

Stability and Performance Enhancement of Lead Iodide Perovskite Solar Cells using Salt Additives

Chih-Wei Chu, and Karunas Karan

Research Center for Applied Science, Academia Sinica, Taipei 115, Taiwan

E-mail: gchu@gate.sinica.edu.tw

Abstract

The main issues in planar perovskite solar cells are the coverage and crystallinity of the perovskite film on the PEDOT:PSS layer. In order to improve these features, we introduced alkali metal halides (salts) as additives in the perovskite precursor solution used in a two-step preparation method. These alkali metal halides chelate with Pb^{2+} ions and enhance the crystal growth of PbI_2 films. These salts promote homogeneous nucleation and larger crystallite sizes, thereby enhancing the morphology and crystallinity of the perovskite films. The alkali metal halides recrystallize the small grains and passivate the grain boundaries and interface states, allowing effective charge generation and dissociation in perovskite films. The power conversion efficiency of the device incorporating a small amount of a salt additive increased by approximately 33%—from 11.4 to 15.08%. This device was more stable than a corresponding device prepared without the additive.

1. Introduction

Organometal halide perovskites are emerging materials in photovoltaics (PVs) because of their high absorption coefficients, long carrier life times, micrometer diffusion lengths [1,2]. Indeed, an exceptionally high solar cell performance of 20.2% has been achieved when using a solution-processed lead halide perovskite. High temperature annealing is required to make crystalline TiO_2 layers in mesoporous-type solar cells, whereas planar heterojunction-based solar cells can be fabricated at low temperature—more suitable for roll-to-roll manufacturing of large-area, flexible solar cells [3]. Unlike mesoporous-type devices, in which the perovskite material can infiltrate within the porous matrices, perovskite films with pinholes and non-uniform coverage are usually present in planar-type perovskite solar cells; these factors are mainly responsible for their poor device performance.

Controlling the morphology and crystallization of perovskite thin films in planar architectures is a main challenge affecting our ability to develop high-performance devices. Incorporation of additives in the perovskite solution process has become a simple way to improve the film coverage and crystallinity. Several additives, including NH_4Cl , 1,8-diiodooctane, 1-chloronaphthalene, polyvinylpyrrolidone, hydroiodic acid (HI), and

hydrochloric acid, can be used to form smooth, continuous, and uniform films with flawless perovskite nanocrystals, significantly improving device performance [4-8].

2. Results and discussion

We investigated the influence of alkali metal halides as additives on the performance of perovskite solar cells.

2.1 Crystallinity and morphology analysis

The X-ray diffraction (XRD) intensities of the perovskite peaks were enhanced significantly when using the alkali metal halides as additives, suggesting the formation of highly crystalline perovskite films. Scanning electron microscopy (SEM) shows the crystallinity and crystallite size of the perovskite film prepared with additives were greater than those of the films prepared without additives. The film prepared additive featured more uniform and dense nanocrystals of large grain size than without additives.

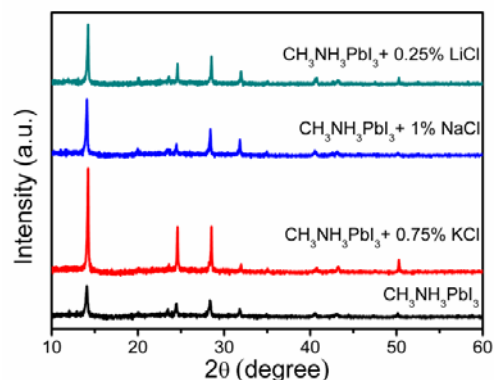


Figure 1 XRD patterns of perovskite thin films prepared with and without KCl, NaCl, and LiCl as salt additives.

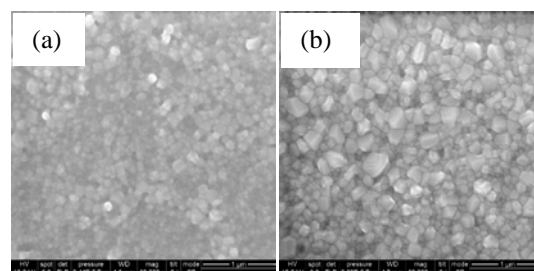


Figure 2 SEM surface images of perovskite film (a) without salt additive (b) with additive

2.2 Device performance

We fabricated devices having the architecture glass/ITO/PEDOT:PSS/perovskite/PC₆₁BM/C₆₀/B CP/Al and investigated their PV performance. The device prepared with 0.75% KCl achieved the highest PCE (15.08%), with a short-circuit current density (J_{sc}) of 19.42 mA cm⁻², an open-circuit voltage (V_{oc}) of 1.04 V, and a fill factor (FF) of 74.67%. The highly crystalline, large-crystal-size perovskite formed in the presence of alkali metal halide additives resulted in power conversion efficiency of 15.08% for a resulting planar heterojunction perovskite solar cells.

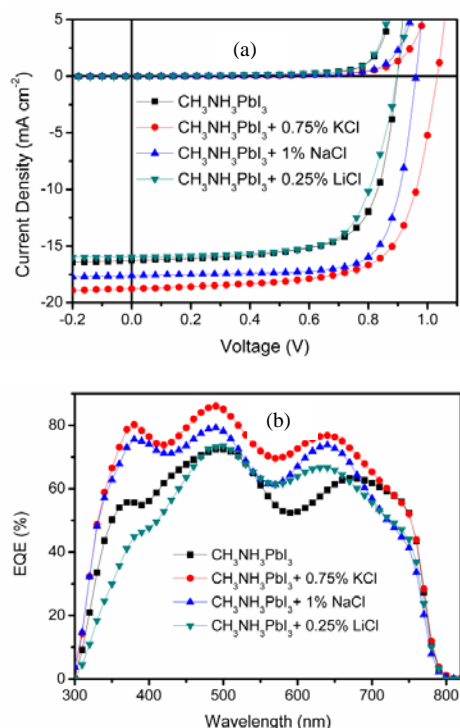


Figure 3 (a) J - V characteristic (b) EQE spectra of perovskite solar devices prepared using various salt additives.

EQE spectra of the perovskites formed with and without the various additives at their optimized concentrations. The additive-containing devices displayed broader and improved EQEs from 350 to 800 nm, relative to that prepared without additives. The EQE of the top-performing KCl-based device reached over 85%, whereas that of the device prepared without additives reached only 70%.

2.3 Stability

We investigated the stability of our perovskite devices prepared with and without KCl as an additive. The stability of the device prepared with the KCl additive was better than that of the device

prepared without the additive, due to the formation of more compact film with larger crystallites. The KCl-added perovskite device exhibited long-term stability: only 16.5% degradation over 50 days under stored in dark environment.

Table 1 Photovoltaic performance parameters of devices prepared with various salt additives.

Salt	V_{oc} (V)	J_{sc} (mA cm ⁻²)	FF (%)	PCE (%)
0.75% KCl	1.04	19.42	74.67	15.08 ^a (14.12) ^b
1% NaCl	0.96	17.59	75.62	12.77 ^a (12.14) ^b
0.25% LiCl	0.91	15.97	68.67	9.98 ^a (9.35) ^b
Without salt	0.90	16.60	76.31	11.40 ^a (10.86) ^b

3. Summary

A significant enhancement (ca. 33%) in PCE, from 11.40 to 15.08%, occurs after incorporating a small amount of KCl as an additive in a planar heterojunction perovskite solar cell. The additives improved the crystallinity and morphology of the perovskite films and, thereby, enhanced their absorption. This approach using rational additives opens up a new path for enhancing the performance and stability of perovskite solar cells.

Acknowledgments

Dr. Chu thanks the Ministry of Science and Technology (MOST) of Taiwan (104-2221-E-001-014-MY3 and 104-2221-E-009-096-MY3) and the Career Development Award of Academia Sinica, Taiwan (103-CDA-M01), for financial support.

References

1. A. Kojima, K. Teshima, Y. Shirai, T. Miyasaka, J. Am. Chem. Soc., 2009, 131, 6050.
2. M.M. Lee, J. Teuscher, T. Miyasaka, T.N. Murakami, H.J. Snaith, Science, 2012, 338, 643.
3. J. You, Z. Hong, Y. Yang, Q. Chen, M. Cai, T.-B. Song, C.-C. Chen, S. Lu, Y. Liu, H. Zhou, Y. Yang, ACS Nano, 2014, 8, 1674.
4. P.-W. Liang, C.-Y. Liao, C.-C. Chueh, F. Zuo, S.T. Williams, X.-K. Xin, J. Lin, A.K.Y. Jen, Adv. Mater., 2014, 26, 3748.
5. H. Choi, J. Jeong, H.-B. Kim, S. Kim, B. Walker, G.-H. Kim, J.Y. Kim, Nano Energy, 2014, 7, 80.
6. L. Yang, J. Wang, W.W.-F. Leung, ACS Appl. Mater. Interfaces, 2015, 7, 14614.
7. C. Zuo, L. Ding, Nanoscale, 2014, 6, 9935.
8. C.-C. Chueh, C.-Y. Liao, F. Zuo, S.T. Williams, P.-W. Liang, A.K.Y. Jen, J. Mater. Chem. A, 2015, 3, 9058.