# **Up-Scaling Strategies of Perovskite Solar Cells and Modules**

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### Abstract

On the basis of concerted research efforts worldwide, there is no doubt that outstanding power conversion efficiency (PCE) can be achieved in perovskite solar cells. However, to move forward this technology towards commercialization, developments of up-scaling processes with high PCE and stability is important. At OIST, a team of researchers in the Energy Materials and Surface Sciences Unit has been making concerted efforts to develop processes aiming at high PCE, high-throughput, and minimum batch-to-batch variation, and compatible with large-area perovskite solar cells and modules. In this talk, we will present our recent progress using chemical vapor deposition and spray coating to fabricate perovskite solar cells and modules. Also, we will introduce a novel methylamine gas-induced crystallization process, which provides insights into the formation of perovskite films.

#### 1. Introduction

Organic-inorganic hybrid perovskites (e.g. MAPbI<sub>3</sub>, FAPbI<sub>3</sub>, etc; where MA = CH<sub>3</sub>NH<sub>3</sub><sup>+</sup>; FA = CH(NH<sub>2</sub>)<sub>2</sub><sup>+</sup>) with mixed compositions including Cs have been intensively studied for solar cell applications [1, 2]. Based on published works, there is no doubt that exceptional solar-to-electricity power conversion efficiencies (PCEs) can be achieved with perovskites. However, the current state-of-the-art perovskite solar cells have relatively small active areas (<0.3 cm<sup>2</sup>). Development of up-scaling processes with high solar energy PCEs and stability is important for moving forward this technology towards commercialization [2-6]. At OIST, a team of researchers in the Energy Materials and Surface Sciences Unit has been making concerted efforts to develop processes aiming at high PCE, high-throughput, and minimum batchto-batch variation, and compatible with large-area perovskite solar cells and modules.

In this talk, we will present our progress to use hybrid chemical vapor deposition (H-CVD) [7-11] and spray coating [12, 13] to fabricate perovskite solar cells and modules. In particular, CVD is a technology widely employed in many industrial applications demonstrating potential for scale up. In addition to high throughput sample processing, the CVDbased batch processing improves sample-to-sample uniformity. Our H-CVD process allows fabrication of MAPbI<sub>3</sub> and FAPbI<sub>3</sub> based solar cells with PCEs up to 15.6% (MAI,  $0.09 \text{ cm}^2$ ) and  $5 \times 5 \text{ cm}^2$  modules with 9.5% (FAI, 5-cell modules, total active area 8.8 cm<sup>2</sup>) and 9.0% (FAI, 6-cell modules, total active area 12 cm<sup>2</sup>). We have also investigated the scaling issues by fabricating modules using an established MAPbI<sub>3</sub> solution process, and demonstrated maximum PCEs of 18.3% (MAI, ~0.1 cm<sup>2</sup>), 14.6% (MAI, ~1 cm<sup>2</sup> single cells), and 8.5% at  $5 \times 5 \text{ cm}^2$  (MAI, 6-cell module, total active area 15.4 cm<sup>2</sup>) [8]. In addition, a method to fabricate Cs<sub>x</sub>FA<sub>1-x</sub>PbI<sub>3</sub> by performing Cs cation exchange on H-CVD grown FAPbI<sub>3</sub> with the Cs<sup>+</sup> ratio adjustable from 0 to 24% will be reported. The champion perovskite module based on Cs<sub>0.07</sub>FA<sub>0.93</sub>PbI<sub>3</sub> with an active area of 12.0 cm<sup>2</sup> shows a module PCE of 14.6% and PCE loss/area of 0.17% cm<sup>-2</sup>, demonstrating the significant advantage of this method toward scaling-up [11].

Also, we will introduce a novel methylamine (CH<sub>3</sub>NH<sub>2</sub>) gas induced crystallization process [14-16], which provides valuable insights into the formation of perovskite films. We described for the first time the formation of stoichiometric perovskite in ambient air by exposing PbI<sub>2</sub> films to a simple CH<sub>3</sub>NH<sub>2</sub> gas precursor (as opposed to CH<sub>3</sub>NH<sub>3</sub>I solid powders). This non-stoichiometric reaction produces Pb oxides as by-products, which can be reconverted by further HI gas exposure. With combined measurements of the X-ray diffraction, X-ray photoelectron spectroscopy, and UV-vis properties, we elucidate the chemical reaction mechanisms underlying these gas-induced processes [15]. In addition, we demonstrated that a post-annealing treatment using methylamine greatly reduces impurities at the perovskite grain boundaries and promotes continuity between adjacent grains [16]. Our fabricated solar cell devices based on methylamine gas induced crystallization process and methylamine post-annealing treatment led to efficiencies of 15.3% [15] and 18.4% [16], respectively. Finally, we demonstrated that such a gas-induced process enables preparation of high-quality perovskite films with large areas and high stability [15, 16].

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## References

- L.K. Ono, E. J. Juárez-Pérez and Y.B. Qi\*, ACS Appl. Mater. Interfaces 9 (2017) 30197.
- [2] L.K. Ono and Y.B. Qi\*, J. Phys. D. Appl. Phys. 51 (2018) 093001.
- [3] L. Qiu, L.K. Ono, and Y.B. Qi\*, Mater. Today Energy 7 (2018) 169.
- [4] L.K. Ono, T. Kim, Y. Jiang, Y.B. Qi\* and S.F. Liu, ACS Energy Lett. 3 (2018) 1898.
- [5] L.K. Ono, N.-G. Park, K. Zhu, W. Huang, Y.B. Qi\*, ACS Energy Lett. 2 (2017) 1749.
- [6] M. Remeika and Y.B. Qi\*, J. Energy Chem. 27 (2018) 1101.
- [7] L.K. Ono, M.R. Leyden, S. Wang, and Y.B. Qi\*, J. Mater. Chem. A 4 (2016) 6693.
- [8] M.R. Leyden, Y. Jiang, and Y.B. Qi\*, J. Mater. Chem. A 4 (2016) 13125.
- [9] M.R. Leyden, M.V. Lee, S.R. Raga, and Y.B. Qi\*, J. Mater. Chem. A 3 (2015) 16097.
- [10] M.R. Leyden, L.K. Ono, S.R. Raga, Y. Kato, S.H. Wang, and Y.B. Qi\*, J. Mater. Chem. A 2 (2014) 18742.
- [11] Y. Jiang, M.R. Leyden, L. Qiu, S. Wang, L.K. Ono, Z. Wu, E.J. Juarez-Perez, and Y.B. Qi\*, Adv. Func. Mater. 27 (2018) 1703835.
- [12] M. Remeika, S.R. Raga, S. Zhang and Y.B. Qi\*, J. Mater. Chem. A 5 (2017) 5709.
- [13] M. Remeika, L.K. Ono, M. Maeda, Z. Hu and Y.B. Qi\*, Org. Electron. 54 (2017) 72.
- [14] S.R. Raga, Y. Jiang, L.K. Ono and Y.B. Qi\*, Energy Technol. 5 (2017) 1750.
- [15] S.R. Raga, L.K. Ono, and Y.B. Qi\*, J. Mater. Chem. A 4 (2016) 2494.
- [16] Y. Jiang, E.J. Juarez-Perez, Q. Ge, S. Wang, M.R. Leyden, L.K. Ono, S.R. Raga, J. Hu, and Y.B. Qi\*, Mater. Horiz. 3 (2016) 548.