

Towards Highly Efficient and Low-Cost Quantum Dot Solar Cells

Mengmeng Hao¹, Yang Bai¹, Lianzhou Wang¹

¹ Nanomaterials Centre, Australian Institute for Bioengineering and Nanotechnology and School of Chemical Engineering, The University of Queensland, St Lucia, QLD 4072, Australia

Colloidal quantum dots (QDs) with great features such as facile tunable optoelectronic properties, low synthesis cost and printable in large-scale on variety of substrates, are promising candidates as light absorber for low-cost, highly efficient, and lightweight solar cell devices, which is beyond the capability of traditional Si solar cell.

Perovskite QDs that possess a defect-tolerant nature have shown great potential over traditional chalcogenide QDs when incorporated into PV devices. CsPbI₃ QD solar cells (QDSCs) have delivered a larger open-circuit voltage (V_{OC}) than thin-film counterparts yielding a certified power conversion efficiency (PCE) of 13.4%, which breaks the record efficiency of PbS QDSCs. However, both CsPbI₃ and FAPbI₃ QDs are metastable, which makes them highly susceptible to moisture and polar solvents during post-synthesis purification and device fabrication process. This poses significant challenges for building highly efficient and stable QDSC devices. To address the above obstacles, we developed a ligand-assisted cation exchange technology to synthesize mixed-cation Cs_{1-x}FA_xPbI₃ QDs which are more desirable than pure CsPbI₃ and FAPbI₃ QDs in terms of stability and charge transport properties. We found that the suitable amount of oleic acid (OA) is crucial during the cation exchange reaction and played a key role to obtain QD products with improved stability and reduced defects. The reaction and surface passivation mechanism in QDs was comprehensively studied by monitoring the photoluminescence property, DFT calculation and Transmission electron microscopy.

With optimized device structure design and surface ligand management about QDs, high efficiency and stable solar cell devices were developed, with a champion device showing PCE of 16.6% and breaking the efficiency world record in QDSC category.

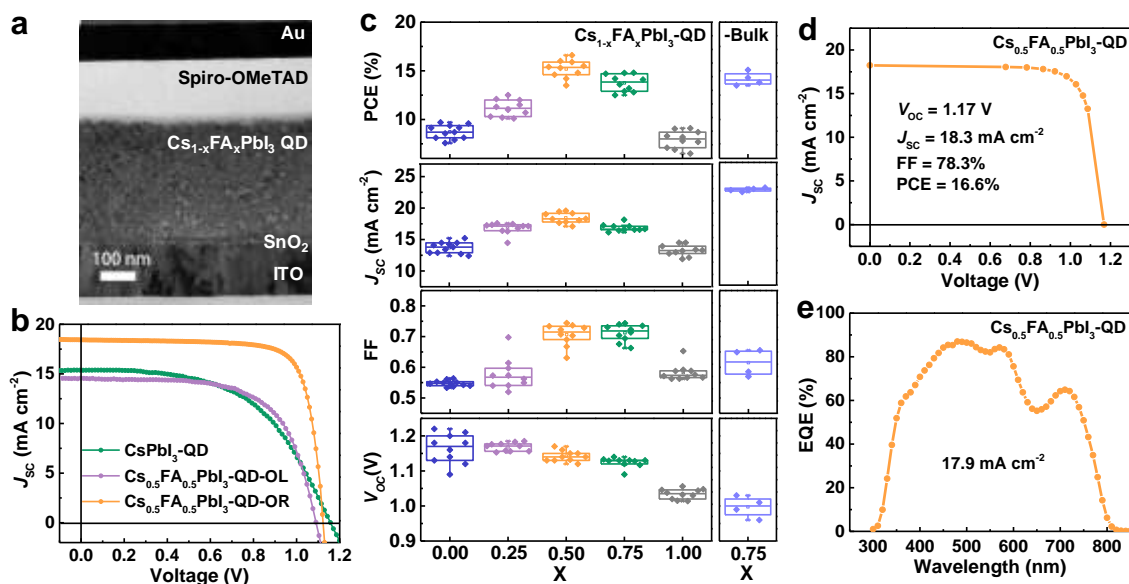


Figure 1. Photovoltaic performance of QD devices. (a) Cross-section TEM image of the Cs_{1-x}FA_xPbI₃ QDSC. (b) Typical J-V curves of QDSCs using different QDs. (c) Performance evolution (PCE, J_{SC} , FF, and V_{OC}) of solar cells based on Cs_{1-x}FA_xPbI₃ QDs of different

compositions and bulk $\text{Cs}_{0.25}\text{FA}_{0.75}\text{PbI}_3$. (d) Certificated J-V curve and (e) EQE spectrum of the hero device measured at Newport PV Lab, USA, the integrated J_{sc} is indicated.

The QDSCs exhibit enhanced V_{OC} and operational stability than devices based on bulk thin film perovskite with same chemical composition. The improved V_{OC} is attribute to the suppressed non-radiative recombination in the QD thin film, which could be originated from the passivation effect of surface ligands and charge spatial confinement. The suppressed ion migration in QD thin film plays the key role for the enhanced operational stability of QDSCs, confirmed by PL stability monitoring (shown in figure 2 a and b) and calculated activation energy for ion migration (figure 2 c and d).

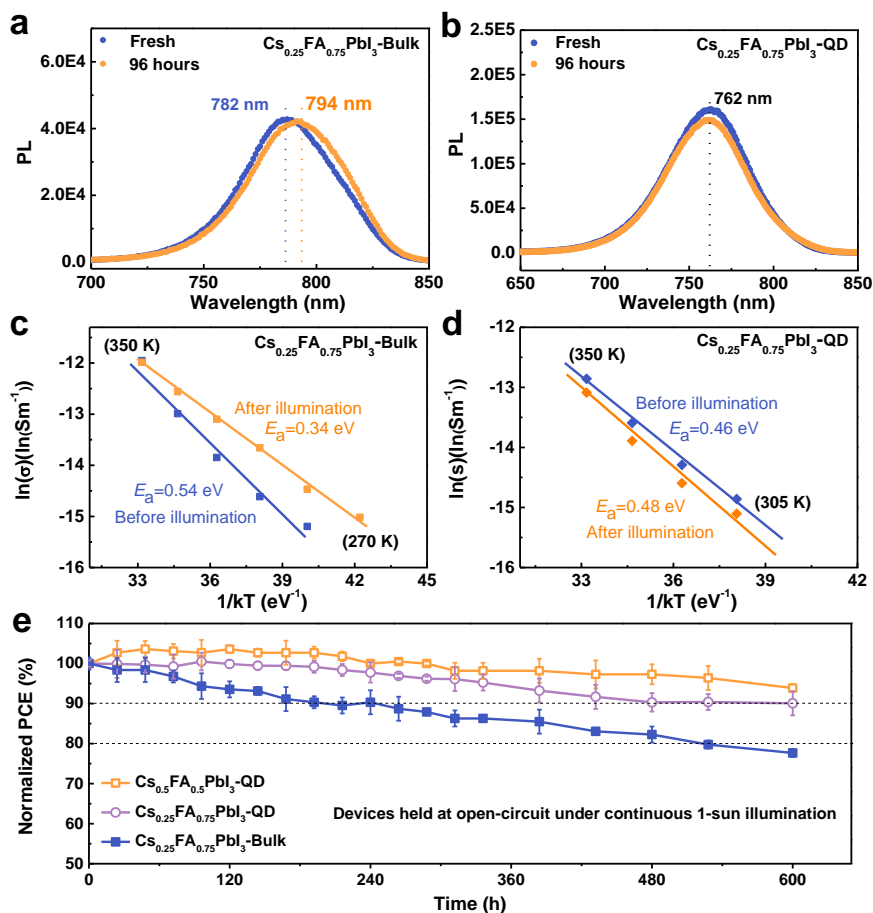


Figure 2. Stability of QD thin film and devices. PL spectra of $\text{Cs}_{0.25}\text{FA}_{0.75}\text{PbI}_3$ -Bulk film (a) and $\text{Cs}_{0.25}\text{FA}_{0.75}\text{PbI}_3$ -QD film (b) on SnO_2 -coated ITO substrates before and after illumination for 96 hours in N_2 , respectively. Arrhenius plot of the ion conductivity of $\text{Cs}_{0.25}\text{FA}_{0.75}\text{PbI}_3$ -Bulk film (c) and $\text{Cs}_{0.25}\text{FA}_{0.75}\text{PbI}_3$ -QD film (d) before and after illumination (25 mW cm^{-2}) for 3 hours, respectively. (e) Long-term stability of unencapsulated solar cells fabricated with $\text{Cs}_{0.25}\text{FA}_{0.75}\text{PbI}_3$ -Bulk film, $\text{Cs}_{0.25}\text{FA}_{0.75}\text{PbI}_3$ -QD film, and $\text{Cs}_{0.5}\text{FA}_{0.5}\text{PbI}_3$ -QD film monitored at open-circuit under 1-sun illumination in N_2 atmosphere.

$\text{Cs}_{1-x}\text{FA}_x\text{PbI}_3$ QDSCs with large bandgap absorber, like most other single-junction devices, have limitation in PCE according to the Shockley-Queisser analysis. The concept of QDs tandem solar cell based on large-bandgap perovskite QDs (with tuneable bandgap between 1.5 eV to 1.9 eV) and low-bandgap PbS QDs (with tuneable bandgap between 0.9 eV to 1.3 eV) was raised to further improve the PCE of QDSCs. The first step was the development of semi-transparent perovskite QDSCs with considerable transmittance and PCE. Simultaneous efforts were put into development of PbS QDSCs with optimized charge transport layer to achieve high PCE. With design of a novel solar cell structure, the damage of perovskite QDs bottom solar cell during the

fabrication of PbS top solar cell could be avoided. This work paved a fundamental way to fabricate QDs tandem solar cells, which may further improve the device efficiency significantly.

Figure 3. Semi-transparent $Cs_{1-x}FA_xPbI_3$ QDSC, (a) photo of device, (b) device structure, (c) I-V curve, (d) device transmittance. (e) TEM image of PbS QD, (f) Xrd spectrum of PbS QD film.

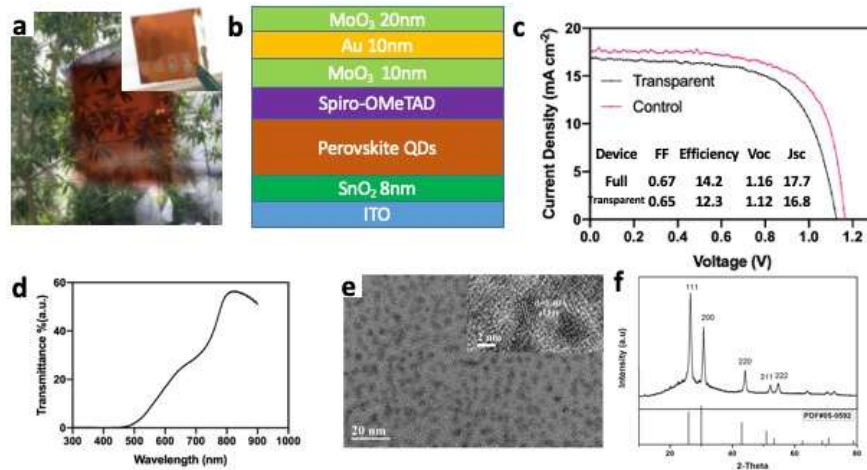


Table I. Parameters of PbS QDSCs with varies hole transfer materials

HTM	V_{oc} (V)	J_{sc} ($mA \cdot cm^{-2}$)	FF	Efficiency (%)
P3HT	0.516	22.2	55	6.314
EDT-PbS	0.526	24.6	57.3	7.43
PTB7	0.605	18.2	56.7	6.246
Spiro-OMeTAD	0.571	21.6	59.9	7.389

References

Hao, M., Bai, Y., Zeiske, S., Ren, L., Liu, J., Yuan, Y., ... & Wang, L., 2020, 'Ligand-assisted cation-exchange engineering for high-efficiency colloidal $Cs_{1-x}FA_xPbI_3$ quantum dot solar cells with reduced phase segregation', *Nature Energy*, 5, p79-88.