Monolayer-modified PTAA facilitates the preparation of high-efficiency wide-bandgap perovskite solar cells and all-perovskite tandem cells with efficiencies exceeding 25%

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1. Introduction

Although the efficiency of the organic-inorganic hybrid perovskite solar cell (PSCs) has reached 25.7%, it still has not surpassed silicon-based solar cells[1]. At the same time, poor stability also makes the competitiveness of single-layer PSCs not obvious compared with the silicon solar cells. Since 2015, narrow bandgap PSCs have been widely reported due to their better light absorption[2]. However, wide bandgap perovskites should not be neglected as they are an important component for fabricating tandem PSCs[3]. Here, we demonstrate a series of monolayer molecules with different alkyl chain lengths as interfacial modifiers to modify the PTAA and perovskite layer for improving the optoelectronic properties of PSCs by improving the quality of perovskite films and increasing the transport and extraction of interfacial carriers. The target device achieves a PCE of 16.57%, which is one of the highest PCE of the WBG-PSCs. Besides, the all-perovskite tandem solar cells with the WBG-Pb-perovskite solar cells and the narrow bandgap Sn/Pb perovskite solar cells gave the high PCE of 25.24%.

2. Device Fabrication:

1 mg of monolayer molecules was dissolved in 1 ml DMF and this solution was spin-coated on the PTAA at a speed of 5000 rpm for the 30s. Then the substrate was put on the hot stage at a temperature of 100 °C for 5 minutes. The surface was washed with 50 μL DMF by using the spinning coater with a speed of 5000 rpm for 30s. The perovskite film was deposited using the two-step spin-coating method at 2000 rpm for 10 seconds and 6000 rpm for the 40s, with 150 μL of CB antisolvent dripped over perovskite films for 30 seconds before the program was terminated, and the film was subsequently annealed at 100 °C for 10 minutes.

3. Result and discussion

Figure 1a shows the typical *J-V* curves of the control and target PSCs. The champion control device with a PCE of 14.46% (a short circuit current (J_{SC}) of 17.15 mA/cm^2, an open-circuit voltage (V_{OC}) of 1.117 V, and a fill factor (FF) of 75.53%), while the devices based on 3,3PrPACz, 4,3BuPACz, and 6,3HePACz modification got the highest PCE of 15.49% (a J_{SC} of 17.59 mA/cm^2, a V_{OC} of 1.141 V and an FF of 77.20%), 16.57% (a J_{SC} of 17.98 mA/cm^2, a V_{OC} of 1.175 V and an FF of 78.44%), and 15.29% (a J_{SC} of 17.53 mA/cm^2, a V_{OC} of 1.146 V and an FF of 76.15%), respectively. Figure 1b exhibits the corresponding incident photon-to-electron conversion efficiency (IPCE) spectra of the best-performing control device and PTAA/monolayer molecules devices, with the integrated current densities of 16.99 mA/cm^2 for the control device, 17.34 mA/cm^2 for the 3,3PrPACz-modified device, 17.72 mA/cm^2 for the 4,3BuPACz-modified device, and 17.00 mA/cm^2 for the 6,3HePACz-modified device, in good agreement with the J-V characterization. Finally, all-perovskite tandem solar cells were fabricated to further improve the solar energy utilization of the cells. The structure of the solar cell was exhibited in Figure 1c. Figure 1d presents the *J-V* curves of the cells modified by 4,3BuPACz. A high PCE of 24.21% was obtained (a J_{SC} of 16.210 mA/cm^2, a V_{OC} of 1.793 V, and an FF of 83.3%) with forwarding scan and 23.92% with reverse scan (a J_{SC} of 16.028 mA/cm^2, a V_{OC} of 1.757 V and an FF of 84.9%), which shows the negligible hysteresis.

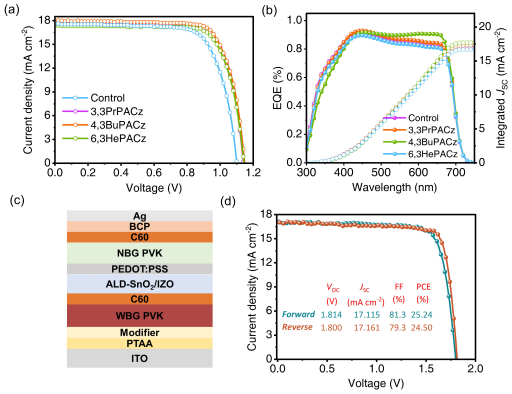

Figure 1 a) *J-V* curves and (b) IPCE curves of the champion devices without and with monolayer molecules modification. (c) Schematic structure of all-perovskite tandem solar cell employed in this work. (d) *J-V* curves of the champion all perovskite tandem solar cells with 4,3BuPACz modification.

4. Conclusion

In conclusion, several new monolayer molecules have successfully been used to modify the interface between PTAA and WBG perovskite layer. The experimental results show that after the introduction of monolayer molecules, the quality of the film has significantly improved. In addition, the stress of the perovskite film is released due to the change of the substrate. On the other hand, the introduction of monolayer molecules reduces the interfacial non-radiative recombination of the device. Finally, 4,3BuPACz-based PSCs achieved a high PCE of 16.57%, meanwhile, the target tandem solar cells also get a PCE of 25.24%.

Reference

[1] PARK J., KIM J., YUN H. S., PAIK M. J., NOH E., MUN H. J., KIM M. G., SHIN T. J. and SEOK S. I., Controlled growth of perovskite layers with volatile alkylammonium chlorides [J]. *Nature*, 2023, 616(7958):724-730.

[2] KAPIL G., BESSHO T., SANEHIRA Y., SAHAMIR S. R., CHEN M. M., BARANWAL A. K., LIU D., SONO Y. Y., HIROTANI D., NOMURA D., NISHIMURA K., KAMARUDIN M. A., SHEN Q., SEGAWA H. and HAYASE S., Tin-lead perovskite solar cells fabricated on hole selective monolayers [J]. *ACS Energy Letters*, 2022, 7(3):966-974.

[3] XIAO K., LIN Y. H., ZHANG M., OLIVER R. D. J., WANG X., LIU Z., LUO X., LI J., LAI D., LUO H., LIN R., XU J., HOU Y., SNAITH H. J. and TAN H., Scalable processing for realizing 21.7%-efficient all-perovskite tandem solar modules [J]. *Science*, 2022, 376(6594):762-767.