



Review Article

Perovskite photovoltaic nanostructured materials: device architectural design, challenges and recent progress

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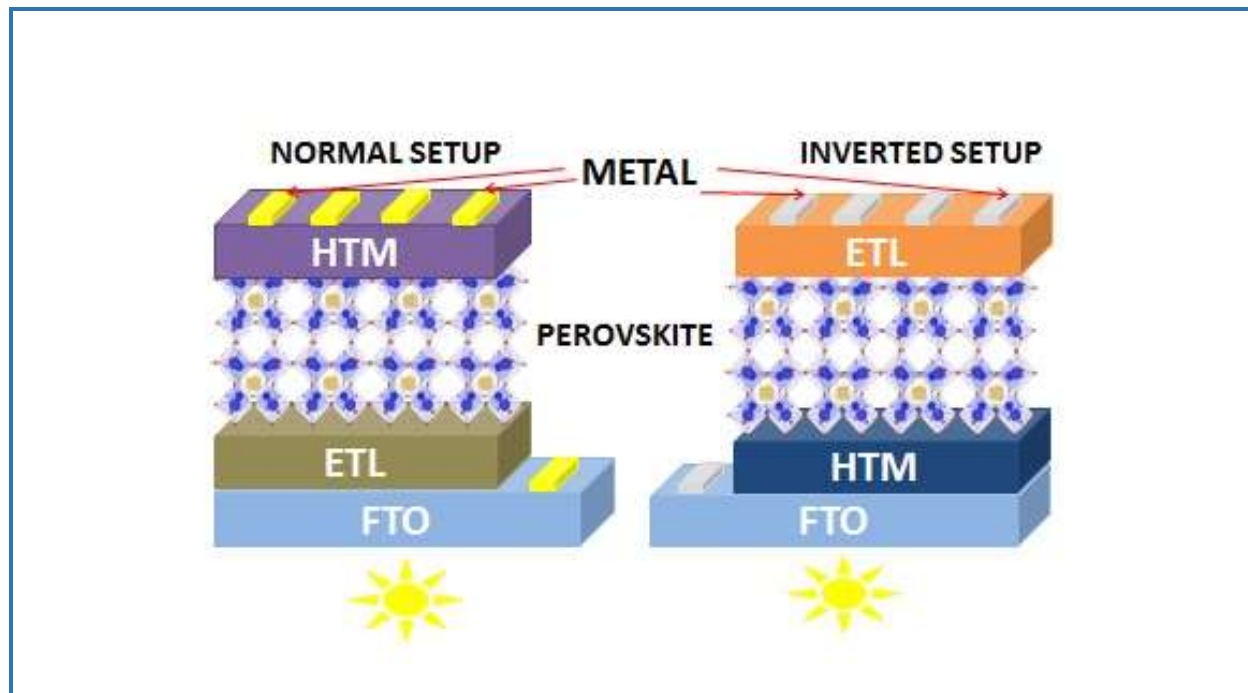
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ABSTRACT

The necessity of cheap energy from a clean and abundant source has naturally stirred a large research effort to find low cost and high efficiency devices to convert sunlight to electricity. A range of solutions which can process organic and hybrid organic-inorganic solar cells, such as dye-sensitized solar cells (DSC) and Bulk Heterojunction (BHJ) organic solar cells, have been intensely developed during the last two decades. But, the conversion efficiencies which required competing the energy market have not yet been realized. Nowadays, researchers focusing their attention to the field of solution have processed photovoltaic for the lead halide perovskite solar cell (PSC). The engineering improvements of perovskite formulations and fabrication routines have led to significant increases in power conversion efficiency (PCE%) with recent devices reaching over 22%. This review discusses the progress of perovskite solar cells focusing on device architectural design, major challenges, recent progress, and its future perspectives.

Graphical Abstract



Biographies



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Introduction

The term “perovskite” was attributed to the crystal structure of calcium titanate (CaTiO_3), which was discovered by the German mineralogist *Gustav Rose* in 1839 and named in honor of the Russian mineralogist *Lev Perovski* [1]. Meanwhile, a perovskite structure was anything that had the generic form ABX_3 and the same crystallographic structure as perovskite (The mineral). However, Perovskite was comprised of different crystalline materials with a wide range of chemical combinations. Because most people in the solar cell world are not involved with the concept of minerals and geology, perovskite and its structure are used interchangeably [2]. The power conversion efficiency of PSCs was considerably high in case of these types of cells when compared to other types (of solar cells) such as organic and dye sensitized photovoltaic devices [3]. During recent years, especially from 2012 to 2016, their efficiency had shown a tremendous increase from 9.8% to 22% overtaking the former generation of photovoltaics cells such as thin films and multicrystalline silicon cells [4]. The efficiency of perovskite was greatly attributed to its various aspects like rate of charge recombination, strong optical absorption and ease of fabrication. Efficient large area perovskite solar cells and modules had been successfully demonstrated [5, 6], indicating incipient success of up-scaling approaches. As a result of such rapid improvements, perovskite solar cells were already approaching thermodynamically achievable efficiencies. These developments had recently triggered intense activities on measure designed to go beyond the Shockley–Queisser limit predicted for a single-junction cell under standard solar illumination [7, 8]. One strategy in this area is to fabricate tandem solar cells [9, 10], whose dual absorber design allowed the utilization of low-energy photons without the open-circuit voltage (V_{oc}) drop otherwise incurred in a single-junction cell. The hybrid perovskite was prepared by simple synthetic methods which made them easy to be exploited as compared to the various existing PV technologies like excitonic dye sensitized photovoltaic cells (DSSC) and the organic solar cells (OSC). Perovskite was routinely synthesized by solid-state mixing of constituent elements or compounds at high temperatures in the range of >1300 K [11]. They could also be synthesized by drying the solution of precursor salts and the ones offering semiconductor properties

which were found to have important applications in printable electronics due to solution process ability [12, 13].

One of the most important aspects for developing solar cell devices for high performance applications was that the mobility of the charge carrier should be high [14]. Perovskite solar cells which utilize organic-inorganic metal halide semiconductors are known as “perovskite”. This perovskite active layer is comprised of halide components in ABX_3 structures with high absorption coefficient and high charge carrier mobility [15]. The perovskite family which has been typically used has been based on organic-inorganic lead perovskites with the polycrystalline structure $CH_3NH_3PbX_3$, where X is a halide atom (I, Cl, Br or a combination of some of them). These types of materials showed advantageous properties to be used as photovoltaic absorbers, namely: 1) strong optical absorption due to s-p antibonding coupling; 2) high electron and hole mobility and diffusion lengths; 3) superior structural defect tolerance and shallow point defects; 4) low surface recombination rate; and 5) favorable grain boundaries since they do not promote the electron-hole recombination [16, 17]. In fact, as compared to the most common photovoltaic systems, perovskite semiconductors showed tunable band gap, balanced electron/hole transportation, low recombination rate, carrier lifetime over 100 ns and a diffusion length over 1000 nm [18]. The high absorption coefficient of the perovskite nanocrystals made the perovskite layer very thin within the PSCs to be about the range of 400 nm.

The perovskite lattice arrangement is demonstrated in [Figure 1](#) which can be represented in multiple ways due to its various crystallographic structures. The simplest way to think about a perovskite was as a large atomic or molecular cation (Positively charged) of type A in the centre of a cube. The corners of the cube were occupied by atoms B (Also positively charged cations) and the faces of the cube by a smaller atom X with a negative charge (Anion).

Depending on atoms/molecules used in the structure, perovskite can have an impressive array of interesting properties; including, superconductivity, giant magnetoresistance, spin dependent transport (Spintronics) and catalytic properties. Therefore, *Perovskite* represented an exciting playground for physicists, chemists and material scientists [19]. These organic–inorganic perovskite materials possess some enticing ability and potential to be used in numerous applications because of their amazing properties which are as follows;

- i) They possessed very excellent characteristics of harvesting light and also served as very capable hole transporting mediums [20].
- ii) They have potentially low cost for processing.

iii) Solar cells have a promising option when the processing was done at a low temperature using the printing techniques. The printing techniques also made its deposition possible when they were deposited on a flexible substrate [21].

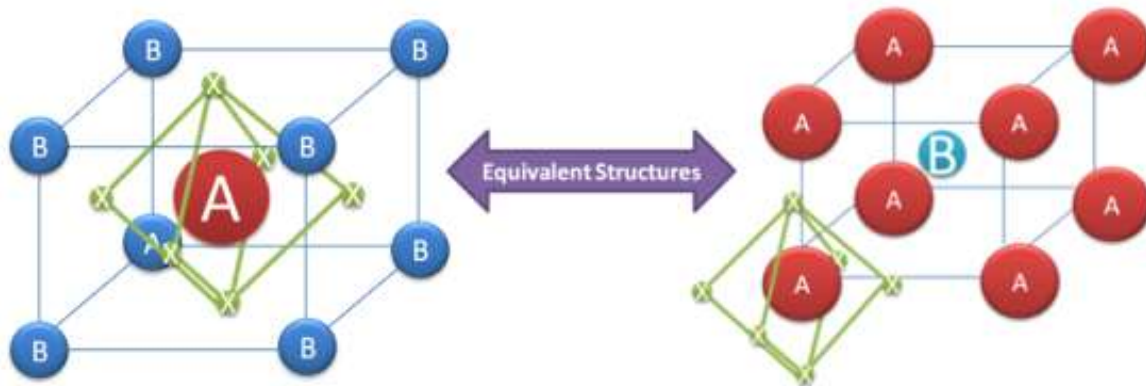


Figure 1. A generic perovskite crystal structure of the form ABX_3

iv) Due to its high absorption coefficient, the amount of light absorbed by the solar cell was also increased which ultimately increased the amount of the generated charge carriers due to the light incident i.e., photogeneration rate is increased. Accordingly, the loss in energy was also reduced and also the collection of charge carriers increased at the respective electrodes [22].

v) Because of low cost of processing and the high efficiency, these devices took less time to return back the equivalent amount of energy which was used or expended on the manufacturing of such devices. It is called the energy payback time which was low for this kind of material owing to its less cost involved in the production and also the higher performance [23].

Fundamental physics and operational mechanisms of perovskite solar cell

The common working principle of any existing solar cell was energy absorption from light source, electron-hole separation, charge collection and electron-hole recombination [24, 25]. The operational principle of a perovskite solar cell was visualized in Figure 2. In a PSC, the perovskite layer was sandwiched between a p-type layer, also called HTM (Hole Transport Material), and a n-type layer, also called ETL (Electron Transport Layer), creating a p-i-n configuration. An example of the set of materials used as ETLs and HTMs is shown in Figure 2 (left). The electron affinity of the ETL must be slightly larger than that of the perovskite absorber in order to permit an energetically efficient extraction of electrons. The working mechanism of PSC resembles the certain extent of a p-i-n heterojunction, with the intrinsic perovskite sandwiched between two extraction layers with some variations depending on the architecture [26]. Differently from inorganic homojunction silicon p-i-n diodes, the n and p are made of different materials; some of which are also in an undoped form.

The bottom layer could be the ETL or the HTM, but the processes involved were the same as schematized in Figure 2c.

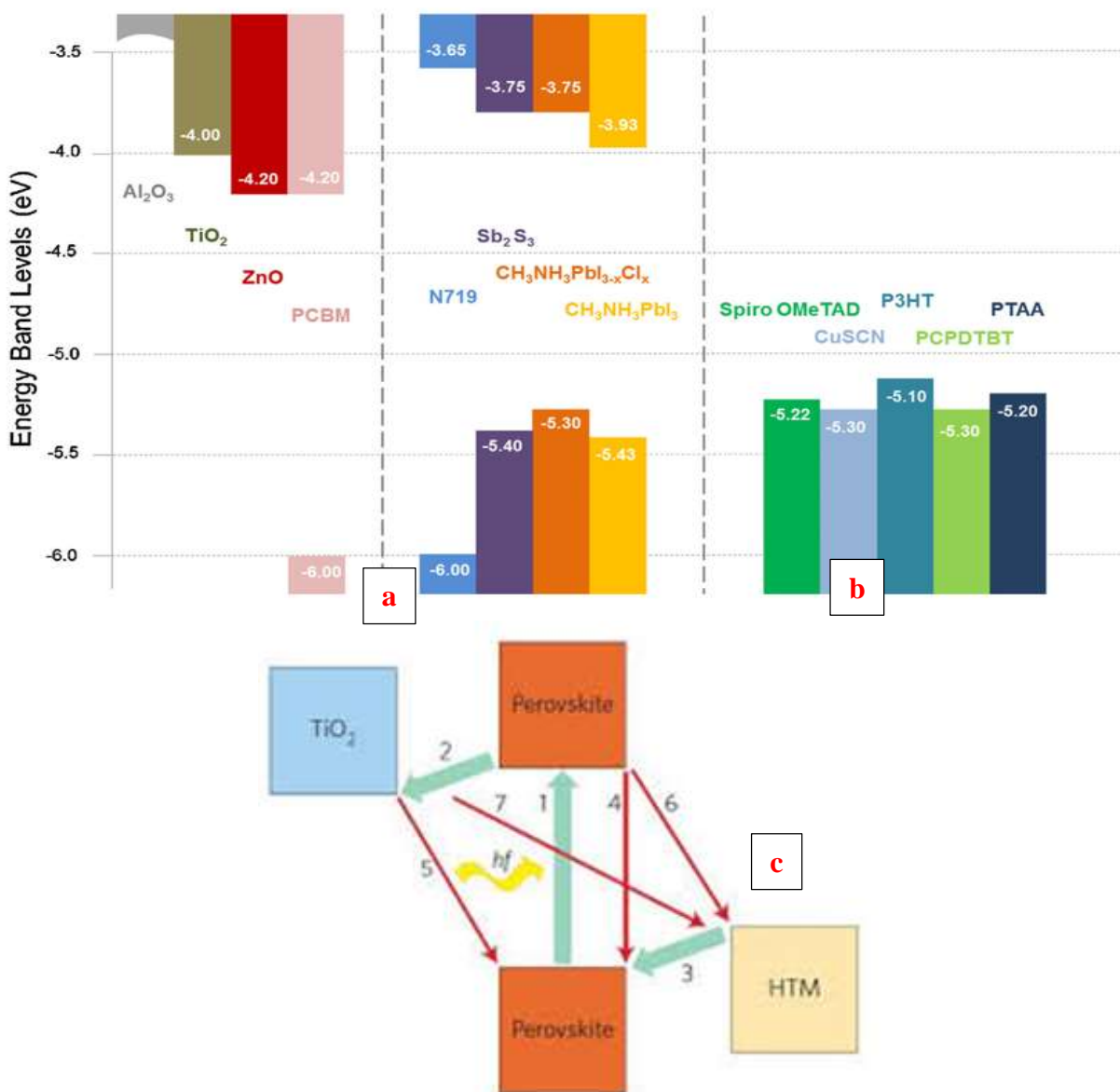


Figure 2. a) Energy levels for different materials acting as ETLs, b) absorbers and c) HTMs in perovskite solar cells. Working mechanism (Green arrows) and recombination paths (Red arrows) in a perovskite solar cell. TiO₂ can be replaced with other ETLs

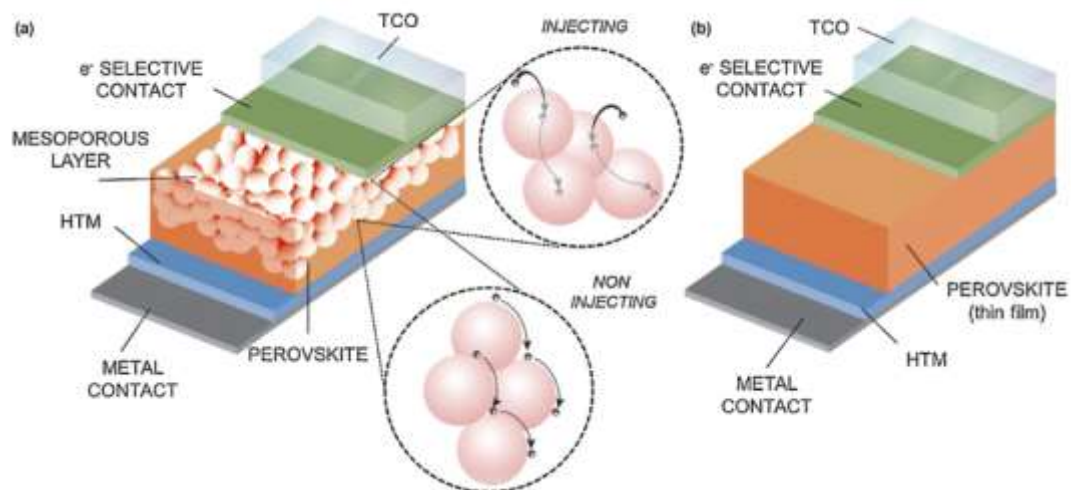
Light promoted electrons from the perovskite valence band to the conduction band (1). The weakly bound exciton split into free charges, and thus PSCs can be better represented by a free carrier model [27]. The electrons and holes can thus drift/diffuse towards the selective contacts as the charges were finally extracted (2 and 3) [28]. Charge generation and collection rates were

competing with those of the recombination. Electron-hole recombination could occur in the perovskite layer itself (4), between electrons in the ETL and holes in the perovskite (5), between electrons in perovskite and holes in the HTM (6), and between electrons in the ETL and holes in the HTM (7), especially if these come in contact due to non-uniform perovskite films. To improve the efficiency of a PSC, one has to improve light harvesting and charge extraction while also suppressing as much as possible the recombination processes.

PSCs had been fabricated in a variety of different configurations as a result of the versatility of the perovskite and extraction layer combinations. The latter had often been developed for second and other types of third generation solar cells (e.g. OPV and DSCs) [29]. To classify the different architectures used in PSCs, two main classifications were analyzed here. First, PSCs could be divided in mesostructure and planar. In the former, at least one among the ETL or HTM stack components was fashioned into a mesoporous layer, while in the latter both ETL and HTM could be considered planar/flat films (Figure 3).

Even if the top extraction layer is considered to be “planar”, there is evidence that it could infiltrate into the perovskite structure, especially at its grain boundaries [30, 31]. Using materials like phenyl-C₆₁-butyric acid methyl ester (PCBM), which were able to passivize some of the perovskite defects, enables one to achieve improved performance [32]. Moreover, the additives used in the HTM were not only dopants of the HTM itself, but also affected the perovskite layer [33].

A second classification can be made on the basis of the nature of the bottom layer on which the perovskite was deposited. Historically, the architecture was defined direct if the bottom layer was the ETL (n-i-p) or inverted in the other case (p-i-n). Four main classes of PSCs had been defined in this way: planar n-i-p PSC, mesoporous n-i-p PSC, planar inverted (p-i-n) PSC and mesoporous inverted (p-i-n) PSC.



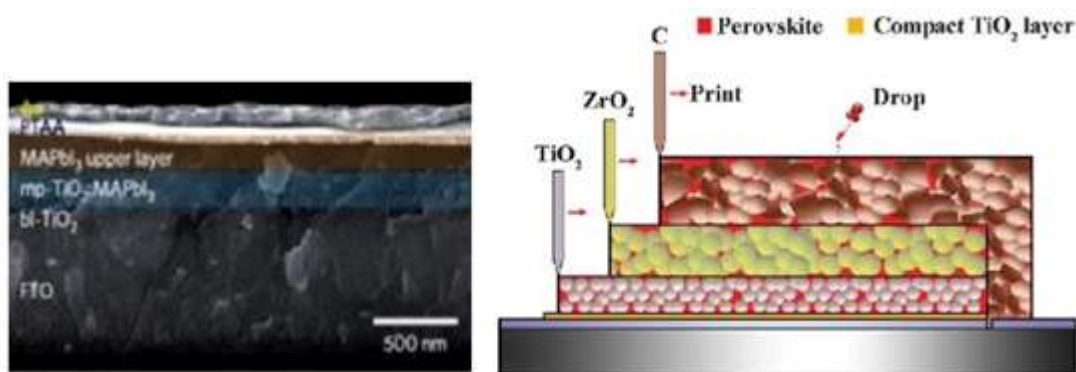


Figure 3. (Top panel) Scheme of a mesoporous a) and planar b) perovskite solar cell. (Below, left) cross-sectional SEM image of an n-i-p mesoporous PSC using TiO₂ as ETL and polyarylamide (PTAA) as HTM of the type shown in scheme a). (Below, right) scheme of the “triple-layer” perovskite structure, where the perovskite is infiltrated in a mesoporous TiO₂-insulator-Carbon stack [29]

Recent challenges of perovskite photovoltaic technology

Despite being a promising photovoltaic technology and showing impressive results in terms of efficiency and performance, perovskite solar cells still have some issues that need to be resolved before they can be commercialized. However, there were some challenges in up scaling PSCs from laboratory to commercial production; including, long-term cell stability and toxicity of the materials.

The research in developing new low-cost photovoltaics not only helped answering the constantly growing need for energy, but also created new opportunities to utilize solar cells, in for example portable, building integrated or indoor applications. Perovskite solar cells have also quite a few characteristics that make them potential candidates for portable photovoltaic applications.

In this section, the current challenges in up scaling perovskite solar cells into portable applications were reviewed. However, the poor long-term device stability of PSCs was 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 was an issue that needs to be addressed urgently for PSCs. Some researchers had shown interest in the issue of stability and given guiding opinions on improving stability [34].

Degradation of a solar cell can be caused by environmental conditions such as elevated temperature, UV illumination and exposure to ambient atmosphere, but it can also be the result of intrinsic instability [35, 36]. Another problem, particularly troublesome considering portable, personal devices, was the possible toxicity of lead included in the perovskite material.

In order to assist creating better solar cells in the future, researchers had been investigating the cause of rapid degradation of these perovskite solar cells (PSCs). For instance, the methylammonium lead iodide perovskite (MAPbI₃) had been the most widely studied. Solar cells made of this material

had been able to reach efficiencies exceeding 22% as they are cheaper to manufacture than silicon. However, their short lifespans had prevented them from becoming a viable silicon solar cell alternative.

Dr. Shenghao Wang, author of the publication in *Nature Energy*, has suggested that the degradation of MAPbI₃ perovskite might not be a fixable issue. His research revealed that iodide-based perovskite will universally produce a gaseous form of iodine (I₂), during operation, which in turn caused further degradation of perovskite.

While many researchers had pointed to other sources, such as moisture, atmospheric oxygen and heat as the cause of perovskite solar cells degradation as shown in [Figure 4](#), the fact that these solar cells continued to degrade even in the absence of these factors led some researchers to believe that the intrinsic property to these PSCs was caused the breakdown of material.

It can be observed that these PSCs are self-exposed to I₂ vapour at the onset of degradation, which led to accelerate decomposition of the MAPbI₃ perovskite material into PbI₂. *Wang* explained, because of the relatively high vapour pressure of I₂, it could quickly permeate the rest of the perovskite material causing damage to the whole PSC".

Most of the PSCs uncertainty could be attributed to the unstable perovskite materials, which are readily decomposed under environmental stress [37]. Perovskite solar cells were remarkably sensitive to humidity because of the hygroscopic ammonium ion. This was previously discovered with the very first PSCs derived from dye-sensitized solar cells, when the perovskite material was corroded by the liquid electrolytes [38]. The perovskite (here CH₃NH₃PbI₃) tends to decompose into PbI₂ and CH₃NH₃I, resulting in a decrease in the power conversion efficiency (PCE%) of the cell. PSCs stored in either dry air or nitrogen maintain 80% of their initial performance after 24 hours and 20% after six days, whereas similar cells stored in ambient air retain less than 20% of their initial performance after 24 hours and 6% after six days [39]. It appeared that humidity had an effect on the degradation of perovskite solar cells [35].

Along with cell stability, another issue that needed to be addressed was the possible toxicity of lead and overall environmental impact of perovskite solar cells, as well as the disposal and recycling of cells at the end of their service life. The European Union regulated the amounts of hazardous substances in consumer electronics. This included, for example, the lead currently used in the best-performing perovskite solar cells. Stationary photovoltaic installations were excluded from the regulations, however, portable devices such as solar powered calculators or telephones were not. The lead content in electronic devices could not be larger than 0.1% of weight [40]. Therefore, in order to realistically produce portable PSC devices with the current legislation, either the lead content had to fall under the limit, or it had to be completely replaced.

The preparation of large continuous films of perovskite in a glove box was also challenging which posed a restriction and limited its production for large scale. But, the deposition of the constituent components in a sequential manner could be done to form a thin continuous film for a wider area without degrading the efficiency of the cell [41].

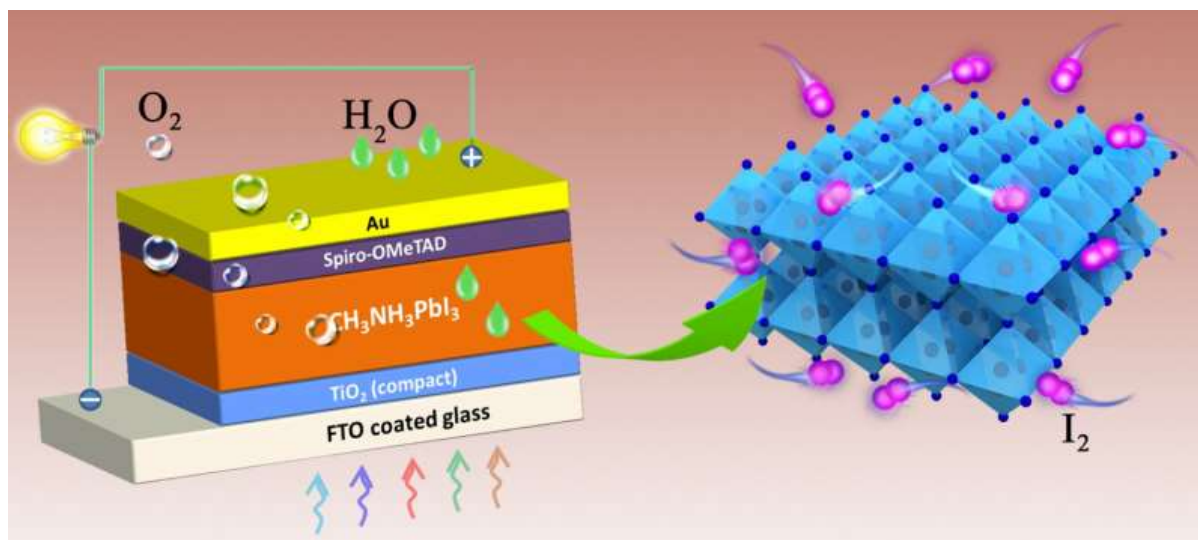


Figure 4. The schematic drawing showing that various factors (e.g., moisture, oxygen, light illumination, applied electric field, etc.) during the operation of MAPbI₃ perovskite solar cells can generate iodine, which leads to degradation of solar cells. Credit: Okinawa Institute of Science and Technology [35, 36]

Perovskite solar cell device architectures

The performance of solar cells based on organic–inorganic perovskite strongly depends on the device architecture and processing conditions. It had been shown that solvent engineering enables the deposition of very dense perovskite layers on mesoporous titania, leading to photovoltaic devices with a high light conversion efficiency and no hysteresis. However, nowadays, the most perovskite solar cells have a similar working principle as the thin-film solar cells. Because currently the most commonly used absorber material in perovskite photovoltaics is methylammonium lead trihalide [42], the working principle of perovskite photovoltaics will be focused on these materials.

The first models of perovskite photovoltaics were based on the dye-sensitized photovoltaics (Sensitizing architecture). In these photovoltaic devices, perovskite materials were deposited over a porous TiO₂ layer, and acted as an absorber coating, with excited carriers injected into hole and electron-transporting materials for collection at the electrodes. Later studies by *Loi et al.* [42] showed that perovskite layer can act as absorbent material, and also have the ability of electron-transfer ability. In this sense, more simple designs of perovskite photovoltaics, based on the thin-film

structure, were developed. Some studies have also tried to achieve higher efficiencies for both of the designs of perovskite photovoltaics. By our knowledge, a dominant design with highest efficiency and best performance has not been established yet. The main difference between these two photovoltaic designs was how the perovskite layer was positioned? In the sensitizing-architecture, the active layer, consisting of mesoporous TiO_2 , was coated with the perovskite absorber. The active layer was contacted with n-type and p-type materials for electron-extraction and hole-extraction, respectively. Thus, the perovskite layer acts only as a light absorbent material, and other materials were accountable for the charge carrier function [42]. In an alternative design, thin-film perovskite solar cell, perovskite layer as a flat layer, was sandwiched between hole-transporting layer and TiO_2 layer. In this architecture perovskite layer acted as an absorber material and a charge carrier layer. [Figure 5a](#) and [b](#) show two design architectures of the perovskite solar cells.

In the [Figure 6a](#), perovskite layer has only the function of charge carrier layer, while [Figure 6b](#) is described below;

- Glass layer: Functions as a protection for the other parts of the solar cell.
- Fluorine-doped tin oxide (FTO): An electrically conductive and transparent coating layer. Because it is stable under the atmospheric conditions and resistant to high temperatures to perform well in solar cells. It is cheaper than indium tin oxide (ITO) which is another material that can perform as same as that of FTO.
- TiO_2 layer: It behaves as a semiconducting layer and acts as an electron charge absorber and charge carrier.
- Perovskite layer: The main layer of cell, it absorbs light and it functions as a charge carrier.
- Hole transporting material: An organic hole-carrier material. In these materials the electric conductivity depends on uncontrolled oxidative process.
- Metal/Gold layer: A highly conductive layer of metal for the extraction and transportation of electrons.

Conclusion and future perspectives

Perovskite solar cells have attracted great interest during the recent years, because of their excellent light absorbing properties, simplicity of processing and wide tenability. Moreover, they offer a competitive alternative for conventional photovoltaics, and perform well in many areas where the current commercial technologies fail. While their lab-scale efficiencies are impressive, however similar values are yet to be obtained or have been reported in literatures. Nevertheless, perovskite solar cells may well be the best-performing low cost photovoltaic technology.

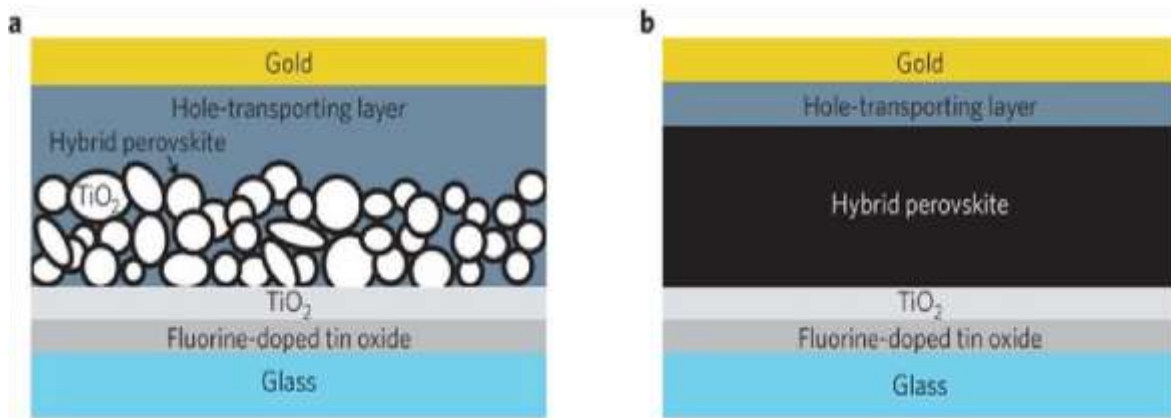
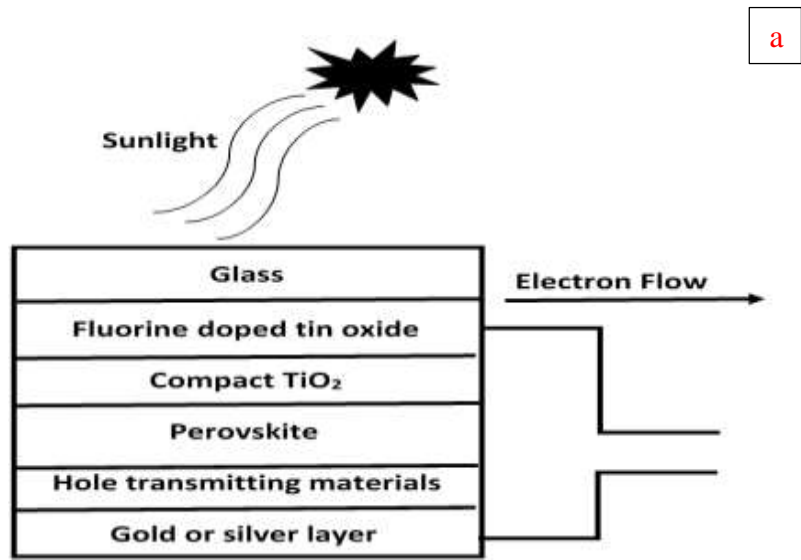
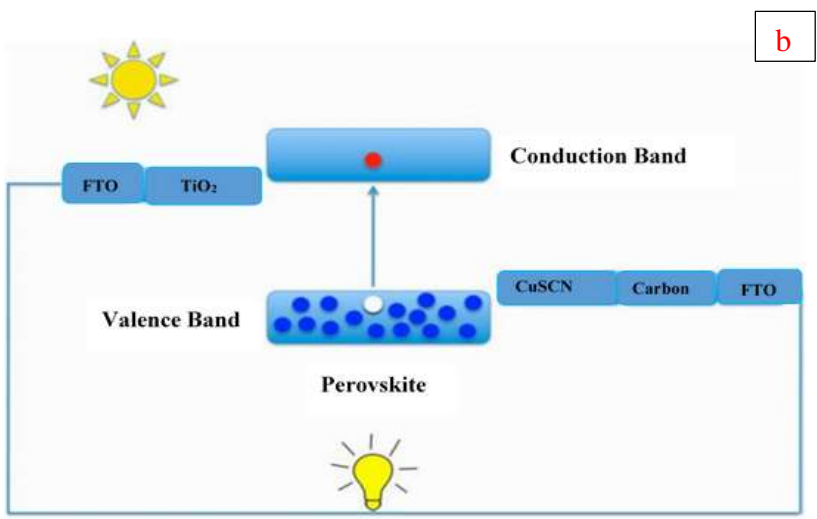


Figure 5. Architectures of perovskite solar cells, a) Hybrid perovskite sensitized architecture solar cell on mesoporous TiO_2 , b) Planar hybrid perovskite solar cell

Figure 6. a) Architecture of a perovskite solar cell , b) Mechanism of perovskite solar cell to produce electricity from sun light



a



b

Even though very high efficiencies have been reported, the stability of these devices is still a matter of concern; hence the commercialization of PSCs will be only possible when the poor stability of the perovskite solar cells under ambient conditions is solved. This drawback has challenged the research community to develop good sealing strategies to avoid degradation mechanisms mainly related to oxygen and moisture exposure under outdoor operation conditions, and also the intrinsic phenomena as ion migration, electro migration and interface reactions are still under scrutiny.

A broad range of architectures and fabrication methods have been proposed, as well as several perovskite compositions and charge selective layers. Unfortunately, the present information obtained on “how PSCs operate?” is rather insufficient at least for now, because the complete working principle is not well explained [43, 44]. Therefore, deriving novel clues on 1) light absorption, 2) charge separation, 3) charge transport, and 4) charge collection are needed during research since these four key issues are general solar cell parameters to identify the principal working of solar conversion process.

The novelties in PSC technology attract a large amount of attention in our recent world due to its significance and effectiveness in electricity production from free and abundant sunlight. The main prospects for the future cover the strategies in terms of reducing the band gap, enhancing the light absorption capacity, and improving the conversion efficiency. In addition, exposing the charge transport properties and improving interfacial engineering methods in device fabrication are important. Especially, the chemistry work on chemical composition has great value. Hence, future work should explain the photophysical mechanism of solid-state solar cells with respect to different chemicals.

Although, due to the constantly dropping cost of silicon solar cells, and also because of the issues with cell stability, perovskite solar cells may have a hard time replacing silicon-based photovoltaics in energy grid integration. However, they do exhibit better performance in non-ideal lighting conditions and can achieve mechanical properties, such as flexibility, that silicon cells cannot. Therefore, they could be considerable candidates for the portable photovoltaics.

Finally, researchers are apt to announce that the future perovskite solar cells should be developed with a reduced concentration of iodine or a reinforced structure that can suppress iodine-induced degradation, in addition to desirable photovoltaic properties. In this sense, the difficulties, which are mainly about stability, are overcome, and also the humanity would face a new and affordable solar energy harvester for future use. Hopefully, better and cheaper solar cells will entice more people to utilize this technology.

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Disclosure statement

No potential conflict of interest was reported by the authors.

Abbreviations	
<i>SFE</i>	<i>Supercritical fluid extraction</i>
<i>SE</i>	<i>Solvent extraction</i>
<i>SD</i>	<i>Steam distillation</i>
<i>MAHD</i>	<i>Microwave assisted hydrodistillation</i>
<i>USWE</i>	<i>Ultrasound-enhanced subcritical water extraction process</i>
CaTiO_3	<i>Calcium titanate</i>
V_{oc}	<i>Open-circuit voltage</i>
<i>OSC</i>	<i>Organic solar cells</i>
<i>DSSC</i>	<i>Dye sensitized photovoltaic cells</i>
<i>HTM</i>	<i>Hole transport material</i>
<i>ETL</i>	<i>Electron transport layer</i>
<i>PCBM</i>	<i>Phenyl-C61-butyric acid methyl ester</i>
MAPbI_3	<i>Methylammonium lead iodide perovskite</i>
I_2	<i>Iodine</i>
<i>PCE%</i>	<i>Power conversion efficiency</i>
<i>FTO</i>	<i>Fluorine-doped tin oxide</i>
<i>ITO</i>	<i>Indium tin oxide</i>
%	<i>Percentage</i>
<i>OPV</i>	<i>Organic Photovoltaic</i>
<i>DSCs</i>	<i>Dye sensitized Solar cells</i>
<i>PSC</i>	<i>Perovskite solar cell</i>
<i>ns</i>	<i>Nanoseconds</i>

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