A Fundamental Review of Trends in Perovskite Solar Cells Based on Introduction of Buffer Layers to Enhance Stability

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ABSTRACT

Perovskite solar cells (PSCs), as photo-electric conversion devices, exhibit adequate power conversion efficiency (PCE) and low processing cost and have thus become one of the most promising devices to replace conventional silicon-based solar cells thereby addressing current prevailing energy issues. Despite rapid progress in the perovskite solar cell efficiency, there have been concerns about issues which could affect the measurement accuracy and/or practical applications of these devices, amongst which are stability, scaling up (large area devices), and possible environmental effects related to the use of lead-based active material. Current PSCs use non-biodegradable petroleum-based polymer substrates, discarding of which will aggravate environmental pollution. In this study, a review of the fundamentals and progress in the perovskite solar cell (PSC) is discussed. The processing cost of metal electrode deposition in PSC is very costly, hence, the replacement of the metal electrode by some other low-cost counter electrode is discussed.

INTRODUCTION

Emerging photovoltaic technologies have made it possible and easy to convert the energy from the sun into electricity at a low cost and with relatively high conversion efficiencies. Gradually, primary energy resources such as fossil fuels, coal, and natural gas are depleting, while global energy consumption is increasing. Solar, wind, biomass, tidal, and geothermal sources of renewable energy are emerging as solutions to the energy deficiencies and other related issues in our world today. These renewable energy resources are freely available in nature, non-polluting, and help in reducing global CO_2 emissions.

The advancement in solar cell technology is to produce a cheap (cost-effective and easy to produce), high efficiency, and long lifetime (stable) solar cell which is a better replacement for energy generation from fossil fuel. Research has been going on, to meet these ultimate goals in photovoltaic technology which has led to the discoveries of new materials and new techniques in solar cell fabrication. Solar cells are categorized into three main groups known as generations based on their order of appearance in the market (Balema, 2009). Available PV cells can be broadly classified into first, second, and third-generation solar cells. First-generation cells are silicon-based crystalline cells while second-generation cells are thin film-based, and third-generation cells comprise new emerging technologies.
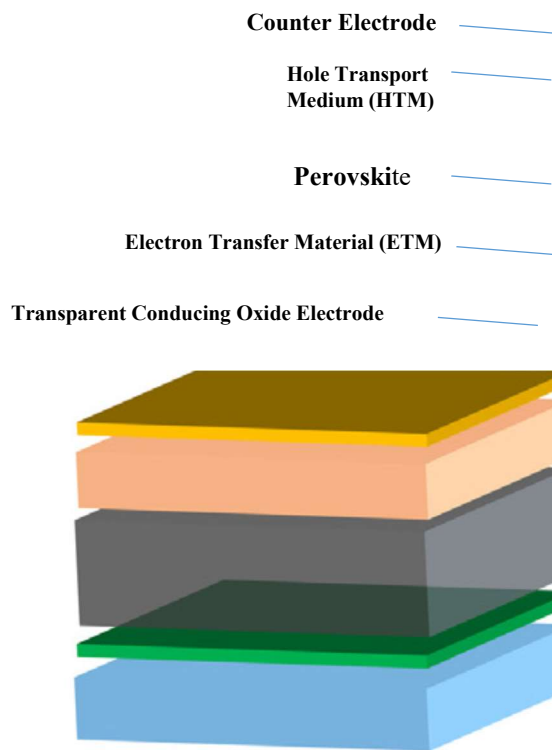
With the relentless effort of researchers in photovoltaics, a new type of solar cell which is based on organic-inorganic hybrid solar cell known as perovskite was discovered in 2006 by Miyasaka and his co-workers (Miyasaka & Kojima, 2009). This new material has the highest reported efficiency of approximately 22.1% (Shin, et al., 2017). These hybrid solar cell technologies such as perovskite-based solar cells are classified as such because their photoactive layer is made of organometallic material. This work will focus purely on this organic-inorganic perovskite-based solar cell which also shall be limited to a particular type known as methylammonium lead tri-iodide ($CH_3NH_3PbI_3$).

THEORETICAL FUNDAMENTALS
Perovskite Solar Cells (PSCs)

Perovskites are a class of materials that share a similar structure, which displays a myriad of exciting properties like superconductivity, magneto-resistance, and more. Perovskite was discovered in 2006 by Miyasaka and his co-workers (Miyasaka & Kojima, 2009). A perovskite solar cell is commonly a kind of solar cell with a perovskite structured compound, most typically a hybrid organic-inorganic lead or tin halide-based material, as the light-harvesting active layer (Hossain, 2018). This work will focus purely on this organic-inorganic perovskite-based solar cell which also shall be limited to a particular type known as methylammonium lead tri-iodide ($CH_3NH_3PbI_3$). These readily synthesized materials are seen as the potentials of solar cells since their photovoltaic conversion efficiencies and cost are determined by their distinctive structures. These PSCs have been predicted to play a role in the next-generation electric vehicle batteries, sensors, lasers, and many others.

Counter Electrode

Hole Transport Medium (HTM)

Perovskite

Electron Transfer Material (ETM)

Transparent Conducing Oxide Electrode

Figure 1: The Architecture of the PSCs (Francesco, Azhar, Rajan, & Thomas, 2016)

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From figure 1 above, depending on the architecture of the device, different interfaces are formed and consequently different trends in the solar cells have been observed. Generally, PSCs are formed from the following fundamentals layers:

a. Photo-Anode as Electron Transport Layers (ETL); which is a composite of n-type Fluorine-Tin Oxide materials as a transparent conductive layer and TiO_2 as a compact layer on Transparent conducting oxides (TCO) of PSCs.

b. Absorption layer; composites of TiO_2 or Al_2O_3 or both, carrying perovskite crystalline materials, the scaffold of perovskite materials, and serving as light harvester light in organic PSCs.

c. Hole Transport Layers (HTL); which is a p-type semiconductor such as the widely used HTMs like spiro-OMeTAD, CuI, C, or polyaniline.

d. Top contact electrode; such as Al, Ag, or Au thin film electrode which is deposited on the HTM layer. The transparent electrode and the metallic electrode are connected to create a closed circuit cell.

Characterization of Photovoltaic Performances

The performance of the DSC is mainly characterized by the following parameters such as incident photon to current efficiency (IPCE), short circuit photocurrent density (J_{sc}), open-circuit photovoltage (V_{oc}), the fill factor (FF), and the overall efficiency of the photovoltaic cell (η).

Incident Photon to Current Conversion Efficiency (IPCE)

The spectral response of the dye-sensitized solar cell depends on the absorption properties of the dye. The incident photon to current conversion efficiency (IPCE), also known as an external quantum efficiency (EQE), is one of the key parameters of quantitative characterization of the solar cell performance for the fact that IPCE shows the spectral response of solar cells to various incident wavelengths. It measures how efficiently the incident light of a specific wavelength is converted into electron/hole pairs, by measuring the ratio between the number of electrons generated by the solar cell and collected at the external circuit and the number of photons of a given wavelength shining on the solar cell. The IPCE can be calculated according to the equation.

$$IPCE = \frac{1240.J_{sc}}{\lambda.P_{in}}$$

(2.1)

Where J_{sc} is the short circuit photocurrent density (mA/cm^2), λ is the wavelength (nm), P_{in} is incident power (mW/cm^2).

Short Circuit Photocurrent Density (J_{sc})

The short-circuit current density is the maximum photo-generated current delivered by a solar cell when the terminals of the solar cell are in contact with each other (i.e. short-circuited). J_{sc} is the photocurrent per unit area (mA/cm^2) when an illuminated cell is short-circuited where the external output voltage is zero. It depends on several factors such as the light intensity, light absorption, injection efficiency, regeneration of the oxidized dye, and the efficiency of charge transport in the TiO_2 film to the counter electrode. It is strongly related to the IPCE and the theoretical values of the J_{sc} can be calculated from the integrated sum of IPCE measure over the entire solar spectrum.

$$J_{sc} = \int_0^\infty IPCE(\lambda).I_{sun}(\lambda)d\lambda$$

(2.2)

$$J_{sc} = J(V) + J_o \left(e^{qv/mK_BT-1} \right)$$

(2.3)

Where V is the voltage across the junction, T is the absolute temperature, J_o is the dark saturation current density and m is the ideality factor with values between 1 and

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2. The electric charge $q = 1.603 \times 10^{-19}C$ and the Boltzmann's constant, $K_B = 1.38 \times 10^{-23}JK^{-1}$

Open Circuit Photovoltage (V_{oc})

V_{oc} is measured when the circuit is open or no external load is connected. Under this condition, there are no external current flows between the two terminals i.e. I = 0 and V = V_{oc}. It depends on both the Fermi level of the semiconductor and the level of dark current. It is the maximum voltage that a solar cell can deliver. The theoretical maximum of the V_{oc} of the cell is determined by the difference between the Fermi level of the semiconductor and the redox potential of the electrolyte. The V_{oc} depends on the photogenerated current density J_{ph},

$$V_{oc} = \frac{mK_BT}{q}\ln\left(\frac{J_{ph}}{J_o} + 1\right)$$

(2.4)

Optimum Voltage (V_m)

V_m is the voltage at the optimum operating point at which the PSC output power is maximum.

Optimum Current (J_m)

The value of J_m gives the maximum photocurrent obtainable at the maximum PowerPoint.

Output Power (P_{out})

For a given bias voltage the output power of the cell is the product of the measured current density and the bias voltage

$$P_{out} = V \cdot J$$

(2.5)

P_{out} generally increases when V is increased from zero, goes through a maximum, and decreases to zero at V = V_{oc}. The maximum power point Pm is the product of J_m and V_m

Fill Factor (FF)

Fill factor is a parameter related to the maximum power point and is often described as a measure of the "squareness" of the J-V curve and describes the degree to which the voltage at the maximum power point (V_m) matches V_{oc} and that the current at the maximum power point (J_m) matches J_{sc}. Its value is determined by the ratio of the maximum power (V_mJ_m) to the maximum power attainable by the solar cell ($J_{sc}V_{oc}$), with a value between 0 and 1.

$$FF = \frac{V_mJ_m}{J_{sc}V_{oc}}$$

(2.6)

Solar Energy to Electricity Conversion Efficiency (η)

The overall solar energy to electric power conversion efficiency (η), the key parameter of the device, measures how much power is converted by the cell in comparison to the amount of absorbed light that reaches the device. It is given by the ratio of the maximum output power (V_mJ_m) to the incident solar power (P_{in}). The overall sunlight to the electric power conversion efficiency of a DSC is given by the following expression

$$\eta = \frac{P_m}{P_{in}} = \frac{J_{sc}V_{oc}FF}{P_{in}}$$

(2.7)

ADVANCED RESEARCH IN PEROVSKITE SOLAR CELLS

The first record of perovskite-based solar cell efficiency was reported by Miyasaka et al. (2009), only less than one decade ago. They reported an efficiency of 3.8% based on a dye-sensitized solar cell structure. Due to the application of liquid electrolyte in the hole-transporting material (HTM), the stability of the solar cells was very weak and did not attract much attention. A similar trial was done by Park and his group (2011) with increased efficiency of 6.5% but stability was still the main problem because of the instability of HTM layer due to the liquid medium.

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The application of solid-state HTM (2, 20, 7, 70 – tetrakis (N, N-di pmethoxyphenylamine)-9, 90-spirobifluorene, i.e., Spiro-OMeTAD), rather than liquid HTM, onto the highly-crystallized perovskite layer triggered the efficiency boosting during the past several years. Lee et al. (2012) reported a breakthrough device efficiency of 10.9% with the open-circuit voltage higher than 1.1 V. Wang et al., (2014) introduced graphene into PSCs and acquired an efficiency of 15.6% while the application of another perovskite material, formamidinium iodide (HC (NH$_2$)$_2$ PbI$_3$) together with poly-triarylamine (PTAA) as a new HTM brought a remarkable 20.1% efficiency in 2015 (Yang, et al., 2015). Another record efficiency of PSCs was 22.1%, created in 2016 by Shin et al. They also accomplished a long-term and stable efficiency of 21.2% in another work (Shin, et al., 2017).

Buffer Layers in PSCs

Buffer layers are commonly used in the optimization of thin-film solar cells. In addition to affecting the open-circuit voltage of a cell, it is commonly reported that buffer layers affect stress-induced degradation and transient phenomena in PSCs. The primary function of a buffer layer in a heterojunction is to form a junction with the absorber layer while admitting a maximum amount of light to the junction region and absorber layer.

Guarnera et al, (2015) deposited the HTM layer within an insulating mesoporous buffer layer comprised of Al$_2$O$_3$ nanoparticles to prevent metal electrode migration while allowing for precise control of the HTM thickness. This was reported to have enabled an improvement in the solar cell fill factor and prevented degradation of the device after 350 h of operation.

Yeh and his group in 2016 demonstrated a PSC using solvent treatment on active layer and a thin buffer layer. The morphology of the perovskite active layer

agglomerates owing to methylbenzene treatment and the PSC showed an increased Jsc. The buffer layer 1, 3, 5-tris(N-phenylbenzimidazol-2-yl)benzene (TPBi) enhanced the device performance by formation of well interfacial contact and restraining the hole from transporting to the cathode. The optimized PSC showed a PCE of 7.81%

Nouri et al (2017) introduced graphene oxide as a buffer layer and promoted soluble n-butyl-substituted copper phthaocyanine as an efficient HTM in organometal halide PSCs. Constructed and characterized under ambient conditions of 50%-60% humidity, soluble n-butyl-substituted copper phthaocyanine yielded a modest PCE of 7.3% but with the introduction of a buffer layer of either Al$_2$O$_3$ or graphene oxide between the perovskite and the hole transporting layers, the PCE extensively increased and reached 14.4%. Corresponding data obtained by employing the standard spiro-OMETAD as HTM gave equivalent performance.

Bathocuproine (BCP) was employed by He et al (2018) as the interface buffer layers between PCBM and Ag layers to improve the performance of the PSC based on NiOx HTL. The effect of the different evaporation times on the performance of solar cells at the same evaporation temperature was carefully investigated. The optimum device efficiency of 14.47% was obtained by evaporating BCP for 36 s suggesting an enhancement of 14% as compared to the cell without BCP modification.

Recently, Shibayama et al (2019) reported that, in the inverted structure PSCs, a buffer layer is generally used at the interface between the n-type semiconductor layer and the metal electrode with a controlled thickness of bathocuproine (BCP) deposited by thermal evaporation. The ideal factor greatly decreased from 5 to 1.4 by introducing the BCP buffer layer and it was

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confirmed that the interface between the n-type semiconductor layer and the metal electrode gradually changed from Schottky barrier diode to an ohmic contact. Excessive BCP film thickness caused the series resistance to increase and also induced recombination. As a result of optimizing the perovskite layer and the BCP buffer layer respectively, the PCE of 17% was obtained.

HTL-Free Perovskite Solar Cells

One of the ways to reduce the fabrication cost of PSC is a fabrication of ETL or HTL free perovskite solar cell. The counter electrode is directly coated either on perovskite film or ETL. Perovskite film acts as an absorber layer as well as the HTM.

The first HTM-free heterojunction PSC was reported by Etgar et al in 2012 by deposition of perovskite nanoparticles from a solution of CH_3NH_3I and PbI_2 in Y-butyrolactone on a 400 nm thick film of TiO_2 nanosheets exposing facets. Au film was then evaporated on the perovskite as a back contact. The PSC yielded an impressive photovoltaic performance with J_{sc} = 16.1 mA/cm^2, V_{oc} = 0.631 V and FF = 0.57 corresponding to a PCE of 5.5% under AM 1.5 solar light of 1000 W/m^2 intensity.

Shi et al. (2014) demonstrated a modified two-step deposition method to fabricate a uniform perovskite capping layer with high coverage and thickness of 300 nm on mesoporous TiO_2. The perovskite layer showed high light-harvesting efficiency and long carrier lifetime over 50 ns and achieved a PCE of 10.47%.

Zheng and coworkers in 2016 explored the use of poly (3-hexylthiophene) (P3HT) for the cathodes of Hole transport free mesoscopic PSCs, simultaneously achieving high performance, high stability and low-cost PSCs. The thin P3HT modifier acts as an electron blocker to inhibit electron transfer into carbon nanotubes (CNTs) and a hydrophobic polymer binder to tightly cross-link the CNTs together to compact the carbon electrode film and greatly stabilize the cell. The presence of CNTs greatly improved the conductivity of P3HT. By optimizing the concentration of the modifier (2 mg/mL), there was an improved PCE of up to 13.43%. Hysteresis and degradation of the CNTs@P3HT based PSCs were strongly retarded as compared to cells employing the pure CNTs electrodes when exposed to ambient conditions of 20-40% humidity.

This research indicates that the incorporation of the interlayer is helping to block holes and promote electron transport and hence enhances the efficiency of the device.

Replacement of Metallic Counter-Electrode by Cost-Effective Carbon

As we can see almost all PSC need a metal electrode to complete the structure. A metal electrode (the counter electrode) is very much useful to prevent the PSC from degradation. The deposition method used for metal coating needs costly instrument (physical vapour deposition) and high vacuum, which requires a very high cost for solar cell fabrication. An effective way to reduce the fabrication cost is the use of another less costly counter electrode. Carbon is the best alternative material for replacement of metallic electrode as it does not require high vacuum and costly instrument for deposition. It can be coated by simple screen printing or doctor's blade method. Many researchers have used carbon paste/carbon nanotube/carbon black as counter electrode and got good efficiency.

Ku et al (2013) developed a mesoscopic methyl-ammonium lead iodide perovskite/TiO_2 heterojunction solar cell with low-cost carbon electrode and full printable process which presented high stability and power conversion efficiency of 6.64% relatively higher than that of flaky graphite based device and comparable to the conventional gold version.

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Wei and his colleagues in 2014, developed a planar PSC that incorporates a nanocarbon hole-extraction layer by an inkjet printing technique with a precisely controlled pattern and interface. By designing carbon plus CH_3NH_3I ink to transform PbI_2 in situ to $CH_3NH_3PbI_3$, an interpenetrating seamless interface between the $CH_3NH_3PbI_3$ active layer and the carbon hole-extraction electrode was instantly constructed with markedly reduced charge recombination compared to that with the carbon ink alone. As a result, a considerably higher PCE up to 11.60% was delivered by the PSC.

Zhang et al (2015) applied mesoporous graphite/carbon black counter electrodes using flaky graphite with different sizes in HTM-free PSCs by screen printing technique. The results indicated that, graphite which acts as a carbon counter electrode could significantly affect the square resistance, thus resulting in differences in fill factor and PCE of the devices.

Duan and his group in 2017 employed Boron-doped graphite for high work function carbon electrode in printable hole-conductor-free mesoscopic perovskite solar cells, which improved the conductivity of the carbon electrode as a result of a higher graphitization carbon of boron-doped graphite resulting in a low charge transfer resistance at the carbon/perovskite interface and an extended carrier recombination lifetime. This yielded a PCE of 13.6%.

Xiao et al (2017) reported the impact of doping Titanium oxide nanoparticles with different amounts of tungsten (W) on the PV properties of HTM-free PSCs that employ carbon counter electrodes. Light doping with W (less than 1000 ppm) improves the PCEs of solar cells by promoting the electron conductivity in the TiO_2 layer which facilitates electron transfer and collection. An efficiency of up to 10.53% was realized (Xiao, et al., 2017)

DISCUSSION OF THE FINDINGS

In the present market, stability issue is likely a considerably more significant problem than the presence of lead in the active layer for wide deployment of the devices. The toxicity of lead-based perovskite solar cells is not a major impediment to large-scale, professional applications, as is evidenced by the fact that CdTe cells have already gained a reasonable market share. Cd or Pb is also present in some CIGS and silicon modules at the same general level as that likely in perovskite modules (Werner, et al., 2011). The danger is that technology relying on toxic materials may be increasingly marginalized as legislation becomes increasingly more pervasive and restrictive, yet the pollution in PSCs is relatively insignificant compared with other lead pollutions (Fabini, 2015), and the production of PSC could be able to use a waste lead from daily waste (Chen, et al., 2014). Researches have established basically that perovskite solar cells have the inherent potentials of reaching very high power conversion efficiencies when proper permutation of the various cell components is done. From the findings, it has been observed that no research into the suitability of starch film as a buffer/desiccant for PSCs has been done, especially with the effective combinations of chitosan-binded TiO_2 as ETM and carbon as the counter electrode. The above research efforts indicate that the PSCs will have a greater potential for commercialization if the stability of cells can be improved.

CONCLUSION

Perovskite solar cells (PSCs), a photoelectric conversion devices, exhibit excellent power conversion efficiency (PCE) and low processing cost and have become one of the most promising devices to replace conventional silicon-based solar cells and address current pressing energy issues. Nevertheless, the majority of research

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efforts have been concentrating on the improvement of efficiency, which has now exceeded 22%. Progress has been made in manufacturing larger area cells as well as modules, which is of interest for commercialization of the technology. However, based on life cycle assessments, the key issue appears to be the lifetime of the devices. Substantial improvements are needed in the stability of PSCs to make them commercially competitive. To achieve these improvements, issues which require attention is the perovskite material composition and film quality, the choice of charge transport layers, electrodes and barrier layers and encapsulation strategies.

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