# Facile Modifications on Buried Interfaces Enable Recent Breakthroughs in Efficiencies of Flexible Perovskite Solar Cells

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## Short Communication

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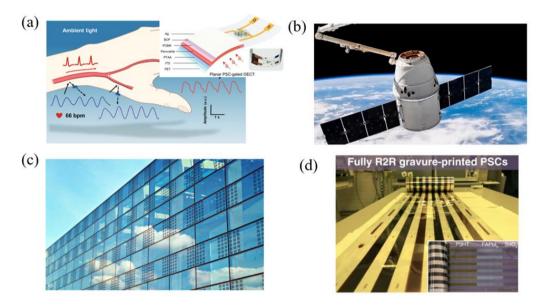
# DESCRIPTION

Flexible Perovskite Solar Cells (FPSCs) have unique merits of lightweight, easy deformation, and high flexibility, making them highly appealing to wearable electronics, smart integrated buildings, and aerospace applications <sup>[1-2]</sup>. In addition, their compatibility with Roll To Roll (R2R) printing fabrication suggest a great potential in realizing the commercialization of perovskite photovoltaic technology (Figure 1).

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**Figure 1**. a) Illustration of PSC-gated OECTs for monitoring PPG signals from human body under ambient light <sup>[3]</sup>. b) A photo of an artificial satellite with solar modules working in the space <sup>[4]</sup>. c) A photo of a building integrated with solar cells <sup>[5]</sup>. d) A photograph of R2R processed FPSCs <sup>[6]</sup>.



#### INTRODUCTION

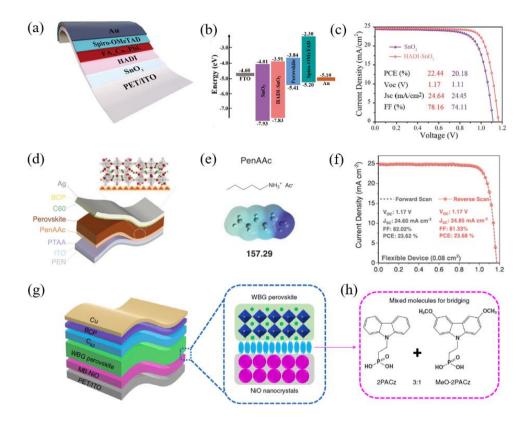
In 2013, the first FPSC with a PCE of 2.62% was reported by Kumar, et al. where a device was deposited on a Polyethylene Terephthalate (PET) substrate <sup>[7]</sup>. After that, numerous efforts have been made to improve the efficiency and mechanical stability of FPSCs, and great achievements have been obtained. At the initial stage of FPSCs, the explorations on flexible substrates, transparent electrode, low-temperature processed charge transporting layers, and high-quality perovskite films make great efforts in achieving high-performance FPSCs with the PCE over 21%, while the efficiency of FPSCs still lags behind the rigid counterpart <sup>[1-8]</sup>.

As well-known, the interface quality plays a critical role in perovskite solar cells with layered structure. It is more important in FPSCs since more severe mechanical stress would be externally applied during the fabrication and utilization (twisting, bending, and stretching)<sup>[9]</sup>. The external stress could lead to large strain or delamination at the interface, which results in serious non-radiative recombination and even prevents the charge transfer in FPSCs <sup>[10,11]</sup>.

A compact and band-aligned interface is thus highly desired to further improve the photovoltaic performance of FPSCs at the next stage where FPSCs can reach a comparable efficiency with rigid counterpart. Among the interfaces in the flexible device, the buried interface needs to pay more attention due to the weak adhesion resulted from the rough surface of flexible substrates and polycrystalline nature of perovskite films, which significantly limits the efficiency and mechanical stability of FPSCs (Figures 2a-2h).

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**Figure 2**. a-c) A device structure, schematic energy level alignment, and J-V cures of a FPSC with modified interface with HADI, respectively <sup>[12]</sup>. d) Schematic illustration of the device structure. e) The molecular structure of PenAAc and its electron density distribution. f) J-V curves of champion FPSC with PenAAc modification <sup>[13]</sup>. g) Device structure. h) Molecular structures of bridging molecules <sup>[14]</sup>.



In the last year, several works have been reported to focus on modifications of the buried interface making the breakthroughs in efficiency of FPSCs over 23%, which is comparable with rigid counterparts. In a work published by Yang, et al. a novel histamine diiodate (HADI) is employed to modify the buried SnO<sub>2</sub>/perovskite interface (Figures 2a-2c) <sup>[12].</sup> The HADI with multi-functional groups could passivate the buried perovskite surface, strengthen the adhesion between SnO<sub>2</sub> and perovskite layers and realign the band at the interface, which improved the efficiency of FPSCs to 22.44%. Gao, et al. reported a FPSC with a certified efficiency of 23.35% achieved by pentylammonium acetate (PenAAc) modifying the buried PTAA/perovskite interface (Figures 2d-2f) <sup>[13].</sup>

Through synthetic manipulation of anion and cation where the PenA<sup>+</sup> and Ac<sup>-</sup> have strong chemical binding with defects on perovskite films, a compact PTAA/perovskite interface is formed enabling highly efficient charge transfer. Li, et al. fabricated a high-performance flexible tandem solar cell by modifying the buried NiOx/perovskite interface in the subcell, which pushes the efficiency to 24.7% (certified 24.4%) <sup>[14]</sup>. In this work, two types of molecules, named 2PACz and MeO-2PACz, were mixed to modify the NiOx/perovskite interface in the wide-bandgap perovskite solar cell (Figures 2g and 2h). Due to the suppressed interfacial non-radiative recombination by efficient passivation, a wide-bandgap perovskite solar cells (WBG PSCs) based on a perovskite composition of FA<sub>0.8</sub>Cs<sub>0.2</sub>Pbl<sub>1.95</sub>Br<sub>1.05</sub> (bandgap of ~1.75 eV) achieved a PCE of 16.2%. A flexible tandem perovskite solar cells with a PCE of 24.7% was prepared by combining the WBG PSC with a Narrow-Bandgap (NBG) perovskite with a composition of FA<sub>0.7</sub>MA<sub>0.3</sub>Pb<sub>0.5</sub>Sn<sub>0.5</sub>I<sub>3</sub> and a bandgap of about 1.22 eV.

### CONCLUSION

It can be concluded from the above study of FPSCs with efficiencies of about 23% that the modification of buried interfaces makes great contribution to push the efficiency of FPSCs to a new level. In view of efficiency improvement, three critical factors about passivators at the buried interface need to be considered.

- The binding strength with CTL and perovskite layers should be strong enough to maintain the interaction under external stress.
- > The introduction of passivator/modifier should enable better matched band alignment at the interface.
- > The modifier could efficiently passivate the defects on CTL and perovskite films.

The above factors could guarantee the efficient charge transfer at the buried interface, which is highly favourable to the high efficiency of FPSCs. Mechanical stability is another requirement for the practical application of FPSCs. In FPSCs with a layered structure, strained interfaces or detached interfaces are usually formed due to the difference in Young's modulus of each layer under bending operation. The buried interface is more susceptible to bending operation due to the weak adhesion resulted from the rough surface of flexible substrates and polycrystalline nature of perovskite films. The strain release or the adhesion enhancement at the buried interface are thus highly necessary to improve the mechanical stability of FPSCs, while the corresponding investigations are insufficient in the above studies of FPSCs, additional mechanical properties, such as Young's modulus, adhesion properties, should be considered. Moreover, novel interfaces at the interface, such as van der Waals interaction, are deserved to being explored to release the stress, which could significantly enhance the mechanical stability. Therefore, the modification of interfaces, especially buried one, is of great importance to fabricate FPSCs with high efficiency and mechanical flexibility.

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