SHORT COMMUNICATION

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Inorganic perovskite/organic tandem solar cells with efficiency over 20%

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The rapid development of perovskite solar cells is beyond our imagination. The power conversion efficiency (PCE) of organic–inorganic hybrid perovskite solar cells has reached 25.5% (https://www.nrel.gov/pv/cell-efficiency.html). However, the unsatisfactory stability of hybrid perovskites is an obstacle for their commercialization, which results from the volatile and hygroscopic organic cations[1]. Recently, inorganic perovskites ($CsPbI_xBr_{3-x}$) are receiving more and more attention due to their good thermal stability[2]. Among them, $CsPbI_2Br$ is the most popular material for solar cells due to the appropriate Goldschmidt tolerance factor[3, 4]. But the photovoltaic performance of $CsPbI_2Br$ is much lower than many other perovskites due to its large bandgap (1.92 eV), which means it can only absorb the light below 650 nm, leading to low photocurrent[5]. Narrow-bandgap organic solar cells have much broader photoresponse than $CsPbI_2Br$ solar cells, but their relatively high energy loss and insufficient absorption for the short-wavelength light limit their PCE. Making tandem solar cells is an effective approach to break the efficiency limit.

Tandem solar cells based on perovskite/silicon[6], perovskite/Cu(In,Ga)Se_2 (CIGS)[7], perovskite/perovskite[8], and perovskite/organic[9] structures have been reported. Compared with perovskite/silicon and perovskite/CIGS tandem cells, perovskite/perovskite and perovskite/organic tandem cells have the advantage of low-temperature solution-processing. Perovskite/perovskite tandem solar cells with tin-containing perovskite cell as the rear cell offered a certified PCE of 24.2%, which is the highest PCE for perovskite/perovskite tandem solar cells so far[8]. But it is hard to obtain long-term stable tin-containing perovskites because Sn^{2+} is easy to be oxidized to Sn^{4+}. Recently, the fast development of organic solar cells with narrow-bandgap non-fullerene acceptors provides new choice for tandem cells[10]. After we reported the first two-terminal inorganic perovskite/organic tandem solar cells[5], the PCE of inorganic perovskite/organic tandem cells increased from 15.04% to 18.38%[11] *via* using better perovskite and organic subcells. But the PCEs are still much lower than the other perovskite-based tandem cells, which may be due to the lack of high-performance organic solar cells. Recently, we developed a new polymer donor D18 for organic solar cells, demonstrating a PCE of 18.56%, which is the highest PCE for organic solar cells to date[12, 13]. Introducing high-performance D18:Y6 cells into inorganic perovskite/organic tandem cells may further improve the PCE.

In this communication, we report an inorganic perovskite/organic tandem solar cell with a $CsPbI_2Br$ front cell and a D18:Y6 rear cell. The energy loss in the tandem cell was reduced by using SnO_2/ZnO electron-transport layer. We optimized the photocurrent matching of the two subcells by adjusting the thickness of $CsPbI_2Br$ layer and D18:Y6 layer. The tandem solar cells delivered a record PCE of 20.18%.

The active layer for the organic solar cells consists of a wide-bandgap polymer D18 (E_g^{opt} = 1.98 eV)[12] and a narrow-bandgap non-fullerene small molecule Y6 (E_g^{opt} = 1.81 eV)[14] (Fig. 1(a)). The D18:Y6 blend film presents an absorption peak at 810 nm and an absorption onset at 931 nm (Fig. 1(b)). The $CsPbI_2Br$ film shows an absorption onset at 650 nm and a shoulder peak at 628 nm. $CsPbI_2Br$ can compensate the absorption of D18:Y6 below 628 nm, leading to better light harvesting when applied in a tandem structure.

The $CsPbI_2Br$ cells and D18:Y6 cells (Fig. S1) were fabricated to find the suitable charge-transport materials and preparation conditions. Unlike the frequently used SnO_2, SnO_2/ZnO was used as electron-transport layer. The $CsPbI_2Br$ cells with SnO_2/ZnO electron-transport layer show enhanced open-circuit voltage (V_{oc}) and short-circuit current density (J_{sc}) (Fig. S2), suggesting reduced energy loss in the cells. The best $CsPbI_2Br$ cell and D18:Y6 cell gave PCEs of 14.48% and 15.23%, respectively (Fig. S3 and Table S1). The $CsPbI_2Br$ cells show external quantum efficiency (EQE) above 80% at 390–640 nm (Fig. S3(b)). The high EQE before 640 nm and high V_{oc} for $CsPbI_2Br$ cell make it suitable to be the front cell in a tandem cell. The D18:Y6 cell presents broad photoresponse up to 970 nm, leading to high integrated current density (23.43 mA cm^{-2}) than the $CsPbI_2Br$ cell (14.56 mA cm^{-2}). The high EQE of D18:Y6 cell beyond 640 nm makes it suitable to be the rear cell.

The tandem cells were fabricated with the structure shown in Fig. S4(a). The SnO_2/ZnO layer, $CsPbI_2Br$ layer, PTAA/MoO_3/Au/ZnO nanoparticles (ZnO NPs) interconnecting layer (ICL), D18:Y6 layer, and MoO_3/Ag layer can be seen clearly in the cross-section scanning electron microscopy (SEM) image of the tandem cell (Fig. 1(c)). The thicknesses for $CsPbI_2Br$ layer and D18:Y6 layer are 260 and 120 nm, respectively. The thin Au layer (1 nm) between the two subcells is important for reducing the charge-transport barrier, thus improving device performance[5]. The charge-transport layers form a

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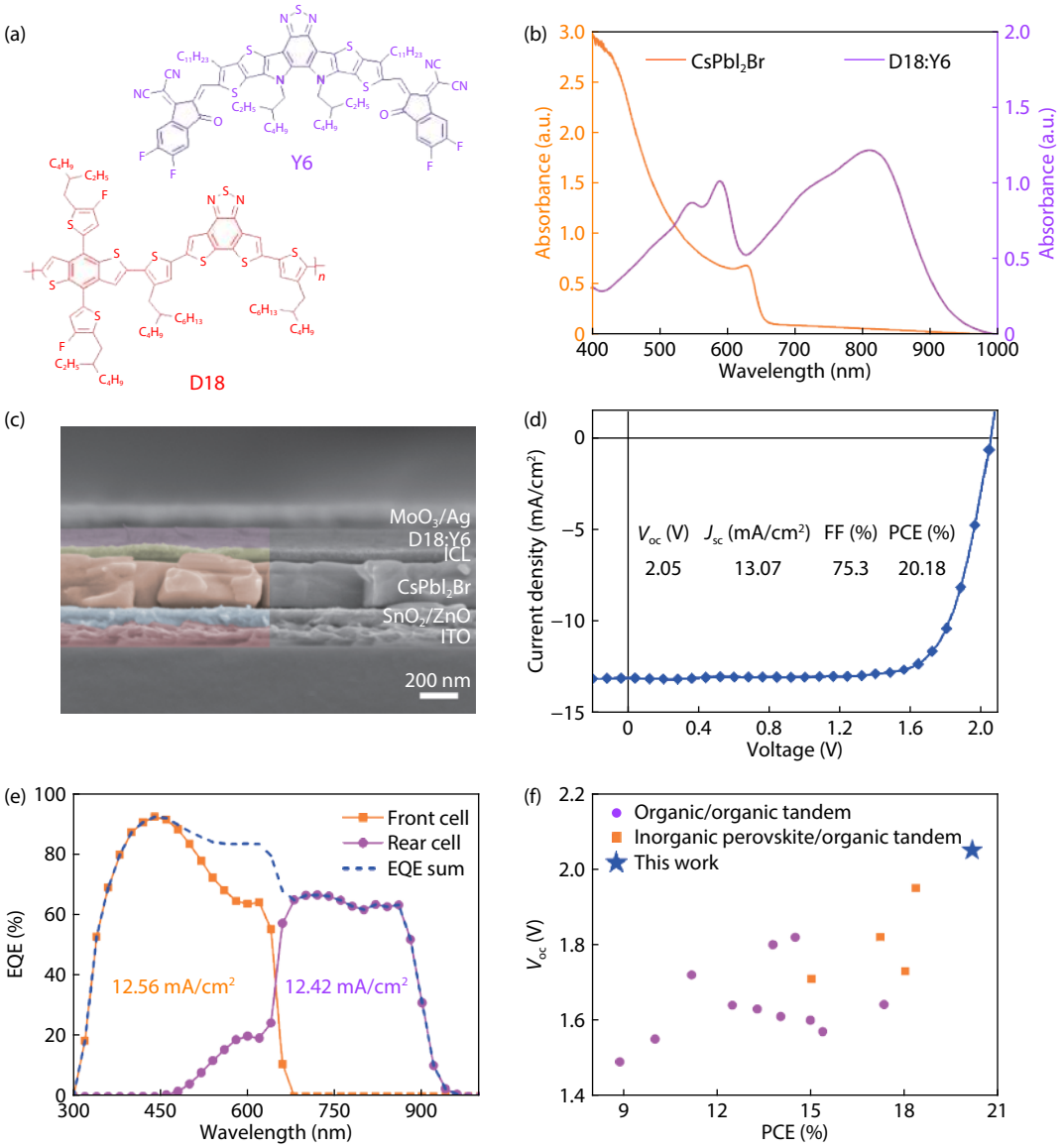

Fig. 1. (Color online) (a) Structures for D18 and Y6. (b) Absorption spectra for $CsPbI_2Br$ and D18:Y6 (1 : 1.6) films. (c) The cross-section SEM image for the tandem cell. (d) *J*–*V* curve for the best tandem cell. (e) EQE spectra for the front subcell and rear subcell. (f) Summary of the V_{oc} and PCE for the tandem cell in this work and the reported organic/organic and inorganic/organic tandem cells.

good energy cascade with $CsPbI_2Br$ layer and D18:Y6 layer (Fig. S4(b)), benefitting charge transport.

The photocurrent from a tandem cell is limited by the subcell with the lower photocurrent. So, the photoresponse of the front cell and the rear cell should be well adjusted to obtain a balanced photocurrent while keeping the optimal overall light harvesting. To this end, the thicknesses for $CsPbI_2Br$ layer and D18:Y6 layer were optimized (Figs. S5 and S6, Table S2 and S3). Finally, a best PCE of 20.18% is obtained (Fig. 1(d)). The *J*–*V* curves show slight hysteresis under forward and reverse scan (Fig. S7), which is caused by the hysteresis of the inorganic perovskite cell. A steady-state PCE of 19.84% is obtained by measuring at the maximum power point (Fig. S8). The EQE spectra for the subcells were measured (Fig. 1(e)). The perovskite subcell and the organic ksubcell present integrated current density of 12.56 and 12.42 mA cm^{-2}, respectively, with only 1% mismatch.

The tandem solar cells present a V_{oc} of 2.05 V, which is close to the sum of that for each subcell (2.08 V), suggesting good ohmic contact between the subcells. Compared with the reported organic/organic[15–25] and inorganic perovskite/organic[5, 11, 26, 27] tandem cells, the tandem cells here show much higher V_{oc} (Fig. 1(f)), which is due to the high V_{oc} of $CsPbI_2Br$ cell. The PCE is also much higher than the reported results, because of the use of efficient D18:Y6 subcell, reduced energy loss, and balanced photocurrent of the subcells. The PCE remains above 19% when the light intensity varies from 9.80 to 111.76 mW cm^{-2} (Fig. S9), making the tandem cells suitable for outdoor applications with instable light intensity. The tandem cells maintain 96% of their initial PCEs after being stored in a N_2 glovebox for 30 days, suggesting good stability (Fig. S10).

In summary, inorganic perovskite/organic tandem solar cells with $CsPbI_2Br$ perovskite front cell and D18:Y6 organic rear cell were made. The V_{oc} of the tandem cells was enhanced by using SnO_2/ZnO electron-transport layer. The photocurrent of the tandem cells was optimized by fine-tuning the photocurrent of the subcells. The tandem solar cells deliver a

decent PCE of 20.18%. To the best of our knowledge, it is the highest PCE from inorganic perovskite/organic tandem solar cells. Further enhancement in device performance can be realized *via* optimizing the structure details of the tandem cells and applying high-performance innovative materials.

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Appendix A. Supplementary materials

Supplementary materials to this article can be found online at https://doi.org/10.1088/1674-4926/42/2/020501.

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