et al., Nature Reviews Materials, 3 (2018)
9. M. Saliba, T. Matsui, K. Domanski, et al., Science, 354,309 (2016)
10. H.S. Kim, C.R. Lee, J.H. Im, K.B. Lee, et al., Sci Rep. 2 (2012)
11. N.G. Park, K. Zhu, Nat. Rev. Mater. 5 (2020)
12. H. Tang, S. He, C. Peng, NRL. 12 (2017)
13. M. Saliba, T. Matsui, J.Y.Seo, et al., Energ Environ SCI. 9, 6 (2016)
14. T. Wang, Y. Zhang, W. Kong, et al, Science, 377, 6611 (2022)
15. S.M. Park, M. Wei, N. Lempesis, et al., Nature, 624, 7991 (2023)