CHALLENGING ISSUES IN PRODUCING NEW GENERATION HIGH EFFICIENCY PEROVSKITE SOLAR CELLS

Sebahattin Tüzemen, Ali Baltakesmez

Department of Physics, Faculty of Science, Atatürk University, 25240 Erzurum, Turkey

Abstract

Flexible high efficiency solar cells with an active layer of organometallic perovskite material with $CH_3NH_3^+$ organic cation surrounded by metal halide MX_6 (M=Pb or Sn, X=I, Br or Cl) octahedron structures is extraordinarily promising as efficient light absorbers since their controllable bandgap ranging from 1.5-2.3 eV corresponding to the optimal solar absorption band. Modification of the bandgap is possible with the precise adjustment of halide concentration. Theoretically this allows the material to have very high solar conversion efficiencies of detailed balance or so called the Shockley-Queisser limit of around 31%. Most recently reported efficiencies are around 20%. However reliable and repeatable devices are reported to have around 12-18% efficiencies. The popular challenge is that what makes a proper process, combination and design to have very high solar conversion efficiencies up to 31%. In this study we will introduce a comparative review of present studies in order to find out an optimal conditions to reach higher efficiencies in perovskite solar cells in terms of various aspects such as processing, chemical combination and device modeling. In particular, we introduce a unique spin coating process named dual coating combining the static and dynamic coating processes, giving rise to repeatable and stable power conversion efficiencies of around 15% at a beginning stage in research laboratory conditions.

INTRODUCTION

On top of the some important advantages such as low coast and easy process, due to some unique mechanical, electrical and optical properties of organometallic perovskites, there are tremendous recent attractions for generating new applications, especially in solar technology. Although there are also some other scientific applications in superconductivity [1], magneto-resistivity [2], ferromagnetism and ferroelectricity on the agenda due to their rich physical properties, the main focus is on optoelectronics resulting in devices such as photodiodes, LEDs and solar cells.

Flexible high efficiency solar cells with an active layer of organometallic perovskite material with $CH_3NH_3^+$ organic cation surrounded by metal halide MX_3 (M=Pb or Sn, X=I, Br or Cl) octahedron structures is extraordinarily promising as efficient light absorbers since their controllable direct bandgap ranging from 1.5-2.3 eV corresponding to the optimal solar absorption band [3]. Modification of the bandgap is possible with the precise adjustment of metal halide stoichiometry and/or with different combinations of metal and/or halide components. Theoretically this allows the material to have very high solar conversion efficiencies (SCE) of detailed balance or so called the Shocley-Queisser limit of around 31%. Most recently reported SCE observed from perovskite solar cells are around 22% or even more [4]. However reliable and repeatable devices are reported to have around 12-18% efficiencies. The popular challenge is that what makes a proper process, combination and design to have very high solar conversion efficiencies up to 31%. Even more efficiencies are possible with some assisting applications such as multi-junction cells, light concentration, quantum dots and photon upconversion upon regular device fabrication. For instance, photon upconversion can often take place in containing ions of d-block materials such as Ln, Ti and Ni, and Ti seems to be the best candidate to alloy with the M component of the perovskite material [5].

IMPORTANT ISSUES

There exist two important parameters; the PCE and the energy conversion rate of photon to electricity, exhibiting the motivation and significance of perovskite solar cells. While commercial and conventional state of the art organic and inorganic semiconductor solar cells reach to less than 20% of PCE in more than quarter century research, investigations on organometallic perovskites exceeds the same amount of efficiency in less than a decade of time. On the other hand, the conversion process from light to photo-current is as low as around 28% in organic solar cells, and as a competing material perovskites has nearly 60% of light-to-current conversion which is nearly as good as most commonly used conventional and commercial semiconductor solar cells such as GaAs, Si and GaInP which covers the vast amount of industry.

Several deposition methods of perovskites materials onto the conductive substrates such as ITO [6] and AZO [7,8] are possible using one and two step spin coating, and recently introduced dual one step spin coating by our group [9]. Various device design alternatives such as planar mesostructured cells have been proposed by many groups. Thin films using spin coating can be produced around 1 μm thickness. Diffusion length of carriers in perovskite materials is >1 μm [10]. Electronic mobility is between 10 cm^2/Vs and 100 cm^2/Vs which allows an appropriate photo-current transportation. Electrons and holes are free excitons rather than bound excitons since the exciton binding energy is low. Therefore, photo-current can be transported from the active layer without any difficulty.

In general argumentations of solar cells, considering all the loss factors such as recombination losses, spectrum losses and impedance losses, the probability of one photon to produce one electron-hole pair, the PCE can only be $\eta \leq 0.31$. While these factors can all considered to be extrinsic, there are also some other intrinsic factors that can reduce the power conversion efficiency (PCE): these are limited mobility, non-radiative recombination and Auger recombination. For instance the last has been an important factor in also LEDs as shown by Avrutin et al [11].

Although there appear to have high efficiency devices from perovskites deposited by various techniques which mostly utilizes single solution based spin coating, there are still remaining following problems in such devices;

- Stability
- Hysteresis

Stability is mainly affected by the high water solubility of the organic compound and injected holes from the oxide layers due to the UV component of the sun light. For the effect of moistures and wet air conditions, various encapsulation techniques have been introduced such as encapsulation by carbon nanotubes and polimer composites [12].

On the other hand it is reported by Bella et al that photopolymer coating of the perovskite devices reduces the effect of both water and UV due to the reduced water diffusion and the conversion of UV to visible, resulting in highly efficient and stable solar cells [13].

However hysteresis still remains as a big issue in all these state of the art devices, mainly governed by ion mobility, polarization, ferroelectric effects and occupation of the trap centers mostly occurred in the interfaces. Therefore the improvement of the interface quality with some novel attempts is necessary to resolve these kinds of important matters.

In this work, in order to overcome these crucial problems and improve the PCE, we have comparatively studied the film properties and the device performance of the perovskite thin films prepared by static and dynamic based SSC methods and the combination of the two methods without any extra step. The results show that high quality perovskite films can be achieved by the application of a modified SSC method using the combined processes. We named this approach as dual-SSC (D-SSC) approach. The perovskite films prepared by the D-SSC approach have high surface coverage, smooth surface, and very low pinhole density, together with improved device performances and enhanced stability.

METHODS AND SOLUTIONS

Although, conventional Si solar cells needs to have high temperature (>1000 ºC), expensive and practically difficult processes, perovskite solar cells can be deposited using cheap, easy and environmentally friendly wet chemical processes at low temperatures (<150 ºC) in laboratory conditions. Process can easily be performed by the solution in one or double step using coating system. PbI_2, PbF_2 or $PbCl_2$ can be mixed up with methyl ammonium halide by various coating methods such as one step spin coating [14], two-step spin coating [15] and solution vapor-assisted [16] as shown in Fig. 1.

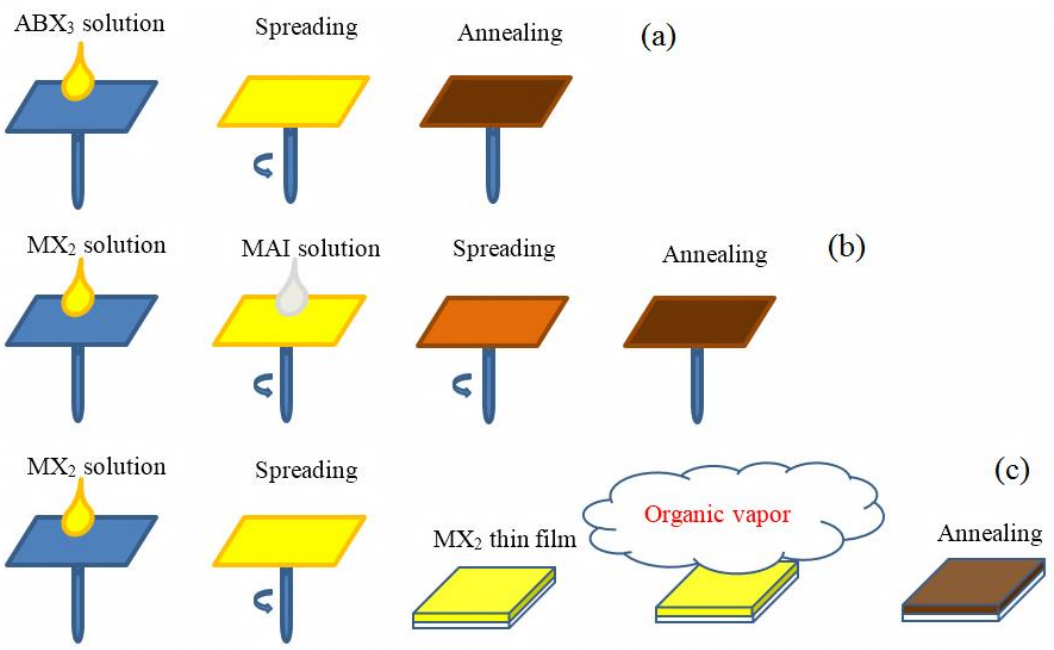

Fig. 1. Three general solution based methods for preparing perovskite thin film.

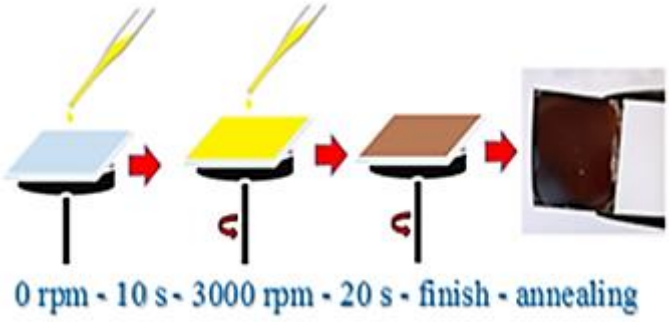

Fig. 2. Dual single solution coating process.

Finally, there are also other possible utilizations in processing in order to enhance the efficiencies. For this, we introduce a unique dual-single solution coating process (D-SSC) [9]. This unique process is illustrated in Fig. 2. It shows low pin hole density and more homogeneity in D-SSC samples. From the I-V curves in Fig.3, it was shown that nearly 50% increase is possible with the unique D-SSC process. The films coated by D-SSC method is formed as layer to layer onto the first crystal grains or templates, remaining high stability and low hysteresis. Hence, it is caused that not only an improved performance of the perovskite solar cell with the PCE increased to around 15%, but also enhanced stability of the

films under ambient conditions with filled pin-holes and grain boundaries of the films and uncoated surface of the substrate. It must be noted that these moderate efficiencies are obtained with single cells under un-concentrated light in our laboratory conditions with limited furnishing. Utilization of the same processes introduced here may result in very high efficiencies around 50% in an industrially sophisticated laboratory, using the discussed applications that can eliminate PCE reducing factors in this limited review.

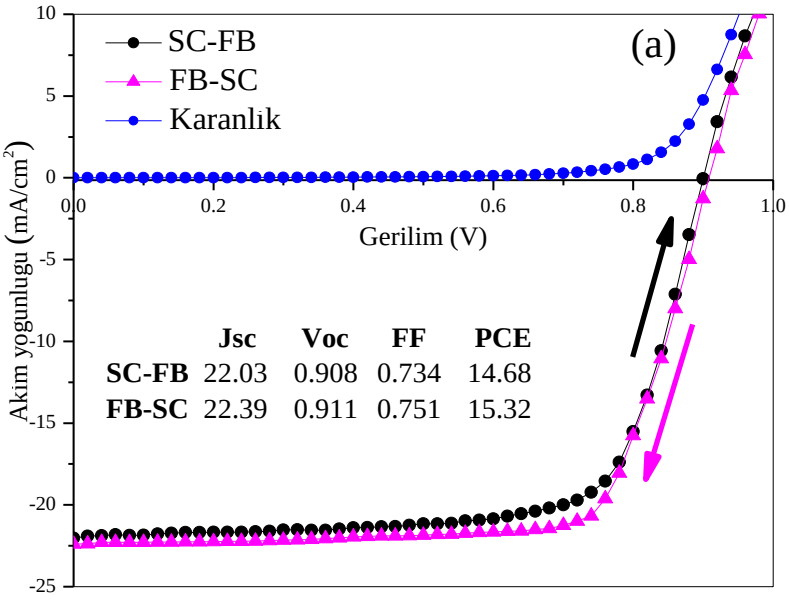

Fig. 3. J-V curves of perovskite solar cells prepared by D-SSC approach.

CONCLUSIONS

We introduced a useful coating approach named dual-single solution coating (D-SSC), resulting in reproducible high morphological, structural and optical quality and void-free perovskite films with a few pinholes, which are comparable with high quality perovskite films prepared by vacuum and enhanced solution processes. Besides, the coating approach could be used for coating other films for the improvement of the quality in optoelectronic device fabrication. In the dual coating approach, the static stage plays an important role as a designed under layer surface such as surface energy and nucleation layer, leading to high quality perovskite thin films coated by the dynamic SSC approach. Thus, the films prepared by the dual coating approach with hot deposition show better properties in all the characterization. The SEM measurements show that perovskite films prepared by the dual coating approach have a good morphology in comparison to the static and dynamic coated ones. The film crystallinity was improved by dual coating with hot deposition, overcoming the problems of stability and hysteresis.

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