Charge carrier management for developing high-efficiency perovskite solar cells

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MIT and KRICT scientists recently reported the highest certified power conversion efficiency of 25.2% for perovskite solar cells. They highlighted that charge carrier management is essential to improve the photovoltaic parameters and overcome the theoretical efficiency limit.

In recent years, organic-inorganic perovskite materials have shown great promise for usage as solar cells because of their excellent optoelectronic properties such as strong light absorption, bandgap tunability, high carrier mobility, and simple solution-processed deposition. Pioneering reports on perovskite solar cells (PSCs) include a liquid-state electrolyte in 2009 by Kojima et al.,[1] all-solid-state in 2012 by Kim et al.,[2] and Lee et al.[3] with a meso-superstructure architecture also in 2012 with power conversion efficiency (PCE) of 3.8%, 9.7%, and 10.9%, respectively. PSCs have been regarded as one of the promising next-generation photovoltaic (PV) technologies having reached a certified 25.2% efficiency by developing composition engineering,[4,5] fabrication protocols,[6] and interfacial passivation.[7,8] The remarkable efficiency rise has put PSCs into a prominent position among emerging PVs such as organic photovoltaics (OPVs), quantum dot solar cells (QDSCs), and dye-sensitized solar cells (DSCs) (see Figure 1A). Nevertheless, PSCs lag behind well-established silicon PV technology, which holds a 27.6% record efficiency after many decades of optimization. However, considering the theoretical PCE Shockley–Queisser limit of 31%, there is still room for improvement of the efficiency of PSCs from the current stage so it can reach, for example, the most efficient gallium arsenide (GaAs) record at 28.8%. Charge carrier recombination reduces photovoltaic parameters such as fill factor (FF) and open-circuit voltage (V_{OC}), which is one obstacle for further improvement. Therefore, efficient charge carrier management facilitates approaching high PCEs in PSCs.

What is the origin of the charge carrier recombination in PSCs? The defect states at the grain boundaries (GBs) and interfaces of perovskite thin films with electron transport layer (ETL) or hole transport layer (HTL) have been highlighted as the main reason for charge carrier recombination. Basically, defects in perovskite materials are shallow level because of their low formation energy, making PSCs defect tolerant. However, due to the ionic nature of perovskite materials, the ions can easily migrate from the defect points during the device performing condition, increasing the defect densities and charge recombination. Therefore, developing passivation methods for better charge carrier recombination management is critical to reaching high-performance values.

In the recent paper published by Yoo et al.,[9] researchers from MIT and KRICT have reported a certified PCE of 25.2% in PSCs (Figure 1A) by an efficient charge carrier management strategy. Significantly, this is one of the most efficient PSCs to date, using a planar architecture, bringing the field one step closer toward the theoretical limit and commercialization of perovskite PV technology. This report emphasizes that simultaneous passivating ETL and perovskite thin film are essential to minimize the charge recombination, improving photovoltaic parameters such as FF and V_{OC} in PSCs.

First, they developed a chemical-bath-processed tin dioxide (SnO_2) layer as an ideal ETL with excellent optoelectronic properties. By optimizing the bath deposition parameters, a pinhole-free SnO_2 layer with beneficial band alignment and low defect density covered the FTO substrate to minimize charge carrier recombination at the ETL/perovskite interface. Figure 1B shows the link between the chemical reaction and surface coverage during the SnO_2 processing with different stages. The PCEs at every stage are presented in Figure 1C. The SnO_2 layer deposited by stage A-ii with a moderated pH of 1.5 showed a high-quality, densely packed morphology with thicknesses from 50 to 100 nm and a highest average efficiency of 24% due to reduced recombination and improved charge extraction at the perovskite interface. However, deposited SnO_2 layers with other stages, A-i, A-iii, and B, were not appropriate to achieve high-efficiency solar cells because of inappropriate coverage, excessive defect states, and unsuitable chemical composition.

Second, besides ETL optimization, they demonstrated that further efficiency enhancement could be obtained by improving perovskite optoelectronic properties. For this purpose, the trace amount of methylammonium lead bromide ($MAPbBr_3$) was added to the formamidinium (FA)-based perovskite thin films, resulting in increased perovskite grains sizes and thin film passivation. As shown in Figure 1D, the

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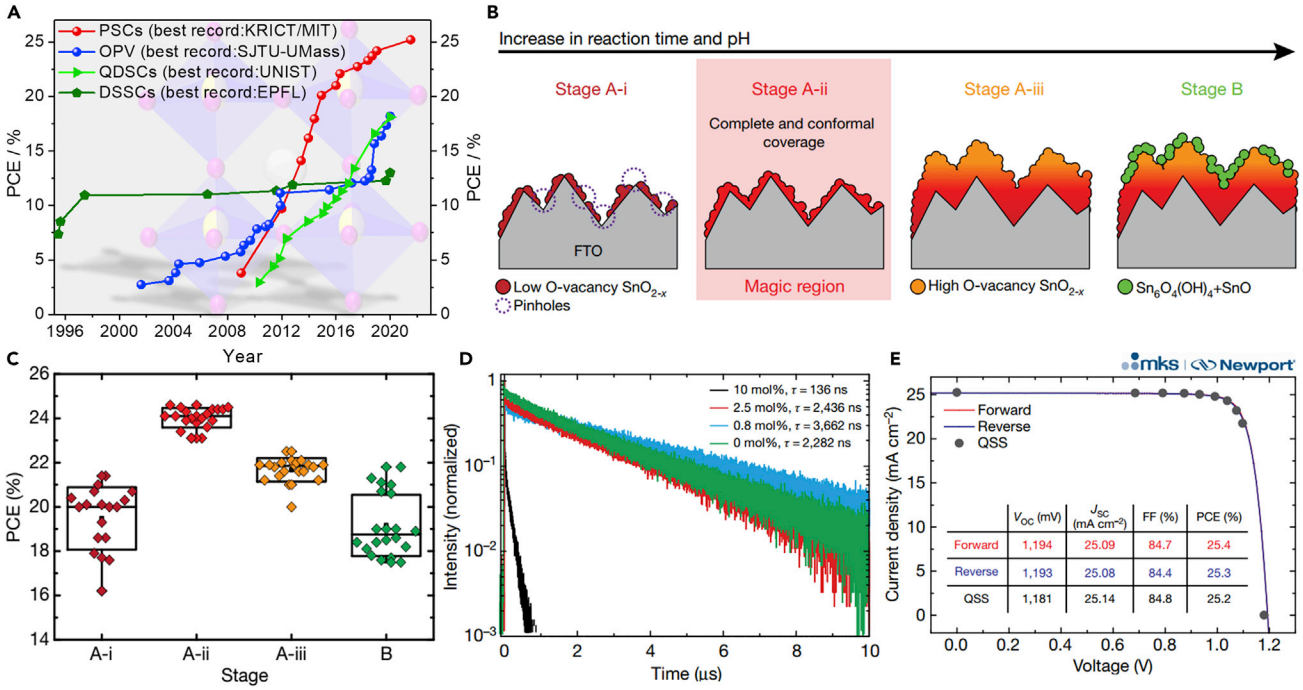

Figure 1. The PSCs performance with various SnO_2 ETLs

(A) Efficiency chart of emerging PV technologies. (B) Schematic illustration of coverage, morphology, and chemical composition of SnO_2 layers. (C) Efficiency statistics fabricated SnO_2-based PSCs with various synthesis stages of SnO_2 layer. (D) Time-resolved photoluminescence of perovskite thin films with different $MAPbBr_3$ content. (E) Certified J-V curve of the champion device fabricated with optimized ETL and passivated perovskite thin film. Adopted with permission from J. J. Yoo et al.[9] Copyright 2021, Nature Publishing Group.

perovskite thin film with 0.8 mol% $MAPbBr_3$ showed the longest carrier lifetime (>3.6 μs) among other concentrations (0%, 2.5%, and 10%), suggesting the lowest recombination due to successful defect passivation.

Finally, as shown in Figure 1E, combining optimized stage A-ii SnO_2 ETL and contained 0.8 mol% $MAPbBr_3$ perovskite thin films leads to a certified quasi-steady-state PCE of 25.2% (mainly due to improved V_{OC} and FF), which is comparable to the theoretical efficiency limit.[10]

More research is needed to better manage the charge recombination in PSCs by defect passivation in different device regions. To reach this goal, better film and device characterization for a comprehensive understanding of the source of defects in PSCs is essential. Additionally, passivation methods from other thin film technologies, such as organic electronics, CIGS, a-Si, and CdTe, may also be suited for PSCs and could be promising avenue in the future.

1. Kojima, A., Teshima, K., Shirai, Y., and Miyasaka, T. (2009). Organometal halide perovskites as visible-light sensitizers for photovoltaic cells. J. Am. Chem. Soc. *131*, 6050–6051.

2. Kim, H.-S., Lee, C.-R., Im, J.-H., Lee, K.-B., Moehl, T., Marchioro, A., Moon, S.-J., Humphry-Baker, R., Yum, J.-H., Moser, J.E., et al. (2012). Lead iodide perovskite sensitized all-solid-state submicron thin film mesoscopic solar cell with efficiency exceeding 9%. Sci. Rep. *2*, 591.

3. Lee, M.M., Teuscher, J., Miyasaka, T., Murakami, T.N., and Snaith, H.J. (2012). Efficient hybrid solar cells based on meso-superstructured organometal halide perovskites. Science *338*, 643–647.

4. Saliba, M., Matsui, T., Seo, J.-Y., Domanski, K., Correa-Baena, J.-P., Nazeeruddin, M.K., Zakeeruddin, S.M., Tress, W., Abate, A., Hagfeldt, A., and Grätzel, M. (2016). Cesium-containing triple cation perovskite solar cells: improved stability, reproducibility and high efficiency. Energy Environ. Sci. *9*, 1989–1997.

5. Saliba, M., Matsui, T., Domanski, K., Seo, J.-Y., Ummadisingu, A., Zakeeruddin, S.M., Correa-Baena, J.-P., Tress, W.R., Abate, A., Hagfeldt, A., et al. (2016). Incorporation of rubidium cations into perovskite solar cells improves photovoltaic performance. Science *354*, 206–209.

6. Wu, C., Wang, K., Li, J., Liang, Z., Li, J., Li, W., Zhao, L., Chi, B., and Wang, S. (2021). Volatile solution: the way toward scalable fabrication of perovskite solar cells? Matter *4*, 775–793.

7. Polavarapu, L., Ye, J., Malekshahi Byranvand, M., Otero Martinez, C., Hoye, R.L.Z., and Saliba, M. (2021). Defect Passivation in Lead-Halide Perovskite Nanocrystals and Thin Films: Toward Efficient LEDs and Solar cells. Angew. Chem. https://doi.org/10.1002/ange.202102360.

8. Malekshahi Byranvand, M., Behboodi-Sadabad, F., Alrhman Eliwi, A., Trouillet, V., Welle, A., Ternes, S., et al. (2020). Chemical vapor deposited polymer layer for efficient passivation of planar perovskite solar cells. J. Mater. Chem. A *8*, 20122–20132.

9. Yoo, J.J., Seo, G., Chua, M.R., Park, T.G., Lu, Y., Rotermund, F., Kim, Y.-K., Moon, C.S., Jeon, N.J., Correa-Baena, J.-P., et al. (2021). Efficient perovskite solar cells via improved carrier management. Nature *590*, 587–593.

10. Guillemoles, J.-F., Kirchartz, T., Cahen, D., and Rau, U. (2019). Guide for the perplexed to the Shockley–Queisser model for solar cells. Nat. Photonics *13*, 501–505.