

The Progress and Development Tendency of Perovskite on Photovoltaic and Electronic-to-optical

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Abstract

The efficiency of perovskite materials solar cells skyrockets from 3.8% in 2009 when it is first developed in this field to 19.3% in 2014, and higher efficiency is also anticipated by researchers and merchants all over the world. Simultaneously, new kinds of LED and semiconductor laser equipments based on perovskite are also developed with the bright future into a new era and the development treads on the heel of perovskite solar cells. In this paper the state-of-the-art and highlight progress in these related fields will be reviewed and the scientific challenges and comments on the current and future issues are mentioned.

Keywords: perovskite, photovoltaic, solar cell

1. Introduction

New energy is developing rapidly all over the world inevitably and Among them photovoltaic (PV) material is playing an important role to drive the world cleaner and develop in a sustainable way. Photovoltaic cells directly convert the photons to electricity. To be a competitive factor with fossil fuels, it is necessary for PV cells to reduce the total cost. Silicon material plays the majority role in PV system, and to find PV material cheaper and higher efficiency than silicon, researchers focus their attention on new technologies such as organic photovoltaic (OPV), dye-sensitized solar cells (DSSCs), and quantum dot solar cells, but all their efficiencies are lower than silicon till today. From 2009, a new kind of solar cell called perovskite come into the view of researchers in this field. Perovskite solar cell is driven rapidly and its efficiency from 3.8% to 19.3% only cost five years, which in compared with ten years of silicon material reached same efficiency level.

Perovskite minerals have been known for nearly two-hundred years, and they have long been of interest for their superconducting and ferroelectric properties, then now perovskite series as the newest PV materials are attracting more and more researchers to endeavor on them to prominent their efficiency to compete with the currently overwhelming PV material- crystalline or polycrystalline silicon. The history of perovskite can be traced back to CsPbX_3 ($\text{X}=\text{Cl}^-$, Br^- or I^-) [1]. MØLLER investigated that these colored materials were photoconductive and suggested that they behave as semiconductors. Dieter replaced caesium with CH_3NH_3^+ to generate the first three-dimensional organic-inorganic hybrid perovskite [2]. Perovskite is generally described as the chemical formula ABX_3 ($\text{A} = \text{CH}_3\text{NH}_3^+$; $\text{B} = \text{Pb}^{2+}$; and $\text{X} = \text{Cl}^-$, Br^- , or/and I^-) with Crystal structure of cubic metal halide, such as $\text{CH}_3\text{NH}_3\text{PbI}_3$, $\text{CH}_3\text{NH}_3\text{PbBr}_3$ and $\text{CH}_3\text{NH}_3\text{PbI}_{3-x}\text{Cl}_x$. The general crystal structure of these materials is that Caesium ions or small organic cations such as methylammonium and formamidinium occupy the cub octahedral voids formed by the twelve nearest-neighbor halide ions. Halide perovskites come into account when they were found to present a semiconductor-to-metal transition with increasing dimensionality [3]. In addition to changes in electrical properties, the band gap decreased with dimensionality increased. A narrow band gap is beneficial

for solar cell applications. Initially, perovskite was used as dye replacements in dye-sensitized solar cells. In that case dyes act as light absorbers coating the surface of a film of titanium dioxide (TiO₂) nanoparticles. When light is absorbed by a dye, electrons and positive-charge carriers known as holes are generated and transferred to different transport materials: to TiO₂ for electrons, and to another material for holes. The transport materials then carry the charges to separate electrodes and so generate a voltage[4]. Their ability to convert the energy from sunlight to electricity was disclosed in 2009, firstly they were added to dye-sensitized solar cells as a sunlight-absorbing layer, and achieved an efficiency of 3.1% for X = Br⁻ and 3.8% for X = I⁻. At that time perovskite is presented in a liquid electrolyte configuration [5]. In the past several years, their efficiency of converting sunlight into electrical energy has drawn much interest from researchers. Efficiency close to 10% was first achieved in solid state perovskite cells. From then on, researchers aimed at improving efficiency of perovskite, and 19.3% is the highest perovskite efficiency reported to date[6]. This is an unprecedented and stunning rise in efficiency for the photovoltaic technology. By controlling the formation of the perovskite layer and careful choices of other materials, Zhou and his team suppressed carrier recombination in the absorber, facilitated carrier injection into the carrier transport layers, and maintained good carrier extraction at the electrodes. By these measures then they improve the efficiency to be 16.6% on average, and the highest efficiency is 19.3%. Based on these prominent behaviors, perovskite solar cell technology was selected as one of the biggest scientific breakthroughs of 2013 by the editors of Science and Nature. In this paper the advantages of perovskite materials are reviewed, and their recent progress in photovoltaic studies and electronic-to-optical are also included. Then some future perspectives with a particular focus on the improvement of efficiency, stability are discussed.

2. Advantages of Perovskite

2.1 Simple Produce Processes and Low Cost

The perovskite solar cells are cheap and easily to be made, which will benefit both science and industry. The thickness of crystalline or polycrystalline silicon solar cells manufactured today are made with approximately 150-micrometre-thick wafers. The thickness of new material merely is a few micrometers on inexpensive substrates such as glass, metal or plastic and compared with traditional silicon it is cheaper enough. The perovskite solar cells are about five times cheaper than current thin-film solar cells, and cells tend to be cheaper to make with a shorter energy payback time.

At the beginning, perovskite is liquid-based and easily solvable in suitable solvent and result in the short stability caused by dissolution of the halides in the electrolyte, then appeared to be a shortcoming that limits its application. The problem was solved by substituting a solid hole conductor for the liquid electrolyte in 2012. The compact TiO₂ layer is required to prevent direct contact between the transparent conductive oxide (TCO) and hole transporting material (HTM). The pores of dye-sensitized mesoporous TiO₂ film should be filled with HTM to induce a heterojunction. CH₃NH₃PbI₃ perovskite was deposited on TiO₂ film using either a one-step spin coating or two-step dipping method. A molecular-type HTM, such as 2,2',7,7'-tetrakis(N,N-p-dimethoxy-phenylamino)-9,9'-spirobifluorenespiro-MeOTAD), is used as a hole transporting material. This kind of structure sandwiches the thin perovskite layer between a rough mesoporous TiO₂ photoanode and a hole transporting layer [7]. These materials dissolve well in common solvents and assemble to form perovskite crystals when dried, making them cheap and simple to make. Not only did the conversion efficiency double, but the stability of perovskite

solar cell also improved greatly as a result of avoiding the use of a liquid solvent. The big surprise to the semiconductor community is to find that such simple process methods still produce very clean semiconductor properties, without the need for the complex purification procedures required for traditional semiconductors such as silicon. For example, the $\text{CH}_3\text{NH}_3\text{PbI}_3$ and $\text{CH}_3\text{NH}_3\text{PbI}_3\text{-xCl}_x$ perovskite precursor solutions were spin-coated on plasma-etched glass at room temperature in air, followed by annealing in air at 100°C for 45 min for the mixed halide and 150°C for 15 min for the triiodide which should simplify manufacturing of large-area perovskite devices that are inexpensive and perform at high levels [8]. This process requires a simple device and does not need high temperature steps, also perovskite can be prepared using large area coating or printing methods. Another important consideration for these perovskites is that they are deposited by low temperature solution methods (typically spincoating). The low energy and ease of deposition is of obvious importance for eventual manufacturing of the cells.

In a word, perovskite can be manufactured using a simple manufacturing process and in a cheap, easy-to-make way.

2.2 High Open-loop Voltage and Tandem

Perovskite solar cells with doped P3HT hole transporter showed a high voltage of open circuit up to 1.01 V [9]. Silicon and most thin-film solar cells typically produce only 0.7 V under same open circuit conditions. Since the high voltage of open circuit observed from metal halide perovskite is likely to be related to high internal photoluminescence quantum efficiency and manipulate the composition of perovskite could further improve voltage of open circuit, thus contributing to a higher efficiency [10]. This team demonstrates efficiency of cesium-doping in methylammonium lead iodide perovskites rise from 5.51% to 7.68% due to increase in short-circuit current density and open-circuit voltage via increased light absorption at optimum device thickness [11].

The direct bandgap of perovskite is 1.55 eV corresponding to an absorption onset of 800 nm, which makes this material a good light absorber over the whole visible solar emission spectrum [12]. Hybrid organometallic halide perovskite $\text{CH}_3\text{NH}_3\text{PbI}_2\text{Br}$ (or MAPbI_2Br) nanosheets with a 1.8eV band gap were prepared via a thermal decomposition process from a precursor containing PbI_2 , MABr , and MACl . The planar solar cell based on the compact layer of MAPbI_2Br nanosheets exhibited 10% efficiency [13]. Because silicon has a smaller bandgap than the perovskites, it absorbs a chunk of the solar spectrum that perovskite does not. Thus we can place a silicon cell underneath a perovskite cell to form a tandem cell [14]. Perovskites could be printed on top of silicon in a way that would add little to the cost, and because perovskites generate a higher voltage than silicon, the tandem cell would be more efficient than a silicon cell. Michael McGeheer reported that his team made 18.6% efficient tandems in which perovskites sit atop alloys of copper, indium, gallium, and selenium (CIGS). And this team pushes efficiency of tandem cells up to 35.67%. They calculate a limiting efficiency of 35.67% for an ideal (no parasitic absorption, ideal contacts) monolithic tandem, assuming a top cell voltage of open-circuit is 1100 mV [15].

2.3 High Capability to Carry Charge

An ideal solar cell material should combine good optical absorption ability with efficient charge transport properties because this character is the key to the efficient. Low-temperature solution-processed photovoltaics suffer from low efficiencies because of poor exciton or electron-hole diffusion lengths (typically about 10 nanometers) [16]. This team provided evidence of balanced long-range electron-hole diffusion lengths of at least 100 nanometers in

solution-processed $\text{CH}_3\text{NH}_3\text{PbI}_3$ [17]. And they deem that the high photoconversion efficiencies of these systems stem from the comparable optical absorption length and charge-carrier diffusion lengths. The excitons produced by light absorption have a weak binding energy of about 0.030 eV, which means that most of them dissociate very rapidly into free carriers at room temperature. The perovskite tends to have high charge-carrier mobility, then the high mobility means that the light-generated electrons and holes can move large enough distances to be extracted as current, instead of losing their energy as heat within the cell combined with high charge carrier lifetimes [18]. However, there is relatively little literature on probing the fundamental photophysics, and even the most basic question of whether photoluminescence occurs via free carrier recombination from the conduction and valence band electrons, or is preceded by exciton formation, remains unknown [8].

1. Versatile Progresses in Perovskite Field

3.1 Photovoltaic

To date, rapid progress has been made on perovskite processing and relevant material design. As a result, various processing methods have been reported.

A mesoporous scaffold made of Al_2O_3 instead of TiO_2 produced similar conversion efficiencies, even though Al_2O_3 is unable to assist in electron extraction due to its large bandgap. These researchers proposed that the electrons percolate along the surface of the Al_2O_3 nanoparticles through an ultrathin conformal coating of $\text{CH}_3\text{NH}_3\text{PbI}_3$. However, until now, the role of the mesoscopic Al_2O_3 scaffold in these devices remains obscure [19]. They observed that TiO_2 nanoparticles play a significant role in electron transport in perovskite solar cells [20]. TiO_2 hierarchical nanostructures with secondary growth have been successfully synthesized on electrospun nanofibers via surfactant-free hydrothermal route. As a result of improved light harvesting, dye loading, and reduced recombination losses, the hierarchical nanofibers yield 2.14% electrochemical conversion efficiency which is 50% higher than the efficiency obtained by plain nanofibers [21]. Al-doped ZnO (AZO) modified ZnO nanorods have been applied in $\text{CH}_3\text{NH}_3\text{PbI}_3$ perovskite solar cells and show a positive effect on open circuit voltage and power conversion efficiency. The average power conversion efficiency is improved from 8.5% to 10.07% and the maximum efficiency reaches 10.7% [22].

One of the new carbazole-based HTM devices exhibit high power conversion efficiencies amounting to 6.0% and 9.8% in dye-sensitized and perovskite solar cells, respectively [23]. The carbazole-based $\text{CH}_3\text{NH}_3\text{PbI}_3$ perovskite cell with SGT-405, having a three-arm-type structure, exhibited a remarkable photovoltaic conversion efficiency (PCE) of 14.79% [24].

Much progress has also been made in the preparation of planar perovskite solar cells, where various methods have been applied ranging from dual-source high-vacuum evaporation [25] to sequential liquid-vapor phase deposition [26] and low-temperature solution casting [27]. Among these methods, the vacuum thermal co-evaporation of $\text{CH}_3\text{NH}_3\text{I}$ and PbCl_2 (or PbI_2) and the resulting perovskite thin films exhibited the most homogeneous morphology and the highest thin film coverage, leading to a high performance of 12-15% efficiency [28]. And only limited reports have utilized vacuum sublimation technique to fabricate perovskite layers. This team provides a method of perovskite thin film deposition via a layer-by-layer sequential vacuum sublimation. This process is relatively simpler than the co-evaporation technique but gains a satisfied result. Because the devices are free of high-temperature-prepared metal oxide layers and the substrates are maintained

under 100 °C throughout the fabrication, this method points a new development orientation [29].

To prevent dendritic grain morphology giving many gaps in the process of planar perovskite films produced by traditional spin coating, a facile gas-assisted solution processing technique is reported to produce very uniform perovskite thin films consisting of densely packed single crystalline grains [30]. Polymer poly-vinylpyrrolidone (PVP) is applied as a surfactant to enhance the surface coverage of perovskite to create perovskite with smooth surfaces and uniform crystal domains [31].

What else, this team found slow transient effects causing hysteresis in the current–voltage characterization of Hybrid organo-metal halide perovskites that can lead to an over- or underestimation of the solar cell device efficiency [32]. And this reminds researchers the correction of measured efficiency.

3.2 Electronic-to-optical

Traditional LEDs should be assembled in high-temperature and high-vacuum environments, but new kind of perovskite LEDs can overcome above shortcomings. To make the perovskite LEDs, a perovskite solution is prepared and simply spin-coated onto a substrate. Thus this new LED is competitive in the cost aspect. They tune the halide compositions in the perovskite to change colors. It will be adopted widely on low-cost display, lighting and optical communication applications. The great benefit of perovskite is their ability to yield different translucent colours, such as red, yellow or brown [33].

On the aspect of use perovskite as laser diodes, the demonstration of amplified spontaneous emission from thin films of perovskites could pave the way to solution-processed low-cost lasers that can be easily tuned across the entire visible spectrum.

One of the main obstacles that prevents the realization of an electrically driven injection laser that uses conjugated organic materials as a gain medium is the difficulty of reaching the lasing threshold density of emissive excited states without compromising the gain by excited-state absorption. The creation of non-radiative states and higher-order excited-state annihilation processes, which also hamper light amplification, can seldom be avoided. Using organic–inorganic halide perovskite, however, both of these shortcomings may now be overcome.

Most importantly, it is highlighted that facile tuning of the emission wavelength across the visible wavelength range can be achieved by varying the composition of the perovskite, especially the ratio of halides that comprise the perovskite structure [34].

4. Open Issues and Challenges

Fundamentally, there still remain many open issues for the perovskite material.

4.1 Efficiency

The most important factor in PV field is efficiency. Efficiency is the research focus due to related installation, occupied area and the supported scaffolds are all tightly related with it. Achieving cost effective, easily processable, efficient and versatile solar cells has always been a challenge for the researchers. As mentioned before, the 19.3% team conduct the process of fabrication perovskite solar cells in air and at low temperatures, and their team has focused on growth techniques that limit defects in perovskite crystals and on improving the interfaces between different material layers in the devices. These defects promote the recombination of electrons and holes, which drain power and reduce voltage, at the same time, impede the improvement of

efficiency. The impressive properties of perovskite could ultimately boost efficiencies as high as 25% in individual devices—as good as today's commercial champions, which are made of costly single-crystal silicon.

4.2 Poisonous Matter

There are concerns that lead toxicity will be a problem, because the perovskites are soluble in water and could be washed out of a leaky module. Lead compounds are very toxic and harmful to the environment. Thus it seems that developing lead-free perovskite is necessary for the widespread deployment of perovskite solar cells. Provide substitution of other elements for Pb is one of the important tasks for environmentally friendly perovskite solar cells.

4.3 Stability

Very few stability studies have been performed so far and the related life span reports including: In the lab, 500 h device stabilities have already been reported in dry ambient [35]. The ass-PSSC based on the PANI delivers a photovoltaic conversion efficiency reduces from 7.34% to 6.71% after 1000 h, indicating the material has a good long 36 term stability [36]. This team replaced the organic hole transport material with polymer-functionalized single-walled carbon nanotubes (SWNTs) embedded in an insulating polymer matrix and acquired a stronger thermal stability compared to cells employing state-of-the-art organic hole-transporting materials [37].

These teams investigate the perovskite solar cell by storing it in different environments. The cell stored in either dry air or nitrogen retain 80% of the initial performance after 24 hours and 20% after 6 days, whereas the devices stored in ambient air retain less than 20% of the initial performance after 24 hours and 5% after 6 days. These results show that better stability with advanced encapsulation techniques is needed for practical use of the perovskite solar cells [6].

This carbon counter electrode-based perovskite solar cell exhibits good stability over 800 h [38]. Another issue is the long-term stability of perovskites. The results of preliminary tests performed on short timescales are promising, but some researchers remain concerned that perovskites will lack the long-term stability as silicon solar cells has been testified can last for more than 25 years.

4.4 Working Mechanism

New attempts and better understanding the physics and chemical properties of perovskite materials never stop, but the fundamental properties, which the performance of these materials based, are currently exploring by researchers and are still being debated. Perovskite solar cell as a competitive candidate for new energy has soared in the improvements of structures as well as efficiency. However, identifying the basic working mechanisms will be crucial to design the optimum configuration and maximize solar cell efficiencies.

This team has investigated the local mobility, recombination, and energetic landscape of charge carriers in a prototype $\text{CH}_3\text{NH}_3\text{PbI}_3$ perovskite using laser-flash time-resolved microwave conductivity (TRMC) technique. Kinetic and frequency analysis of the transient complex conductivity strongly support the superiority of the perovskite. Their study provides a basic conception for understanding perovskite solar cell operation [39]. The transport mechanism needs to be further investigated. Singaporean photophysics expert from NTU's School of Physical and Mathematical Sciences discovered that the electrons generated in perovskite material by sunlight can travel quite far. Only few of these related reports can be found.

5. LookForward

Recent progress in perovskite solar cells has been reviewed above. All aspects about perovskite are under consideration and in experiment processes and this have triggered rapid and continuous improvements in their efficiencies. Higher efficiency is still possible through structural modification along with band gap tuning [40]. Furthermore, other perovskite structured materials such as CsSnI₃ was effectively demonstrated as hole-transporting materials (HTM) and absorber in this field. It is feasible to improve the efficiency through changing the basic structure of perovskite. As well known, the components diversity of perovskite presents the all kinds of combination, for example, Co²⁺, Fe²⁺, Mn²⁺, Pd²⁺ and Ge²⁺. There are other candidates from this family of materials that are suitable for solar cells. Perovskite is a flexible structure type and many elements can be incorporated through various structural adaptations. Thus the milestone of efficiency can be expected with the emergency some new combination one day.

To replace Pb with a less toxic element Sn is also appeared on the agenda. Nevertheless its easy oxidation character make it lowers the photovoltaic performance [41]. Other organic-inorganic halide materials based on Sn have also displayed good ability to carry charge [42], with Sn as one of the obvious candidates. Furthermore, the life span of perovskite solar cell still needs further experiments to support and testify.

The ultimate goal of the solar-cell industry is to make inexpensive devices that are highly efficient at converting sunlight into electricity. The advent of perovskite semiconductors could be the key to reaching this goal. This material can lead to advances in knowledge and breakthroughs in applied science.

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