RESEARCH HIGHLIGHTS

Journal of Semiconductors
(2023) 44, 020201
doi: 10.1088/1674-4926/44/2/020201

Perovskite/organic tandem solar cells

Jie Sun[1, 2] and Liming Ding[1, †]

[1]Center for Excellence in Nanoscience (CAS), Key Laboratory of Nanosystem and Hierarchical Fabrication (CAS), National Center for Nanoscience and Technology, Beijing 100190, China

[2]University of Chinese Academy of Sciences, Beijing 100049, China

Citation: J Sun and L M Ding, Perovskite/organic tandem solar cells[J]. *J. Semicond.*, 2023, 44(2), 020201. https://doi.org/10.1088/1674-4926/44/2/020201

In recent years, the power conversion efficiency (PCE) for single-junction perovskite solar cells (PSCs)[1, 2] has reached 25.7%, approaching the Shockley-Queisser limit (S-Q limit). Further enhancing efficiency is challenging. Tandem solar cells offer an effective way to further increase the efficiency beyond S-Q limit. Currently, perovskite/silicon tandem solar cells (TSCs)[3–5] have achieved a PCE of 31.3%[6]. However, the complicated preparation processes and high cost hinder their commercialization application. In contrast, thin-film perovskite/organic TSCs have the advantages of solution processability, low cost, and flexibility, making them to be promising candidates for the next-generation photovoltaic technology.

The idea of combining perovskite with photoactive organics was first proposed by Ding *et al.*[7] in 2014, and they designed the first integrated solar cell structured with ITO/PEDOT:PSS/$CH_3NH_3PbI_3$/(PDPP3T:PC_{61}BM)/Ca/Al. A bulk heterojunction layer composed of PDPP3T:PC_{61}BM blend was inserted into a PSC structure, which broadened the spectral response to 970 nm. Though this structure is not a tandem structure, as there is no interconnecting layer (ICL), it still inspired the attempts to combine PSCs with organic materials to broaden the photoresponse. After that, Jen *et al.*[8] fabricated the first perovskite/organic TSCs with PBSeDTEG8 as the active layer of front subcell and $CH_3NH_3PbI_3$ as the active layer of rear subcell. The tandem cells gave a PCE of 10.23%. However, the structure with organic solar cell (OSC) as the front cell would cause serious deposition problem, as the high temperature and solvent treatment during the processing of perovskite layer could damage the already deposited OSC. Russell *et al.*[9] designed a ~90 nm thick perovskite-based front subcell and a ~100 nm thick polymer-based rear subcell, yielding a PCE of 15.9%. This structure allows long wavelength light to pass through PSC and to be utilized by the OSC (Fig. 1). Also, it avoids the damage to organic layer during processing.

After years of exploration, many effective strategies have been proposed to improve the photovoltaic performance of perovskite/organic TSCs, including broadening the light absorption, optimizing the ICL, and passivating the bulk and interfacial defects. The PCE for perovskite/organic TSCs has reached 23.1%[10]. The PCE progress for perovskite/organic TSCs is illustrated in Fig. 2, and the device details can be found in Table 1.

Broadening the absorption spectra. Perovskite and organic materials absorb photons of short and long wavelength, respectively. The absorption spectra for both perovskite and organic materials in early TSCs are overlapping, leading to insufficient sunlight utilization. Therefore, designing wider-bandgap perovskite and narrower-bandgap organic materials to enhance sunlight absorption is an effective approach to improve the performance of TSCs. In 2019, Ding *et al.*[11] developed a 2T tandem solar cell with $CsPbI_2Br$ cell as the front cell with high response to light before 650 nm and PTB7-Th:CO_i8DFIC:PC_{71}BM ternary cell as the rear cell absorbing light at 650–1050 nm. The TSC offered a 15.04% PCE. In 2020, Cao *et al.*[12] used a wide-bandgap (WBG) $CsPbI_2Br$ front cell in series with a narrow-bandgap (NBG) PM6:Y6 rear cell and obtained a TSC with a 18.4% PCE. The thickness control of perovskite layer is vital for the optimal utilization of the solar spectrum. If it is too thin, the short-wavelength photons cannot be absorbed completely; when it is too thick, the transmittance would be greatly reduced. In 2021, Ding *et al.*[13] further enhanced the efficiency of inorganic perovskite/organic TSCs to 20.18% by integrating a WBG $CsPbI_2Br$ front cell and a D18:Y6 organic rear cell. Since then, scientists have focused on varying the ratio of I and Br in $CsPbI_2Br$ to tune the bandgap, pushing the PCE to more than 21%[14, 15]. Organic–inorganic hybrid perovskite was also used in perovskite/organic TSCs in recent years. In 2020, Yang *et al.*[16] proposed a semi-empirical model to select the optimum combination of materials for perovskite/organic TSCs. The optimal combination was $FA_{0.8}MA_{0.02}Cs_{0.18}PbI_{1.8}Br_{1.2}$ (1.77 eV) and PBDBT-2F:Y6:PC_{71}BM (1.41 eV), and the TSCs gave high PCE (certified PCE of 19.54%) and high reproducibility. In 2022, Cao *et al.*[17] used $MAPbI_2Br$ doped with $Pb(SCN)_2$ front cell and an organic rear cell to make a TSC with over 20.03% PCE. In 2022, Li *et al.*[18] synthesized a small molecule acceptor BTPV-4Cl-eC9 to expand the photoresponse to infrared region, and they combined PTB7-Th:BTPV-4Cl-eC9 rear cell with $FA_{0.6}MA_{0.4}Pb(I_{0.6}Br_{0.4})_3$ front cell to make high-performance TSCs with a PCE of 22%.

Optimizing the ICL. The ICL connects two subcells to achieve electron and hole recombination and protects the front cell from being destroyed during the deposition process of the rear cell. A good ICL possesses good transmittance, chemical stability, and low resistive losses. In 2016, C_{60}-SB/Ag/MoO_3 was used as the ICL[9], which alleviated the damage of thermal annealing and chemical treatment to polymer front cells during the process of perovskite layer. In 2020, Yang *et al.*[16] demonstrated a simplified ICL structure

Correspondence to: L M Ding, ding@nanoctr.cn
Received 3 DECEMBER 2022.

Fig. 1. (Color online) The structure for perovskite-organic tandem solar cells.

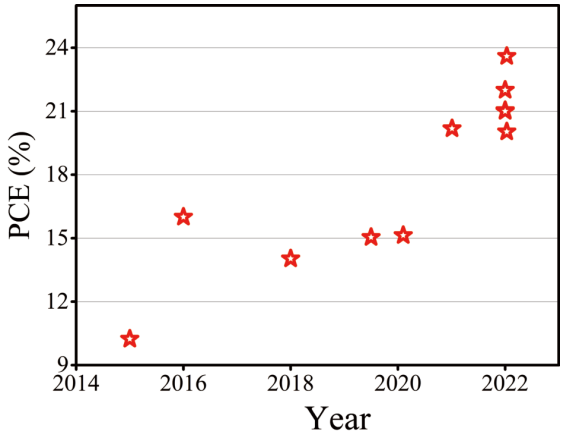


Fig. 2. (Color online) The PCE advance for perovskite-organic tandem solar cells.

Table 1. Performance data for perovskite-organic tandem solar cells. (* indicates the certified PCE)

Front cell	ICL	Rear cell	V_{oc} (V)	J_{sc} (mA/cm^2)	FF (%)	PCE (%)	Ref.
ITO/PEDOT:PSS/PBSeDTEG8:$PC_{61}BM$/PFN/TiO_2	PEDOT:PSS PH500/PEDOT:PSS 4083	$CH_3NH_3PbI_3$/$PC_{61}BM$/PFN/Al	1.52	10.05	67.0	10.23	[8]
ITO/PEDOT:PSS/$CH_3NH_3PbI_3$/$PC_{61}BM$	C_{60}-SB/Ag/MoO_3	PCE-10:$PC_{71}BM$/C_{60}-N/Ag	1.62	12.90	75.0	16.00	[9]
ITO/SnO_2/$CsPbI_2Br$/PTAA	MoO_3/Au/ZnO	PTB7-Th:CO_i8DFIC:$PC_{71}BM$/MoO_3/Ag	1.71	11.98	73.4	15.04	[11]
ITO/PTAA/$Cs_{0.1}(FA_{0.6}MA_{0.4})_{0.9}Pb(I_{0.6}Br_{0.4})_3$/$PC_{61}BM$/BCP	Ag/PEDOT:PSS	PBDB-T:SN6IC-4F/C60-bis/BCP/Ag	1.85	11.52	71.0	15.13	[20]
ITO/ZnO/SnO_2/$CsPbI_2Br$/PDCBT	MoO_3/Ag/ZnO	PM6:Y6/MoO_3/Ag	1.95	12.46	75.6	18.38	[12]
ITO/NiO_x/$FA_{0.8}MA_{0.02}Cs_{0.18}PbI_{1.8}Br_{1.2}$/$C_{60}$	BCP/Ag/MoO_x	PBDBT-2F:Y6:$PC_{71}BM$/TPBi/Ag	1.90	13.05	83.1	19.54*	[16]
ITO/SnO_2/ZnO/$CsPbI_2Br$	PTAA/MoO_3/Au/ZnO	D18:Y6/MoO_3/Ag	2.05	13.07	25.3	20.18	[13]
ITO/SnO_2/$CsPbI_{1.8}Br_{1.2}$/TACl	PBDB-T/MoO_3/Au/ZnO/PFN	PM6:Y6/MoO_3/Al	2.05	13.36	76.8	21.04	[14]
ITO/ZnO/$CsPbI_2Br$/polyTPD	MoO_3/Ag/PFN-Br	PM6:Y6-BO/MoO_3/Ag	1.96	13.30	80.8	21.10	[15]
ITO/Poly-TPD/($MAPbI_2Br$ + $Pb(SCN)_2$)	PCBM/BCP/Au/MoO_3	PM6:Y6/PFN-Br/Ag	1.94	13.12	78.7	20.03	[17]
ITO/poly-TPD/($MA_{0.9}FA_{0.1}PbI_2Br$ + $Pb(SCN)_2$)	PCBM/BCP/Au/MoO_3	PM6:CH1007/PFN-Br/Ag	1.96	13.80	78.4	21.2	[21]
ITO/2PACz/$FA_{0.6}MA_{0.4}Pb(I_{0.6}Br_{0.4})_3$	C_{60}/BCP/Ag/MoO_x	PTB7-Th:BTPV-4Cl-eC9/PDINN/Ag	1.88	15.70	74.6	22.00	[18]
ITO/PTAA/$FA_{0.8}Cs_{0.2}Pb(I_{0.5}Br_{0.5})_3$/PEAI/$PC_{61}BM$/AZO-NP	SnO_x/InO_x/MoO_x	PM6:Y6:$PC_{61}BM$/C_{60}/BCP/Ag	2.15	14.00	80.0	23.10*	[10]
ITO/NiO_x/BPA/$Cs_{0.25}FA_{0.75}Pb(I_{0.6}Br_{0.4})_3$	C_{60}/BCP/CRL/MoO_x	PM6:Y6/PNDIT-F3N/Ag	2.05	14.83	77.2	22.94*	[19]

by using a thin MoO_x/Ag/BCP layer, yielding low optical loss and high reproducibility. In 2021, Ding *et al.*[13] used PTAA/MoO_3/Au/ZnO as the ICL. The thin Au layer (1 nm) reduces the charge-transport barrier and contributes to the high PCE of 20.18%. The above ICLs adopted a thin layer of metal inserted between two charge-transport layers. However, despite the excellent conductivity of metal, it is not transparent, causing optical losses. Very recently, Riedl *et al.*[10] inserted an ultrathin InO_x layer (1.5 nm) made by atomic layer deposition (ALD) to eliminate the Schottky barrier between the low-workfunction SnO_x and the high-workfunction MoO_x. InO_x avoided the use of metal layer and the optical loss is negligible, greatly improving the photovoltaic performance of TSCs. Hou *et al.*[19] developed C_{60}/BCP/IZO/MoO_x ICL. IZO can improve carrier recombination and minimize the optical losses at the same time, yielding a certified PCE of 22.94%. Cao *et al.*[17] investigated the influence of ICL thickness on the performance of TSCs. Thin ICL may lead to poor device performance due to inefficient recombination, while thick ICL can lead to inefficient electron extraction, lower subcell EQE and device efficiency, so the ICL thickness needs to be optimized. Overall, an excellent ICL layer should be chemically inert, conductive, and transparent, providing sufficient recombination sites between the front and rear subcells.

Passivating bulk and interfacial defects. A large number of defects exist in polycrystalline perovskite films, especially at the interface between ICL and perovskite layer, which will cause severe nonradiative recombination. It leads to large V_{oc} loss and undermines the long-term stability of perovskite front cell[13]. So, interfacial passivation is essential for improving the performance of TSCs. In 2020, Jen *et al.*[20] used phenmethylammonium bromide to passivate the WBG perovskite layer $Cs_{0.1}(FA_{0.6}MA_{0.4})_{0.9}Pb(I_{0.6}Br_{0.4})_3$ and the TSCs gave a PCE of 15.13%. In 2021, Cao *et al.*[21] reduced defects at grain boundaries and improved phase stability through compositional engineering. FA^+ was incorporated, and $MA_{0.9}FA_{0.1}PbI_2Br$ doped with $Pb(SCN)_2$ presented homogeneous crystallization and reduced SCN^--induced PbI_2 excessive aggregation. The TSCs with PM6:CH1007 subcell gave a

PCE of 21%. In 2022, Li *et al.*[18] introduced organic cation ClFA$^+$ to $FA_{0.6}MA_{0.4}Pb(I_{0.6}Br_{0.4})_3$ film to passivate bulk defects in perovskite films. A high V_{oc} of 1.25 V and a high FF of 83.0% for the perovskite front cell were achieved. In 2022, Riedl *et al.*[10] used large organic cation-based halides to passivate perovskite surface by forming a two-dimensional (2D) layer. The valence band was lowered to prevent holes from reaching the electron extraction layer, and an efficiency of 23.1% was achieved. In 2022, Hou *et al.*[19] used nickel oxide hole-transport layer passivated by phenylphosphonic acid, and obtained a certified efficiency of 22.94%.

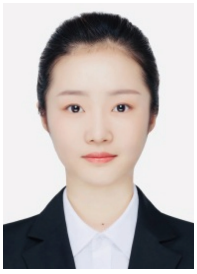
In short, perovskite-organic TSCs already demonstrate a record PCE of 23.1%. Further work should focus on long-term stability, reproducible and reliable fabrication, and low cost, which are very important for future commercialization.

Acknowledgements

L. Ding thanks the National Key Research and Development Program of China (2022YFB3803300), the open research fund of Songshan Lake Materials Laboratory (2021SLABFK02), and the National Natural Science Foundation of China (21961160720) for financial support.

References

[1] Zhang L X, Pan X Y, Liu L, et al. Star perovskite materials. J Semicond, 2022, 43, 030203
[2] Kim M, Jeong J, Lu H, et al. Conformal quantum dot-SnO_2 layers as electron transporters for efficient perovskite solar cells. Science, 2022, 375, 302
[3] Cheng Y H, Ding L M. Perovskite/Si tandem solar cells: Fundamentals, advances, challenges, and novel applications. SusMat, 2021, 1, 324
[4] Chen S, Zuo C T, Xu B M, et al. Monolithic perovskite/silicon tandem solar cells offer an efficiency over 29%. J Semicond, 2021, 42, 120203
[5] Fang Z M, Zeng Q, Zuo C T, et al. Perovskite-based tandem solar cells. Sci Bull, 2021, 66, 621
[6] National Renewable Energy Laboratory. Best Research-Cell Efficiencies. 2022
[7] Zuo C T, Ding L M. Bulk heterojunctions push the photoresponse of perovskite solar cells to 970 nm. J Mater Chem A, 2015, 3, 9063
[8] Chen C C, Bae S H, Chang W H, et al. Perovskite/polymer monolithic hybrid tandem solar cells utilizing a low-temperature, full solution process. Mater Horizons, 2015, 2, 203
[9] Liu Y, Renna L A, Bag M, et al. High efficiency tandem thin-perovskite/polymer solar cells with a graded recombination layer. ACS Appl Mater Interfaces, 2016, 8, 7070
[10] Brinkmann K O, Becker T, Zimmermann F, et al. Perovskite-organic tandem solar cells with indium oxide interconnect. Nature, 2022, 604, 280
[11] Zeng Q, Liu L, Xiao Z, et al. A two-terminal all-inorganic perovskite/organic tandem solar cell. Sci Bull, 2019, 64, 885
[12] Xie S, Xia R, Chen Z, et al. Efficient monolithic perovskite/organic tandem solar cells and their efficiency potential. Nano Energy, 2020, 78, 105238
[13] Liu L, Xiao Z, Zuo C, et al. Inorganic perovskite/organic tandem solar cells with efficiency over 20%. J Semicond, 2021, 42, 020501
[14] Chen W, Li D, Chen X, et al. Surface reconstruction for stable monolithic all-inorganic perovskite/organic tandem solar cells with over 21% efficiency. Adv Funct Mater, 2022, 32, 2109321
[15] Wang P, Li W, Sandberg O J, et al. Tuning of the interconnecting layer for monolithic perovskite/organic tandem solar cells with record efficiency exceeding 21%. Nano Lett, 2021, 21, 7845
[16] Chen X, Jia Z Y, Chen Z, et al. Efficient and reproducible monolithic perovskite/organic tandem solar cells with low-loss interconnecting layers. Joule, 2020, 4, 1594
[17] Xie Y M, Niu T, Yao Q, et al. Understanding the role of interconnecting layer on determining monolithic perovskite/organic tandem device carrier recombination properties. J Energy Chem, 2022, 71, 12
[18] Qin S, Lu C, Jia Z, et al. Constructing monolithic perovskite/organic tandem solar cell with efficiency of 22.0% via reduced open-circuit voltage loss and broadened absorption spectra. Adv Mater, 2022, 34, 2108829
[19] Chen W, Zhu Y, Xiu J, et al. Monolithic perovskite/organic tandem solar cells with 23.6% efficiency enabled by reduced voltage losses and optimized interconnecting layer. Nat Energy, 2022, 7, 229
[20] Li Z, Wu S F, Zhang J, et al. Hybrid perovskite-organic flexible tandem solar cell enabling highly efficient electrocatalysis overall water splitting. Adv Energy Mater, 2020, 10, 2000361
[21] Xie Y M, Yao Q, Zeng Z, et al. Homogeneous grain boundary passivation in wide-bandgap perovskite films enables fabrication of monolithic perovskite/organic tandem solar cells with over 21% efficiency. Adv Funct Mater, 2022, 32, 2112126



Jie Sun got her BS from Minzu University of China in 2021. Now she is a PhD student at University of Chinese Academy of Sciences under the supervision of Prof. Liming Ding. Her research focuses on perovskite devices.

Liming Ding got his PhD from University of Science and Technology of China (was a joint student at Changchun Institute of Applied Chemistry, CAS). He started his research on OSCs and PLEDs in Olle Inganäs Lab in 1998. Later on, he worked at National Center for Polymer Research, Wright-Patterson Air Force Base and Argonne National Lab (USA). He joined Konarka as a Senior Scientist in 2008. In 2010, he joined National Center for Nanoscience and Technology as a full professor. His research focuses on innovative materials and devices. He is RSC Fellow, and the Associate Editor for *Journal of Semiconductors*.