Fabrication Strategy to Promote Performance of Perovskite Solar Cells

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Abstract. Improvements in process of perovskites, materials of auxiliary layers and encapsulation have significantly enhanced the performance of perovskite solar cells (PSCs). However, unified fabrication of PSCs has not been completely settled till now. Whether it is harmful to the perovskite should be concerned when selecting function layer materials and encapsulation materials of PSCs. Encapsulation is the main way to enhance the stability of PSCs. Besides, to cope with the emerging environment issues, the function layer materials can be modified to adsorb lead, preventing the leakage of lead from PSCs. To integrate the advantages of each part of PSCs, interactions between constituent materials are needed to be studied.

1. Introduction
The problem of energy shortage has been a big problem that needs to be solved imperatively. Utilizing of solar energy has become attractive because of its inexhaustibility. Many types of solar cells have been developed such as silicon solar cells, organic solar cells and thin-film solar cells, etc. Over the recent years, organometal halide perovskites have attracted more and more attention and have been rapidly developing[1,2], due to its low-cost and simple processing. After the first report of perovskite sensitized photovoltaic cells, many achievements have been made to promote the performance of perovskite solar cells (PSCs). Many structures have been developed such as mesoscopic structured device and planar heterojunction device. According to the recent research, the power conversion efficiency (PCE) of PSCs has been highly promoted to more than 22%[3].

Adjusting components of perovskites is a valuable research topic. For example, some types of polymer are immingled in perovskites so as to reduce defects in the film, such as aliphatic fluorinated amphiphilic, 4-vinylpyridine, ethylene glycol and some semiconducting polymers, etc. Some researches focus on mixed-cation perovskites. It has been proved that stability of perovskite can be improved by mixing it with Cesium ions[4]. Besides, it has been found that the performance of perovskite is related to the content of Br. Stability can be enhanced with the optimization of chemical elements[5].

Improvments have also been made in fabrication of PSCSs such as process of perovskites and materials of auxiliary layers. These improvements significantly enhanced the efficiency and stability of PSCs. However, unified fabrication of PSCs and material selection have not been completely settled till now. There still remains some issues to be improved, including poor stability under external conditions, initial efficiency and leakage of lead. In this articles, the PCE after fabrication process is termed as initial efficiency. Thus, much effort is still required to be done to commercialize PSCs.
Some feasible strategies on promoting resistance of degradation and avoiding leakage of lead are developed. The object of this work is to present the latest results on this subject.

2. Stability

Poor stability is the main obstacle of the commercialization of PSCs. The long-term stability is intensively impacted by environmental stability, thermal stability and photo-stability\cite{6-8}. Poor stability of PSCs commonly results from degradation, which can be accelerated by heat, ultraviolet light (UV-light), oxygen and moisture, etc\cite{6,7}. For example, it has been demonstrated that methylammonium lead iodide perovskite will degrade when confronting with water molecules\cite{9} and thermal circumstance\cite{10}. Degradation also occurs inside PSCs. It has been tested by experiments lifetime of PSCs is about 0.7 years at 35 ℃ if take 25% degradation as standard\cite{11}. It is rather shorter than lifetime of over 20 years for crystalline silicon solar cells\cite{12}. Typically, the PSCs with a structure of anode/HTL/perovskite/ETL/cathode exhibits excellent performance, in which HTL and ETL stand for hole transport layer and electron transport layer respectively. Causes of degradation can be divided into internal and external factors. Internal factors include heating in necessary process, intrinsic heating under applied voltage, etc. All possible internal degradation in PSCs can be avoided by careful interface engineering, such as good selection of materials, ion hybridization in perovskite layer, etc\cite{13}. Thus the oxygen and moisture are believed as main factors that cause degradation, because of the moisture sensitivity of perovskite materials. Water promotes the decomposition reaction as a catalyst.

Encapsulation is the main way to enhance the stability of PSCs. According to the properties of perovskite, encapsulation should be resistance to UV-light, moisture and thermal induced degradation. To fulfill the function of photovoltaic cells, encapsulation material should exhibit high dielectric constant and light transmission. To offer protection on PSCs, the encapsulation material needs to be chemical inert and to have enough mechanic strength with appropriate oxygen transmission rate (OTR) and water vapour transmission rate (WVTR). Besides, glass transition temperature is another important indicator for organic materials.

It is worth mentioning that there is no unified testing standard for stability. Experiments are carried out under different humidity and temperature to simulate the environmental condition, while some of them are implemented in nitrogen box. Illumination is another significant factor influencing the result of stability. Provisions of test specifications is crucial for further research.

2.1. Carrier transport layer

Many organic transport layer was applied so as to acquire high efficiency. For example, a conducting polymer poly(3,4-ethylenedioxythiophene):polystyrenesulfonate(PEDOT:PSS) has typically been used as HTL and [6,6]-phenyl C61 butyric acid methyl ester (PCBM), C60 and their derivatives have been used as ETL. HTL and ETL are adjacent layer of perovskite. It is reported that, to simplify the process, perovskite functions as both light harvester and hole conductor without HTL. Although the perovskite can also act as a role of electron and hole transporter, the presence of them provides efficient charge separation and reduces recombination of photocurrent\cite{14}. Besides, they are the main contributors to resist the penetration of external material which may induce the degradation of perovskite, and the penetration of moisture and oxygen. However, many organic materials are vulnerable to heat and have detrimental effects on the long-term stability of PSCs. Taking PEDOT:PSS as an example, its high acidity may lead to corrosion of the ITO electrode and its high hygroscopicity would absorb water and accelerate the degradation of PSCs\cite{15}. Compared to the organic transport layer, metal oxide layer has better stability, higher carrier mobility and lower cost.

It has been reported that using CuSCN, Cul, NiO, etc. as HTL and TiO2 nanoparticle, ZnO nanoparticle, SnO2, etc as ETL acquires high efficiency and stability of PSCs. TiO2 nanoparticles have long been used for photocatalyst and photovoltaic (PV) technologies. Although further development on the material and process has been made\cite{16}, there is an obvious shortcoming. It has been verified that TiO2 solar cells decay faster than unencapsulated ones under the exposure to sunlight. The instability is attributed to the unfilled oxygen vacancies at the surface of TiO2, which is caused by the
bandgap excitation of TiO$_2^{[17]}$. ZnO exhibits similar phenomenon. Adding surface blocking layer such as Sb$_3$S$_3$ and Al$_2$O$_3$ can reduce the degradation effect by blocking photocatalytic effect and suppressing the recombination of charges. It has been reported that TiO$_2$ layer is replaced by Al$_2$O$_3$ scaffold$^{[18]}$, because thick Al$_2$O$_3$ is insulated. The UV-light induced degradation also happened on perovskite. Then, F Bella, et al. developed a designed fluoropolymeric layer coating on the both sides which downshifted the luminescence and decreased UV portion of the spectrum$^{[19]}$. Up to now, organic layer is modified to enhance stability$^{[20]}$. ZnO and Al$_2$O$_3$ are considered as promising materials.

2.2. Electrode
The electrodes, especially cathode, can affect the performance of PSCs. Typical electrode materials, such as Ag and Al can chemically react with decomposition products of perovskite by direct contact or under humid condition$^{[16]}$. The halide reaction products such as AlI$_3$ and AgI would facilitate the degradation. Besides, with the absence of oxygen, silver contacts may form shunting paths with the TiO$_2$$^{[21]}$. The detrimental effect can be avoided by two ways: changing the material and isolating perovskite from electrodes using designed layers. The cathode can be substituted by Au or carbon-based composite electrode$^{[22]}$. Adding interlayer is another common solution. It has been shown that metal atoms can diffuse into the carrier transport layer during the process$^{[16]}$. An interlayer effectively lessen the possibility of direct contact between perovskite and metal. It has been demonstrated that a Cr$_2$O$_3$-Cr interlayer effectively prevent contact between electrodes and perovskite$^{[23]}$.

2.3. Encapsulation
Encapsulation plays an important role in commercialization of Silicon solar cell. It is a low-cost and efficient approach to prolong life-time of solar cells. Recently, many studies about the encapsulation of PSCs have been carried out. For example, Teflon deposited on device as a hydrophobic passivation layer effectively improves the resistance to moisture$^{[24]}$. However, it is a tough challenge to meet the requirements as stated above. Some materials such as SiO$_x$Cy serving as a barrier for oxygen and moisture, need complex and expensive process. These materials are also not qualified for encapsulation.

Glass-to-glass encapsulation is the simplest way to prevent the moisture and oxygen. For instance SiO$_2$ layer deposited by electron beam inside glass cover layer has a good long-term stability$^{[25]}$. UV-curable epoxy-based encapsulation method with a glass lid has already been used in organic light-emitting devices (OLEDs). Han Y, et al. enhanced the stability under the simulated condition by placing a desiccant inside the glass lid. They found that perovskites still had the risk of degradation under high humidity and temperature condition which can be easily reached by sunlight and rainfall$^{[26]}$. Various encapsulation materials are investigated to enhance the long-term stability such as ethylene vinyl acetate (EVA), ethylene methyl acrylate (EMA), polyisobutylene (PIB), etc. Typically, the structure of glass-polymer-glass prevents ingress of moisture. The polymers and epoxy resin serve as glass adhesive and edge sealant.

This rigid glass encapsulation method restricts its application space. Flexible encapsulation materials and substrates are desirable. Typically, indium doped zinc oxide-coated polyethylene terephthalate (IZO-PET) films$^{[27]}$ and PET$^{[28]}$ are used as flexible substrate. It has been reported that PCE of flexible device has exceeded 10% without encapsulation$^{[29]}$. Single layer encapsulation method is a common encapsulation strategy. For instance, some commercial plastic$^{[27]}$ are used as barrier film. Thin-film encapsulation (TFE) is one of the promising low cost solution adopted from OLEDs technology. This encapsulation is a combination of organic and inorganic multilayer films deposited on devices. Inorganic layer such as SiO$_2$, Si$_3$N$_4$ and Al$_2$O$_3$ functions as a barrier preventing ingress of moisture. Moisture ingress from lateral sides can not be averted by this strategy. Organic layer buffers interface stress ensuring device flexibility. It has been demonstrated that compact Al$_2$O$_3$ layer made by
atomic layer deposition, super hydrophobic FDTS layer made by thermal evaporation and pV3D3 layer made by chemical deposition on the device forming a TFE improved the stability[29].

2.4. Extra layer
Most of the improvements adjust specific parts of the PSCs. Compared to the device structure mentioned above, sometimes extra layers are introduced to enhance the long-term stability. Generally, interlayer should exhibit high light transmission, perfect electrical properties and no reaction with other parts. They typically play a role of suppressing recombination of charges, modifying surface of provskite, preventing ingress of moisture and diffusion of other material. Thus the presence of interlayer can enhance the stability of PSCs. Many types of interlayers have been mentioned in above section such as carrier blocking layer and separation layer isolating perovskite and electrodes. Perovskite may be damaged by sputtering particles with high kinetic energy. Thus, to protect the provskite, some devices contain buffer layers such as ITO, MoO3, porous Al2O3, etc. There are also some interlayers which are not widely used. Jeng, J. Y., et al. used NiOx electrode interlayer on substrate to promote photovoltaic performance[30].

3. Initial efficiency
As described in above section, encapsulation can enhance the long-term stability. Nevertheless, the encapsulation process also lower the PCE. Matteocci, F. et al. has compared four types of sealing materials and their corresponding sealing procedures. It is proved that encapsulated PSCs could have a higher PCE even after 20 hours, though it has a lower initial efficiency. It is also revealed that high curing temperature, UV-light exposure and UV-induced heating have a great impact on initial efficiency. Because the processing condition is related to the sealing material, it is crucial to keep balance between the long-term stability and initial efficiency. For instance, it has been shown that compared with using surlyn thermoplastic gasket as an edge sealant, UV-light curable epoxy cured under optimum UV-light intensity and low temperature retains higher proportion (83%) of its original performance, proving this edge sealant can effectively protect the PSCs from moisture to enhance the long-term stability[31].

The PCE of PSCs is 32% in theory[32] which is higher than the currently reported maximum. Except reducing the impact of material, finding optimum process of PSCs can also increase the initial efficiency of PSCs. Conventionally, perovskite is made using solution-based techniques. In accordance with planar perovskite, however, the PCE of solution-processed device is far below than that of mesoscopic structured device. Several improvements have been made on low-temperature solution-processing such as interdiffusion and thermal annealing, etc. Such one-step deposition techniques typically troubled by unexpected morphological changes caused by dewetting problems, which lead to unstable photovoltaic performance. Poor morphology of perovskite is detrimental to device performance, mainly because it causes electrical shorting and detrimental impacts on charge dissociation, transport and recombination[33]. It has been demonstrated sequential deposition enhances the morphology of perovskite[34]. Besides, the formation of perovskite films can be regulated by incorporating additives in precursor solution. Bidentate halogenated additives such as 1,8-diiodooctane (DIO) improve the solubility of PbCl2 by temporarily chelating with lead ions during crystal growth, which lead to enhanced size as well as homogeneity of perovskite and efficiency of solar cell[33]. To ameliorate the formation processing of polycrystalline perovskite films, adding alkylphosphonic acid ω-ammonium chlorides to perovskite by inducing organic ammonium cations shows higher initial performance and device stability[35]. Besides, to acquire high performance, process was designed through vapour deposition.

Hole transport material(HTM) and electron transport material(ETM) also affect the initial efficiency through carrier transport rate, conductivity and other electrical properties. Organic transport layer is commonly used to acquire high PCE. 2,2',7,7'-tetakis(N,N-p-dimethoxyphenylamino)-9,9'-spirobifluorene(spiro-OMeTAD), poly[bis(4-phenyl)(2,4,6-trimethylphenyl)amine] (PTAA), poly(3-hexylthiophene-2,5-diyl) (P3HT),
etc. are used as HTM and PCBM, C60 and their derivatives, etc. are used as ETL. As mentioned in above section, low-temperature processed metal oxide is promising material used as ETL. To acquire high PCE, improvements have been made to enhance the surface of ETL film. The main reason is that chemisorption of oxygen of materials may causes energetic disorder induced deep trap states\cite{36}. The electrons from perovskite lowest unoccupied molecular orbital (LUMO) hopping between these trap states can lead to trap-mediated recombination\cite{37}. It has been demonstrated that surface of ETL can be modified through metal doping, UV-Ozone treatment, extra organic ETL and nanostructured network on pristine ETL. Improving efficiency and stability at the same time keeping low costs may bring great challenges to the selection of materials. It is believed that the performance will be better after comprehending fully mechanism of PSCs.

4. Lead leakage

Solar energy is regarded as clean energy, however there are still some pollution from photovoltaic technologies such as processing procedures of solar cells and recycling of waste solar cells. Environmental protection is a significant obstacle to the commercialization of PSCs. Recently, many feasible attempts have been made to reduce environmental impacts. For instance, hole transporting material (HTM) typically is processed with solvents which is toxic to human bodies. Lee, J. et al. have reported a polymeric HTM containing benzothiadiazole (BT) and benzodithiophene (BDT). This material is environmentally friendly, because it is highly soluble in an unhararmful solvent and is dopant-free\cite{38}.

Lead pollution is a major concern regarding the metal halide perovskite. The main reason is lead in degraded perovskite has a high solubility in rainfall (Ksp~10^{-8})\cite{39}. Lead leakage is severe pollution which has notoriously deleterious impact on human body, especially organs and soft tissues. Lead poisoning can give rise to anemia, kidney damage and nervous system diseases\cite{40}. Developing Pb-free PSCs as an alternative has been proposed. The most promising Sn-based halide perovskite does not have a fully three-dimension structure. Although the three-dimension hollow structure has been designed\cite{41}, limitations of electrical properties of Sn-based halide perovskite restrict its performance\cite{42}. Unfortunately, Pb-based halide perovskite is irreplaceable at present. Although some measures are helpful to partially relieve the pollution caused by Lead leakage, encapsulation is the simplest and cheapest method to prevent the Lead leakage.

PSCs work outside, occasionally under severe weather. Hail and storm may break the solar cells. Damaged solar cells can cause Lead leakage when encountering rainfall or other adverse weather. To avoid the leakage, one way is to prevent water from entering the damaged perovskite. It has been reported that taking advantage of the self-healing property of epoxy resin, under the high temperature condition exposed to the sun, the Lead leakage of the perovskites can be effectively reduced by preventing water penetration\cite{43}.

Another way is to absorb Lead ions in water. For example, chelation can effectively remove the Lead ions in water. Coating thin film outside the device to collect the leaked Lead ions has been demonstrated. According to the description, methanediphosphonic acid (DMDP) film with proper thickness is coated on the front transparent electrode and pre-dry film which made by blending Pb-chelating agents such as EDTMP into polymer matrix is fabric ated on the back side. Because the DMDP film is transparent, permeability to water, insoluble in water and strongly binding to Lead ions. It can greatly reduce the lead tolerance in water without influence on PCE\cite{44}.

This on-device amelioration has several obvious shortcomings. The adsorbent may be saturated when exposed to external environment, especially rainfall\cite{45}. It leads to invalidation of the thin film. Besides, the thin film could be damaged by UV-light. Materials which are resistant to UV-light and external friction are much expensive and choice of available materials is rather limited. In order to avoid the problem, amelioration can be made inside the solar cells. As described in the above section, ETL and HTL are adjacent layers of PSCs. They take on the role of preventing the invasion of external substances and enhancing the stability. It has been designed utilizing 2D conjugated metal-organic frameworks (MOF) as electron extraction layer (EEL)\cite{46} and alkoxy-polytetraethylene glycol as
HTL\textsuperscript{[47]}. These materials can trap lead ions by dense array of thiol groups on the MOF and adsorb lead ions by chelation respectively. It is worth mentioning that HTL is on the top side of perovskite. Thus the thickness of HTL will hinder absorption of sunlight. For example, 27-nm-thick alkoxy-polytetraethylene glycol layer is incapable of adsorbing the lead ions completely. Increasing the thickness would lessen efficiency of PSCs.

It has been tested that carbon electrode combined with cation-exchange resins achieves the efficacy of reducing the lead without affecting the PCE\textsuperscript{[48]}. Another constructive suggestion is adding a lead adsorbing layer between HTL and electrode. It has been reported that utilizing mesoporous sulfonic acid cation exchange resins as adsorbent layer can effectively prevent the lead leakage and improve the stability at the meanwhile\textsuperscript{[49]}. Many other ideas have been proposed, such as applying a porous MOF polymer composite, known as FeBTC/PDA, in perovskite solar panels\textsuperscript{[50]}.

| Table 1 : List of probable materials for preventing Pb leakage |
|---------------------|----------------------|------------------|-----------------|
| Materials          | Simulation method of rainfall                                      | Heating temperature | Pb concentration         |
|---------------------|--------------------------------------------------------------------|--------------------|----------------------|
| Epoxy resin\textsuperscript{[43]} | Drip acid water (pH=4.2) for 1.5h at a speed of 5ml/h on damaged PSCs | 45°C               | Without Heating <1.8mg/L |
|                     |                                                                    |                    | After Heating <0.1 mg/L |
| DMDP & EDTMP-PEO\textsuperscript{[44]} | PSCs soaked in 40 ml of pure water for over 3h | 50°C               | Without Heating <9ppm  |
|                     |                                                                    |                    | After Heating <12ppm   |
| Metal-organic frameworks (MOF)\textsuperscript{[46]} | soaked in acid water (pH=5.6) |                   | 38.1 ppm              |
| Cation-exchange resins\textsuperscript{[48]} | PSCs soaked in 200 ml of pure water for over 3h | 85°C               | Without Heating <1ppm  |
|                     | Drip water (pH=7) for 1h at a speed of 5ml/h on damaged PSCs        |                    | After Heating <1ppm    |
|                     | Drip acid water (pH=4.2) for 1h at a speed of 5ml/h on damaged PSCs |                   | 1.92ppm               |
| Mesoporous sulfonic acid cation exchange resins\textsuperscript{[49]} | Drip acid water (pH=4.2) for 1h at a speed of 5ml/h on damaged PSCs |                   | 2.55ppm               |
|                     |                                                                    |                   | 11.9ppb               |

5. Conclusion

Whether it is harmful to the perovskite should be concerned when selecting function layer materials and encapsulation materials of PSCs. Replacing HTM with environmentally friendly one is completely feasible. Encapsulation is the main way to enhance the stability of PSCs. Low cost thin-film encapsulation is an important development direction. Besides, to cope with the emerging environment issues, the function layer materials can be modified to adsorb lead, preventing the leakage of lead from PSCs. An ideal and effective way is adding a buffer layer which can adsorb lead and enhance stability at the same time. To integrate the advantages of each part of PSCs, interactions between constituent materials are needed to be studied.

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