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Recent UpdateonPerovskite/Organic Tandem Solar Cells

The emergence of multiple-junction photovoltaics (PVs) has presented a remarkable opportunity to overcome the Shockley-Queisser limit of single-junction solar cells. Recently, perovskite solar cells (PSCs) and organic photovoltaics (OPVs) are two of the most promising new-generation PVs, which have gained widespread attention in the PV community due to their exceptional rapid growth in their power conversion efficiencies (PCEs). Combining PSCs and OPVs in tandem structures offers numerous advantages, such as the ability to tune the bandgap of absorbers to regulate the absorption bands and enhance transparency. The use of thin-film technology ensures that the devices are lightweight and flexible, which is particularly advantageous for certain applications. Furthermore, both PSCs and OPVs are low-cost, making them attractive for large-scale deployment in the future. These advantages will make PSC/OPV tandem devices promising for applications beyond traditional silicon-based PVs. This review provides an up-to-date account on the recent progress of PSC/OPV tandem PVs. The state-of-the-art fabrication techniques and material engineering on the properties of PSC and OPV sub cells as well as their functional layers are discussed. A perspective guidance is also given to direct the future development of this type of tandem PVs. This paper provides an insight into the development of PSC/OPV tandem PVs, providing researchers with a roadmap to advance this technology further and unlock its full potential in the field of renewable energy.

Keywords: Perovskites, organic photovoltaics, tandem solar cells, device engineering.

Introduction

Due to the high energy demands and the risks caused by global warming, developing alternative energy sources is essential to address these urgent challenges. Solar energy is a promising renewable source of energy, which is safe, cost-effective and clean. In order to utilize solar energy, photovoltaics (PVs) are the key device for converting the solar energy to electrical energy. The history of the PV effect was discovered by the French physicist, Alexandre-Edmond Becquerel in 1839. The first practical application of photovoltaic (PV) technology was realized in 1954 with the development of the first silicon (Si) solar cell, which achieved a power conversion efficiency (PCE) of 6 % [1]. Nowadays PV technologies are classified into three generations. PV cells based on single-junction silicon wafers (monocrystalline and multicrystalline) and gallium arsenide (GaAs) are considered 1st generation PV technologies. Recently, the PCE of the single-junction GaAs and crystalline Si PV have reached 26.8 % and 29.1 %, respectively [2]. The 2nd generation of PV technologies utilizes thin-film materials such as amorphous silicon (a-Si), cadmium telluride (CdTe), and copper indium gallium selenide (CIGS) to achieve a more cost-effective device architecture for single junction devices, while they typically have a lower PCE compared to first-generation PV technologies. Currently, the highest efficiencies of CIGS, CdTe and a-Si: H thin-film solar cells are 23.6 %, 22.1 % and 14.0 %, respectively. The 3rd generation of solar cells is based on the emerging materials such as conductive polymers, organic dyes, quantum dots (QDs) and perovskites, etc. This type of emerging PVs provides promising alternatives of conventional Si-based PVs systems, ensuring more cost-effective manufacturing processes and offering a wide range of potential applications. The certified record PCEs of emerging PVs are 25.8 %, 19.2 %, 18.1 % and 13 % for perovskite solar cells (PCSs), organic photovoltaics (OPVs), QD cells and dye-sensitized cells, respectively [2]. Among different types of third-generation PV technologies, PSCs and OPVs are currently the two most promising examples, with rapidly PCEs being recorded for both technologies. PSCs are based on perovskite materials with a chemical formula of ABX_3 (A — monovalent cations, B — divalent metal cations, and X — halogen ions). The halide perovskite materials have gained tremendous attention from researchers due to their excellent optoelectronic properties, such as high light absorption coefficients, tunable band-gaps, long carrier diffusion lengths and carrier lifetime, as well as high carrier mobility [3]. Recently, the invention of NFAs (NFA) with a broader absorption range and tunable energy levels has also made a significant breakthrough in materials properties of polymer: NFA system, leading to very promising performance of OPVs [4].

It is noteworthy that the single junction devices face critical problems that limit their efficiency. The first challenge is thermalization loss, caused by thermal relaxation when the energy of an absorbed photon is greater than the bandgap (Eg) of the device. The second issue is photon transmission loss, where photons with energy below the bandgap cannot be absorbed [5]. The theoretical PCE limit in single junction devices was determined as 33.7 % (Shockley and Queisser (SQ) limit) [6]. Currently the existing single junction devices are approaching SQ limit. Development of multi-junction solar cells can be the solution to surpass the SQ limit by connecting 2 or more subcells with complementary absorption spectra in a tandem structure. It was calculated that a PCE of around 46.1 % can be theoretically achieved by the tandem devices [5]. Nowadays, a lot of investigations have been performed on PSCs/Si tandem structure and a record PCE of 33.2 % have been recently announced [2]. However, the large difference of lifetime between silicon and PSCs hinders the progress of their commercialization. The expensive fabrication processes and opaque properties of Si also limit its potential for widespread use in conditions that require transparency or flexibility. On the other hand, perovskite/organic tandem solar cells have attracted increasing attention as these two types of PVs possess compatible material and device properties, making them a good pair in tandem structures. PSCs and OPVs can be fabricated by similar cost-effective thin-film deposition techniques on both rigid and flexible substrates. The bandgap of absorbers and transparency of the absorbers can be tuned according to the requirements of the applications, creating more freedom in tailoring the tandem device properties for future innovative products. Nevertheless, the PSC/OPV tandem PVs are still in the stage of research, the potential for further growth in PCE will be significant as the theoretical calculations have shown that a PCE higher than 30 % is achievable for this type of tandem PVs [7-9]. To provide prospective guidance for further studies of PSC/OPV tandem PVs, the recent achievements in this area are summarized, which provides analysis of the properties of each subcell and functional layer in the tandem structures, and outlines future directions for the development of this type of PSC/ OPV tandem technology in the PV community.

Table

Summary of perovskite/organic Tandem solar cells

	Sub cell	2T/ 4T	Tandem configuration	Band-gap, (eV)	V_{oc} (V)	J_{SC} (mA/ cm^2)	FF (%)	PCE (%)	Ref.
1	$FA_{0.8}Cs_{0.2}Pb(I_{0.5}Br_{0.5})_3$	2T	MgF_2/ Glass/ITO/MeO-2PACz/WBG/PEAI /$PC_{61}BM$/ AZO NP / SnO_X / InO_X / MoO_X/NBG /C_{60} / BCP / Ag	1.85	2.15	17.90	80	23.1 %	[10] *(2022)*
	PM6: Y6: PC61BM (1:1.2:0.2)			1.33					
2	$Cs_{0.25}FA_{0.75}Pb(I_{0.6}Br_{0.4})_3$	2T	ITO/NiO_X/BPA/WBG/ C_{60}/BCP/ IZO/ MoO_X/ NBG / PNDIT-F3N /Ag	1.79	2.06	14.83	77.20	23.60 %	[11] *(2022)*
	PM6: Y6: P71CBM			1.36					
3	$MAPbI_2Br + Pb(SCN)_2)$	2T	ITO/Poly-TPD/WBG/ PCBM/BCP/Au/MoO_3/ NBG /PFN-Br/Ag	1.7	1.94	13.12	78.7	20.03	[12] (2022)
	PM6: Y6			1.3*					
4	$MAPbI_{2.5}Cl_{0.5}$	2T	ITO/PVBT-SO_3/WBG/ $PC_{61}BM$/ C_{60}-ionene/Ag/MoO_3/ NBG/C_{60}-ionene/ Ag	1.6*	1.86	12.4	79	19.2	[13] (2022)
	PM6: Y6			1.31*					
5	$FA_{0.8}Cs_{0.2}Pb(Br_{0.4}I_{0.6})_3$	2T	MgF2/Glass/ITO/2PACz/ WBG/C_{60}/SnO_2/PEDOT: PSS/NBG/PFN-Br/ Ag	1.78	1.83	13.8	69.4	17.6	[14] (2022)
	PTB7-Th: IEICO-4F			1.24					
6	$FA_{0.6}MA_{0.4}Pb(I_{0.6}Br_{0.4})_3$ + ClFA	2T	ITO/2PACz/WBG/C_{60}/ BCP/ Ag/MoO_X/NBG/PDINN/Ag	1.79	1.88	15.7	74.6	22.0	[15] (2022)
	PTB7-Th: BTPV-4Cl-eC9			1.22					

7	$MA_{0.96}FA_{0.1}PbI_2Br(SCN)_{0.12}$	2T	ITO/poly-TPD/WBG/PCBM/BCP/Au/MoO$_3$/NBG/PFN-Br/Ag	1.75*	1.96	13.8	78.4	21.2	[16] (2022)
	PM6: CH1007			1.31*					
8	$CsPbI_{1.8}Br_{1.2}$/TACl	2T	ITO/SnO$_2$/WBG/TACl/PBDB-T/MoO$_3$/Au/ZnO/PFN/ NBG/MoO$_3$/Al	1.93	2.05	13.36	76.82	21.04	[17] (2021)
	PM6: Y6			1.4					
9	$CsPbI_2Br$	2T	ITO/ZnO/WBG/poly-TPD/MoO$_3$/Ag/PFN-Br/NBG/MoO$_3$/Ag	1.91*	1.96	13.3	80.8	21.1	[18] (2021)
	PM6: Y6-BO			1.3*					
10	$CsPbI_2Br$	2T	ITO/SnO$_2$/ZnO/WBG /PTAA/MoO$_3$/Au/ZnO/NBG/MoO$_3$/Ag	1.91*	1.96	13.07	80.8	20.9	[19] (2021)
	D18: Y6			1.33*					
11	$CsPbI_2Br$	2T	ITO/ZnO/SnO$_2$/WBG/PDCBT / MoO$_3$/Ag/ZnO/NBG /MoO$_3$/Ag	~1.9	1.95	12.5	76	18.4	[20] (2020)
	PM6: Y6			1.31*					
12	$CsPbI_2Br$	2T	ITO/ZnO/SnO$_2$/WBG/PDCBT / MoO3/Ag/ZnO /NBG /MoO3/Ag	~1.9	1.86	12.9	75	18.1	[20] (2020)
	PTB7-Th: O6T-4F			1.24*					
13	$FA_{0.8}MA_{0.02}Cs_{0.18}PbI_{1.8}Br_{1.2}$	2T	ITO/NiOx/WBG/ C$_{60}$ / BCP/ Ag/ MoO$_X$/ NBG/ TPBi/ Ag	1.77	1.902	13.05	83.1	20.6 (19.54)	[21] (2020)
	PBDBT-2F: Y6: PC$_{71}$BM			1.41					
14	$CsPbI_2Br$	2T	TO/SnO$_2$/WBG/ P3HT/MoO$_3$/Ag/PFN-Br/NBG/MoO$_3$/Ag	1.91	1.82	13.2	71.68	17.24	[22] (2020)
	PTB7-Th: IEICO-4F			1.24					
15	$Cs_{0.1}(FA_{0.6}MA_{0.4})_{0.9}Pb(I_{0.6}Br_{0.4})_3$	2T	ITO/PTAA/WBG/PMABr/ PCBM/BCP/Ag/M-PEDOT/NBG/ Bis-C60/BCP/Ag	1.74	1.85	11.52	70.98	15.13	[23] (2020)
	PBDB-T: SN6IC-4F			1.3					
16	$CsPbI_2Br$	2T	(ITO)/SnO$_2$/WBG/PTAA/MoO$_3$/Au/ZnO/NBG/MoO$_3$/Ag	1.92	1.71	11.98	73.4	15.04	[24](2019)
	PTB7-Th: COi8DFIC: PC$_{71}$BM			1.2					
17	$MAPbI_3$	2T	ITO/PEDOT: PSS/WBG/PC$_{61}$BM/C$_{60}$-SB/Ag/ MoO$_3$/ NBG/C$_{60}$-N/Ag	1.5	1.63	13.1	75.1	16	[25] (2016)
	PCE-10: PC71BM			1.46*					
18	$CH_3NH_3PbI_3$	2T	ITO/PEDOT: PSS/NBG/PFN/TiO$_2$/ PEDOT: PSS PH500/PEDOT: PSS 4083/WBG/PC$_{61}$BM/PFN/Al	1.52	1.52	10.05	67	10.23	[26] (2015)
	PBSeDTEG8: PC$_{61}$BM			1.28					
19	$CsPbI_2Br$	4T	ITO/SnO$_2$/ZnO/ WBG/HTL/ MoO$_3$/ITO + ITO/PEDOT: PSS/NBG/PDIN/Ag	1.92	1.26	13.9	73.99	21.25	[27] (2022)
	D18-Cl-B: N3: PC$_{61}$BM			1.31*	0.84	27.37	78.6		
20	$CsPbBr_3$	4T	ITO/ZnO/WBG/ Spiro-OMeTAD/ transfer laminated PH1000+ ITO/ZnO/NBG/MoO$_3$/Al	2.3	1.38	6.15	70.5	14.03	[28] (2018)
	PBDB-T-SF: IT-4F			1.46*	0.88	12.18	75.2		

** The values are calculated from the published EQE or absorption spectra.*

1. **Narrow-bandgap subcell**

The use of materials and device structures as well as the corresponding latest photovoltaic performance based on PSC/OPV tandem PVs are summarized in Table [10-28]. Nowadays, the majority of works use OPVs as the narrow bandgap subcells with a range from 1.20 eV to 1.46 eV in a tandem structure with PSCs. The absorber layer is composed of blending polymer donor, NFA and/or fullerene acceptors in an optimized ratio. The NFA based material system becomes the dominant absorber in OPVs used in tandem structure with PSC. In comparison with OPVs based on fullerene derivatives, NFA based devices have further enhanced absorption properties due to tunable energy levels and wider absorption range of the active layer [29]. Additionally, a lower driving force is needed for NFA based OPV to overcome the exciton binding energy, which leads to minimum V_{OC}losses and higher J_{SC} [29-30]. Moreover, it is found that NFA based devices are more stable under the light with minor burn-in losses [30]. Y6 and IEICO-4F are commonly used as NFA materials in the OPV subcell of the tandem devices, with the absorption spectra extended to near infrared (NIR) region and a peak absorption at 810 nm and 865 nm, respectively [4]. The OPVs with an active layer composed of non-fullerene acceptors (NFA) blended with common polymer donors, such as PM6, a member of the PBDB-T family, and PTB7-Th, have exhibited high PCEs over 14 % in single-junction solar cells [30]. Due to their excellent optoelectronic properties, this type of OPV has been widely used in tandem structures with PSCs. The recent development of copolymer D18 with a high hole mobility of 1.59×10^{-3} cm^2 V^{-1} s^{-1} blended with Y6 in the single-junction OPV has demonstrated a promising certificated PCE of 17.6 % [31]. The D-A copolymer donor D18 blended with NFA has been also applied in OPVs tandem with PSCs in 2T and 4T structures, for which both of the tandem structures exhibit a PCE of ~21 % [19],[27], indicating a huge potential of this type of OPV subcell for future development of high-efficiency PSC/OPV tandem PVs. The absorption onset for the D18, PM6 and PTB7-Th in thin films is 625 nm, 688 nm and 785 nm respectively, corresponding to an optical bandgap of 1.98 eV, 1.80 eV and 1.58 eV [30-31]. The absorption spectra of narrow-bandgap NFAs are complementary to those of common polymer donors used in OPVs, ensuring a wide absorption range extended to the long-wavelength region, which makes NFA based OPV sub cells well-suited for using in tandem structures with wide bandgap PSCs.

The non-radiative recombination which reduces the charge transport efficiency in OPV subcell, is one of the major challenges in achieving high-performance devices. By using fullerene as additives in the binary bulk heterojunction (BHJ) to form a ternary blend system, which enables better photovoltaic performance of OPVs as well as the overall efficiency of the tandem devices. It is found that adding an optimized amount of fullerene additives to the NFA based bulk BHJ active layer can introduce multiple positive effects, such as improved molecular packing for better carrier transport, enhanced light harvesting power, and better energy alignment etc. [10], [21], [24], [32-33]. In addition to enhancement of carrier transport, ternary OPVs are more flexible in controlling complementary absorption bands of the donor and acceptor components in the active layer to optimize the absorption range of the OPV subcell. Brinkmann *et al.* [10] added an optimized amount of PC61BM into the PM6: Y6 blend for improving the blend morphology of the photoactive layer, resulting in enhanced charge transportation and reduced non-radiative recombination in the PSC/OPV tandem solar cells, yielding a PCE of 17.5 % of OPV sub cell and 23.1 % for tandem with a PSC [10]. The high non-radiative recombination rate is one of the reasons for the low external quantum efficiency (EQE) of the OPVs. The EQE spectrum of binary PM6: Y6 extends beyond 900 nm and ternary cells exhibit enlarged EQE beyond 85 % at $\lambda > 650$ nm [10], which is suitable for use in tandem solar cells with a wide-gap front-cell. Other works reported by Chen *et al.* [21] and Zeng *et al.* [24] also used techniques of adding fullerene derivatives to form a ternary system in fabrication of OPVs. Jeong *et al.* [34] reported an improved PCE from 14.9 % to 15.6 % can be obtained from OPVs based on a ternary blend system by forming the BHJ blend with a fullerene self-assembled monolayer (C60-SAM). The OPVs with this ternary system were also found to retain 90 % of their original PCE after 8 hours of illumination. Similar approach of incorporation of PC71BM into the binary system has been reported by Chen *et al.* [11]. The OPVs based on the system of PBDBT-2F: Y6: PC71BM (1:1.2:0.2) possess balanced charge carrier transports, yielding a PCE of 15.8 % and a high PCE of 20.6 % (certified as 19.54 %) for 2T PSC/OPV monolithic tandem device.

Although the BHJ structure is widely used in OPVs, the random mixing of materials to form donor-acceptor blends creates challenges in controlling the morphology of the resultant active layer, reducing the batch-to-batch reproducibility. Recently, a sequential layer-by-layer (LBL) fabrication technique has been reported to increase the control of morphology formation and promote vertical phase separation in the OPVs. Qin *et al.* [15] have compared the fabrication techniques for forming LBL and BHJ structure and found the advantages of using LBL technique for preparing the rear OPV subcell based on PTB7-Th: BTPV-4Cl-eC9.

It is found that application of LBL leads to a better phase of the donor and acceptor, which can decrease the trap density and inhibit the non-radiative recombination [15]. The thin films in the LBL structure can facilitate charge extraction and transport at the interconnecting layer (ICL). The LBL-OPV subcell exhibits a higher J_{SC} and FF compared to the BHJ based device. The J_{SC} and FF of the LBL-OPV subcell are 28.6 mA cm^{-2} and 69.2 %, respectively, while those of the BHJ-based device are 27.8 mA cm^{-2} and 65.2 %, showing LBL as an effective technique for improving the performance of OPVs. A PCE of 22 % can be realized by fabricating 2T-tandem based on PSC/LBL-OPV. It is also interesting to note that the OPV subcells in monolithic tandem devices are usually fabricated in a regular structure (p-i-n), which can be due to the feasibility of deposition methods of preparing typical high-performance functional layers for OPVs and matching with the well-established inverted structures of organo-metal halide-based PSCs.

2. **Wide-bandgapsubcell**

The perovskite materials are commonly used in the wide-bandgap front cells in the tandem devices. The organic-inorganic hybrid perovskites (i.e. multiple cation mixed halide perovskite) and Cs-based all-inorganic perovskites (e.g. $CsPbI_2Br$, $CsPbI_{1.8}Br_{1.2}$) are the two major absorbers used in the PSCs. The bandgap of the perovskites can be tuned by changing the amount of monovalent cations and halogen ions in the range of 1.5 — 2.3 eV [15]. However, the chemical instability of organic–inorganic perovskites upon exposure to moisture, UV-light, and heat caused by the weakly bound of organic components in the hybrid crystal structure is still a critical problem for the device compliance with specific application requirements [35]. The use of all-inorganic perovskites such as $CsPbI_{3-x}Br_x$(x = 0, 1, 2, and 3) as light-harvesters by completely replacing organic cations with inorganic Cs^+ ions has emerged as a promising technology for improving the thermal stability of PSCs.

The defects in the bulk of perovskites and at the material interfaces are the key factors that lead to non-radiative charge recombination resulting in poor device efficiency and stability. Tremendous works have been done on passivation engineering of single junction PSCs [36], but they are also useful to implement in PSC/OPV tandem devices. *Li et al.* [23] used phenmethylammonium bromide (PMABr) as an interlayer between perovskite film and ETL, to passivate the defects in grain boundaries of $Cs_{0.1}(FA_{0.6}MA_{0.4})_{0.9}Pb(I_{0.6}Br_{0.4})_3$. The PL and TRPL results show that using PMABr for passivation increased the PL photon energy, resulting in a blueshift of PL peaks and suppressed nonradiative recombination, yielding a PCE of 15.13 % for the tandem device [23]. Qin *et al.* used chloro-formamidinium (ClFA) to passivate bulk defects in the $FA_{0.6}MA_{0.4}Pb(I_{0.6}Br_{0.4})_3$ based PSCs. It is also found that the use of CIFA leads to formation of bigger grains and more uniform film, resulting in reduction of trap density and better vertical charge transport, yielding a final PCE of 22.0 % in tandem with OPVs [15]. Intensive research has been also done on improving the quality of the interface between the perovskite and charge transport layers, since defects are likely concentrated at the material interface and lead to severe carrier recombination. Brinkmann *et al.* [10] have implemented passivation strategies, by modification of the perovskite surface with phenethylammonium iodide (PEAI), which promotes the formation of a 2D perovskite capping layer. The insertion of PEAI leads to notable lowering of the perovskite valence band maximum, which avoids photogenerated holes from reaching the electron extraction layer (EEL) and thereby facilitates selective extraction of electrons. As a result, a PSC/OPV tandem device with a remarkable PCE of 23.1 % and excellent stability of more than 1000 hours can be achieved [10]. Chen *et al.* [11] applied benzylphosphonic acid(BPA) for passivating the interface between NiOx and $Cs_{0.25}FA_{0.75}Pb(I_{0.6}Br_{0.4})_3$. Their tandem devices exhibited improved band alignment, reduced non-radiative recombination and better operational stability, yielding a PCE of 23.6 %.

Device architecture also plays an important role in determining the performance of PSC/OPV tandem devices. The PSCs can be fabricated in the n-i-p regular structure or inverted p-i-n structure. Notably, a compelling correlation has been observed based on the analysis of existing scientific literature. The inverted p-i-n structure is commonly employed for inorganic-organic hybrid PSC in tandem structure with the OPV subcell. The adoption of the inverted structure has demonstrated enhanced device stability and PCE in hybrid single-junction PSCs. These improvements are attributed to the increase of charge carrier lifetime and the diffusion coefficient as well as enhancement of charge collection efficiency, resulting in an enhanced performance of PSCs [37]. Meanwhile, owing to non-availability of corrosive additives in the inverted device configuration and hydrophobicity of the PCBM layer, the device stability can be enhanced in the ambient atmosphere [37]. In contrast, an n-i-p structure is almost exclusively used for all-inorganic Cs based PSCs in the tandem devices, with the electron transport layer deposited on the substrate followed by the perovskite active layer and hole transport layer. This is not surprising, as high efficiency Cs-based all-inorganic PSCs have not been demonstrated in p-i-n structure for single-junction devices.

3. Other Constituent layers

The interconnecting layer (ICL) is one of the key components in the development of high-performance 2-terminal monolithic devices. Requirements for good interconnecting layers include high transparency, minimum optical losses, good interface for efficient recombination of holes and electrons generated from WBG and NBG sub cells. Moreover, minimum current leakage is obligatory for the interconnecting layer. The ICL in PSC/OPV tandem devices consists of carrier recombination layer (CRL) sandwiched between hole transporting layer (HTL) and electron transporting layer (ETL) (or known as EEL and hole extraction layer (HEL), respectively). The commonly used CRLs are thermally evaporated ultra-thin metal like Ag, Au or transparent conductive oxides such as indium tin oxide (ITO). The CRL has to be used for tandem devices. Otherwise, devices exhibit poor performance due to the Schottky barrier between the HTL and ETL. By inserting a CRL between the top and bottom cells, this barrier can be reduced, allowing charge carriers to recombine efficiently. Additionally, the CRL can improve the ohmic contact between the top and bottom cells, enhancing the overall performance of the tandem solar cell. Brinkmann *et al.* [10] developed an interconnection between sub cells using InO_x layer prepared by low-temperature atomic layer deposition (ALD) (80 °C) with a precise control of layer thickness. This approach has led to notable improvements in the *J-V* characteristics of PSC/OPV tandem solar cells and rendering the $SnO_x/InO_x/MoO_x$ ohmic contact. Chen *et al.* have found that 4-nm-thick sputtered indium zinc oxide (IZO) ICL can also reduce optical losses as well as V_{oc} loss in the WBG PSC by interfacial passivation. These improvements cause a promising PCE of 23.60 % (certified 22.95 %) for PSC/OPV tandem devices [11]. Xu *et al.* [14] demonstrated metal-free ICL PSC/OPV tandem devices based on ALD grown SnO_2 and solution-processed PEDOT: PSS. The use of metal-free ICL can simplify the device fabrication process and thus reduce the cost for future manufacturing. This type of tandem device has good outdoor stability and exhibits a PCE of 17.6 % [14].

For PSC/OPV tandem devices, the most commonly used HTLs are MoO_x, Poly-TPD, 2PACz/MeO-2PACz while the ETLs are ZnO, SnO_2, NiO_x, PCBM. A double layer of ETL based on ZnO and SnO_2 is also popularly used in all-inorganic Cs based PSCs [19], [20], and [27]. The thickness of the ETLs in the ICLs plays a critical role in affecting the performance of monolithic PSC/OPV devices. A very thin ETL can lead to inefficient electron extraction from the WBG sub cell to the ICLs, while too thick ETL can also inhibit electron extraction and compromise tandem device performance [12].

The deposition techniques used for constructing tandem devices should be also carefully considered. The processing techniques directly impact the film thickness, interface quality, crystal structures, and morphology of the layers, consequently influencing the overall performance of the tandem devices. For 2T-PSC/OPV tandem devices, the majority of the functional layers are prepared by the spin-coating processes, which is one of the most economical approaches among the thin-film deposition techniques such as e-beam deposition, magnetron sputtering and thermal evaporation etc. The spin-coating technique is straightforward to use under laboratory conditions while it is not suitable for large-scale production, and it can lead to composition inhomogeneities in the layers of the devices. Other thin-film deposition methods, which are compatible with solution-processed films, have been attempted during monolithic tandem device fabrication. One of them is thermal evaporation, it is a common deposition technique in PSC/OPV tandem device for preparing the thin metal layers of Ag or Au as the CRL and the electrode, as well as depositing ETL and HTL materials such as C_{60}/BCP/TPBi and MoO_3 respectively [10], [11], [16], [20-22]. The thermal evaporation has its advantages such as capability to control the film thickness, low contamination risks as well as high compatibility with the solution-processed layers. It is also capable of preparing flexible samples at low substrate temperature [38-39].

Apart from ICL, the external coating such as anti-reflective layers on the devices can reduce the light reflection at the surface of incoming light. Brickmann *et al.* [10] and Troughton *et al.* [14] utilized MgF_2 as an antireflective layer. The direct effect of the antireflective coating on the efficiency of the tandem device was not discussed in these studies while it is predicted that approximately 1 % reduction in reflection between the range of 400-800 nm can be achieved [40]. Given the promising results, the use of anti-reflective layers such as MgF_2 or other materials is an interesting area for research to further enhance the light harvesting ability of tandem PVs.

4. Perspective on future development PSC/OPV Tandem

The scientific community has made notable advances in developing efficient tandem PV devices using emerging materials. Among these, PSC/Si-based tandem devices have been extensively researched and currently hold the highest PCE record of 33.2 % [2]. However, the mismatch in the lifetime of perovskite and silicon remains a significant obstacle for practical applications. Furthermore, silicon cannot be used for the construction of semi-transparent tandem solar cells, which limits the potential applications of PSC/Si tandem devices. The certified record PCE of 23.1 % has been achieved for PSC/OPV tandem devices [10], demonstrating the potential of using it as one of the future alternatives to conventional photovoltaics. The tunability of bandgap and transparency in PSCs and OPVs is a distinct advantage, ensuring a wide range of future innovative applications, such as smart PV windows with thermochromic functions and colorful PVs for aesthetically oriented projects and other building-integrated PVs (BIPVs). However, despite the promising progress made in the development of PSC/OPV tandem devices, there is still a scarcity of research focused on their engineering for practical applications. This is because some limitations still need to be overcome before PSC/OPV tandem devices can become commercially viable.

One of the major challenges is large-scale manufacturing of tandem structure which involves more fabrication steps compared to their corresponding single junction devices. Currently, the common reported active areas for PSC/OPV tandem devices range from 1.74 mm² [10] to 1 cm² [13, 23], which are still limited for practical applications that require large-scale deployment, and thus it is very crucial to develop effective thin-film deposition techniques for fabrication of larger devices while maintaining high efficiency is necessary. For example, blade coating, slot-die coating, spray coating and inkjet printing should be intensively investigated for preparing PSCs and OPVs while the vacuum deposition process such as thermal evaporation can be used for preparing uniform large-scale perovskite thin films and metal oxides carrier transport layers. Despite the ability to tune the bandgaps of both the perovskite and organic absorber layer in OPVs, the majority of reported PSC/OPV 2-terminal monolithic tandem devices are based on a front wide-bandgap perovskite subcell and a rear narrow- bandgap OPV subcell. This is due to the higher processing temperature required for PSCs, such as the conversion of precursors and perovskite crystallization, compared to the fabrication processes of OPVs. The high processing temperature steps of perovskite can cause the degradation of the underlying OPV properties. The possible solution is to develop pre-synthesized inks of perovskite, such as quantum dot dispersion solutions [41], and carrier transport materials, and investigate effective deposition of these inks to avoid high-temperature processing steps of PSC fabrication. Only with a maturity of diverse thin-film deposition methods can the freedom of PSC/OPV combination be largely enhanced, and this is crucial for advancing the practical viability of this type of tandem PVs. On the other hand, the development of transparent electrodes and the use of flexible substrates are also essential for enabling more diverse applications of PSC/OPV tandem PV in the future. The materials such as graphene with good flexibility [42] and sputtered ITO with desirable transparency and conductivity have been widely demonstrated for application in OPV and PSCs [43-44]. However, the compatibility of large-scale processing and the potential for induced damages on the thin films underneath during high-energy deposition processes should be carefully addressed.

Device stability and lifetimes of PSC/OPV tandem PVs are other critical considerations in the development of their practical applications. Both PSCs and OPVs are sensitive to environmental factors such as moisture, heat, and UV exposure etc., which can lead to device degradation over time. Researchers are exploring various strategies to improve device stability, including encapsulation techniques [45], material and device engineering [30-31], [46] etc. and achieved significant advancements. Despite the fact that PSC and OPV still have shorter lifetimes compared to Si at the current stage, their low material and fabrication costs can provide a cost-effective replacement option. Therefore, they could potentially be employed in certain applications that do not require a long product lifespan, which could be beneficial to indoor applications and portable electronic devices, where cost-effective replacement options are desirable and product lifetimes are relatively short due to frequent upgrades. This will be an incremental step towards future practical application of PSC/OPV PVs. With continued research and development, this technology will become a commercially viable option for a broader range of applications.

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Перовскитті органикалық тандемдік күн элементтерінің дамуындағы соңғы мәліметтер мен тенденциялар

Бірнеше өтпелі фотоэлектр элементтердің (ФЭ) пайда болуы бірөтпелі күн элементтерінің Шокли-Квиссер шегін еңсеруге мүмкіндік береді. Соңғы уақытта перовскитті күн батареялары (ПКЭ) және органикалық фотоэлектрлік элементтер (ОФЭ) — бұл энергияны түрлендіру тиімділігінің (EPE) өте жылдам өсуіне байланысты фотоэлектрлік қоғамдастықтың кең назарын аударған жаңа буынның ең перспективалы екі фотоэлектрлік элементі. ПКЭ және ОФЭ-ді тандемдік құрылымдарда біріктіру абсорбциялық жолақтарды реттеу және мөлдірлікті жақсарту үшін жарық сіңіруші қабаттардың шектік аймақтарын өзгерту мүмкіндігі сияқты көптеген артықшылықтарды ұсынады. Жұқа қабат технологиясын қолдану құрылғылардың жеңілдігі мен икемділігін қамтамасыз етеді, бұл әсіресе кейбір заманауи қолданбалар үшін тиімді. Сонымен қатар, ПКЭ және ОФЭ екеуі де арзан бағасымен ерекшеленеді, бұл оларды болашақта кең ауқымды коммерцияландыру үшін тартымды етеді. Бұл артықшылықтар ПКЭ/ОФЭ тандемдік құрылғыларын дәстүрлі кремний негізіндегі ФЭ-ден тыс қолданбалар үшін перспективалы етеді. Бұл шолуда ПКЭ/ОФЭ тандемдік ФЭ-ның соңғы жетістіктері туралы өзекті ақпарат берілген. ПКЭ және ОФЭ субэлементтерінің қасиеттеріне, сондай-ақ олардың функционалды қабаттарына әсер ететін материалдарды жасаудың заманауи технологиялары мен инженерлік әзірлемелері талқыланған. Сондай-ақ, тандемдік фотоэлектрлік элементтердің осы түрінің дамуына бағыт беру үшін перспективалық нұсқаулық беріледі. Мақала зерттеушілерге бұл технологияны одан әрі дамыту және оның жаңартылатын энергия көздері саласындағы толық әлеуетін ашу үшін бағыт жоспарын ұсына отырып, ПКЭ/ОФЭ тандемдік ФЭ әзірлеу туралы түсінік береді.

Кілт сөздер: перовскиттер, органикалық фотоэлектриктер, тандем күн элементтері, құрылғы инженериясы.

Т. Серикова, Г. Бижанова, Ханлин Ху, Энни Нг

Последние данные и тенденции в развитии перовскитных органических тандемных солнечных элементов

Появление фотоэлектрических элементов (ФЭ) с несколькими переходами предоставило возможность преодолеть предел Шокли–Квиссера однопереходных солнечных элементов. В последнее время перовскитные солнечные элементы (ПСЭ) и органические фотоэлектрические элементы (ОФЭ) являются двумя наиболее перспективными фотоэлектрическими элементами нового поколения, которые привлекли широкое внимание фотоэлектрического сообщества благодаря их исключительно быстрому росту эффективности преобразования энергии (ЭПЭ). Объединение ПСЭ и ОФЭ в тандемных конструкциях дает множество преимуществ, таких как возможность настройки ширины запрещенной зоны поглотителей для регулирования полос поглощения и повышения прозрачности. Использование тонкопленочной технологии обеспечивает легкость и гибкость устройств, что особенно выгодно для некоторых применений. Кроме того, как ПСЭ, так и ОФЭ отличаются низкой стоимостью, что делает их привлекательными их для широкомасштабной коммерциализации в ближайшем будущем. Эти преимущества сделают тандемные устройства ПСЭ/ОФЭ перспективными для применений, выходящих за рамки традиционных ФЭ на основе кремния. В настоящем обзоре представлена актуальная информация о недавнем прогрессе тандемных ФЭ ПСЭ/ОФЭ. Обсуждены современные технологии изготовления и инженерные разработки материалов, влияющие на свойства субэлементов ПСЭ и ОФЭ, а также их функциональных слоев. Кроме того, дано перспективное руководство для направления будущего развития этого типа тандемных фотоэлектрических элементов. Настоящая статья дает представление о разработке тандемных ФЭ ПСЭ/ОФЭ, предоставляя исследователям план действий для дальнейшего развития этой технологии и раскрытия ее полного потенциала в области возобновляемых источников энергии.

Ключевые слова: перовскиты, органические фотоэлектрики, тандемные солнечные элементы, разработка устройств.