RESEARCH HIGHLIGHTS

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# Monolithic perovskite/silicon tandem solar cells offer an efficiency over 29%

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Organic–inorganic hybrid perovskite semiconductors possessing superior optoelectronic properties (e.g. long carrier diffusion lengths, high optical absorption coefficient, low exciton binding energy, and high defect tolerance) are attracting serious attention. The certified power conversion efficiency (PCE) for single-junction perovskite solar cells have exceeded 25%[1, 2]. As a very promising PCE-enhancement strategy, tandem structure made by stacking a perovskite cell on a market-dominant silicon cell can yield much higher PCEs beyond the Shockley-Queisser limit of single-junction devices without adding substantial cost[3]. To satisfy current-matching in tandem configuration, the top perovskite cell requires an ideal bandgap of ~1.7 eV rather than the ones (~1.5–1.6 eV) typically used for highly efficient single-junction perovskite devices given that the bottom silicon cell holds a bandgap of 1.12 eV[4]. Such wide-bandgap perovskites achieved through I/Br alloying usually suffer from photoinduced phase segregation and relatively low radiative efficiency, which inevitably result in large open-circuit voltage ($V_{oc}$) deficits[5, 6]. Several strategies like adjusting perovskite composition[7, 8], additive engineering[9, 10], and upper surface passivation[11, 12] have been utilized to stabilize these wide-bandgap perovskites and improve film quality to reduce $V_{oc}$ losses. The reported perovskite/silicon tandem devices suffer from low $V_{oc}$ (<1.9 V) and PCEs (⩽28%)[13]. There is still a large room for enhancing PCEs given that the predicted PCE limit is beyond 30% for this tandem technology[14].

Recently, Albrecht *et al.* demonstrated an effective strategy to improve $V_{oc}$ and PCE simultaneously and made perovskite/silicon tandem solar cells with a certified PCE of 29.15%[15]. To be specific, a self-assembled, methyl-substituted carbazole (Me-4PACz) monolayer was used as the hole-transport layer (Fig. 1(a)) instead of common PTAA. The chemical structures for PTAA and Me-4PACz is depicted in Figs. 1(b) and 1(c). The 1.68 eV perovskite film ($Cs_{0.05}(FA_{0.77}MA_{0.23})_{0.95}Pb(I_{0.77}Br_{0.23})_3$) deposited on ITO/PTAA exhibited an obvious phase segregation with a red shift of photoluminescence (PL) peak (Fig. 1(b)), whereas the emission peak position kept nearly stable on ITO/Me-4PACz (Fig. 1(c)). Me-4PACz monolayer not only minimized nonradiative interface recombination losses *via* surface passivation according to quasi-Fermi level splitting (QFLS) measurement, but also enabled ultrafast hole extraction evidenced by charge carrier lifetime analyses, thus leading to effective suppression of photoinduced phase segregation. The corresponding single-junction device with a structure of ITO/Me-4PACz/perovskite/LiF/$C_{60}$/$SnO_2$/Ag offered a $V_{oc}$ of 1.22 V and a PCE of 20.8%. Except the role of phase stabilization, the fast charge extraction paired with efficient surface passivation significantly reduces the diode ideality factor of Me-4PACz-based solar cells to 1.26 (1.55 for PTAA-based devices), accordingly contributing to a fill factor (FF) of 84%. Further FF analysis indicated the formation of a lossless interface when inserting Me-4PACz monolayer between ITO and perovskite (Fig. 1(d)).

Subsequently, such efficient single-junction cells were employed to construct monolithic perovskite/silicon tandem solar cells (Fig. 1(e)). The best tandem device with Me-4PACz under forward scan gave a $V_{oc}$ of 1.90 V, an FF of 79.52%, a short-circuit current density ($J_{sc}$) of 19.26 mA/cm$^2$, and a PCE of 29.05%, yielding a stabilized PCE of 29.15% at the maximum power output, which notably outperformed PTAA counterpart ( $V_{oc}$ = 1.85 V, FF = 75.61%, $J_{sc}$ = 19.19 mA/cm$^2$, PCE = 26.79%) (Fig. 1(f)). The impressive PCE of 29.15% is much higher than that for previous monolithic perovskite/silicon tandem devices[12, 13, 16, 17]. Strikingly, the tandem device with Me-4PACz delivered a maximum $V_{oc}$ of 1.92 V, where the contribution of the perovskite subcell reached 1.2 V in consideration of a $V_{oc}$ of ~715 mV from silicon subcell under the filtered illumination. Additionally, the efficiency of Me-4PACz-based tandem cells without encapsulation just degraded by 4.5% after 300 h operation in air. More encouragingly, fitting results indicated that a 32.4% PCE can be achieved from perovskite/silicon tandem cells.

The performance of monolithic perovskite/silicon tandem cells depends on not only perovskite absorber but also the important yet neglected charge-transport layers, which can affect perovskite phase stability, charge extraction, and interface recombination losses[18]. Despite breaking theoretical efficiency limit (29.1%) of silicon cells[19], tandem configuration may realize >32% PCE through further optimization. More efforts should focus on charge transport losses and current-matching conditions as well as $V_{oc}$ deficit.

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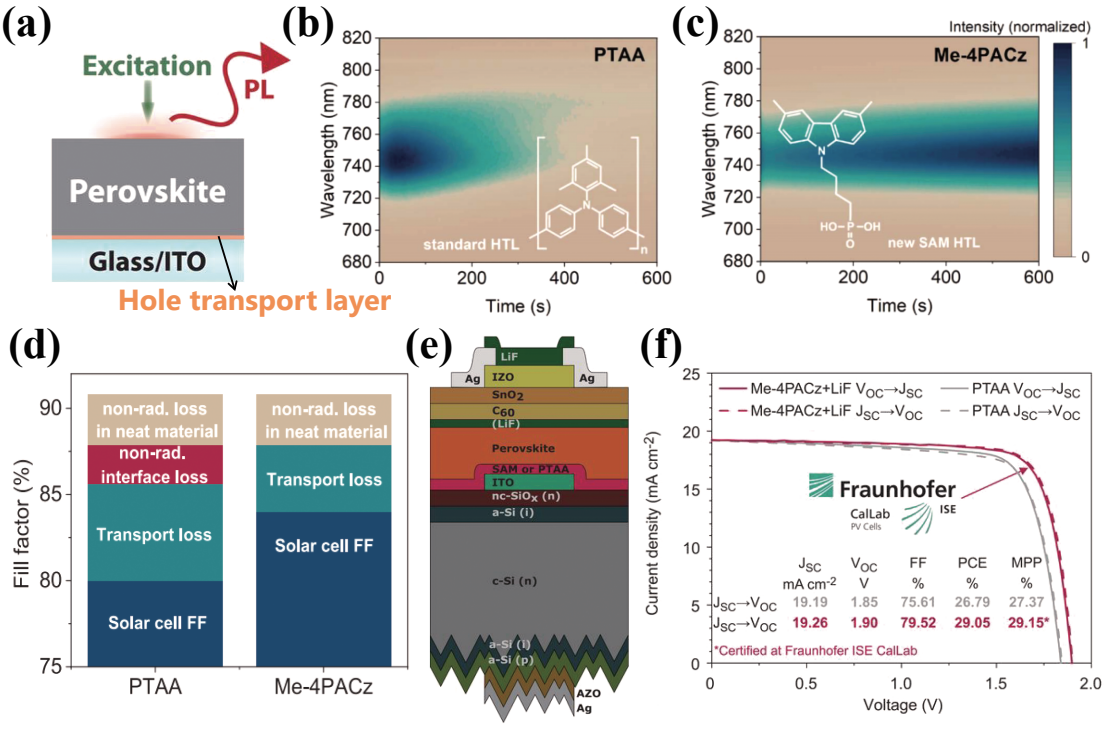


Fig. 1. (Color online) (a) Schematic diagram for PL measurement of perovskite films deposited on ITO substrates with different hole-transport layers. Time-dependent PL spectra (inset: molecular structure) for 1.68 eV perovskite films coated on (b) ITO/PTAA or (c) ITO/Me-4PACz substrates. (d) FF and its losses in PTAA and Me-4PACz based single-junction solar cells. (e) Schematic illustration for monolithic perovskite/silicon tandem device. (f) *J*–*V* curves for champion tandem solar cells with PTAA (in-house measurement) or Me-4PACz (certified at Fraunhofer). Reproduced with permission[15], Copyright 2020, American Association for the Advancement of Science.

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