## Efficient Planar Perovskite Solar Cells via Interfacial Engineering with Anatase or Brookite TiO<sub>2</sub> Single-Crystalline Nanoparticles

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The electron transport layer (ETL)/perovskite interface engineering plays a promising strategy to produce highly efficient planar heterojunction (PHJ) perovskite solar cells (PSCs). The deep trap states of ETL compact TiO<sub>2</sub> surface facilitates a large leakage current and recombination of electron-hole pairs. Interfacial modification of TiO<sub>2</sub> by a novel one-step hydrothermal processed single-crystalline anatase (AT) and/or brookite (BK) TiO<sub>2</sub> NPs with average diameter sizes about 6 and 30 nm, respectively, is applied as an ETL bilayer to enhance the separation of charges and suppress the recombination rate. Herein, a novel TiO<sub>2</sub>/AT or BK TiO<sub>2</sub> NPs bilayer was introduced by comprising spray pyrolysis (SP) deposition and spin-coating (SC) technique, respectively, in PHJ PSCs. A SP-TiO<sub>2</sub>/AT and BK SC-TiO<sub>2</sub> NPs bilayer-based PSCs are facilitated more efficient electron transport, charge extraction, and low interfacial recombination, and thus leads champion efficiencies up to 17.05% and 16.74%, respectively, by a significant decrease of J-V hysteresis, presenting almost 12% enhancement compared to TiO<sub>2</sub> single layer-based PSCs with PCE of 13.33%. The PHJ PSCs exhibited a spectral response that extended from the visible to the near-infrared region with a broad, flat absorption peak of intensity 80%-85% at approximately 380-750 nm. The higher IPCE value of the device with a bilayer in the visible-to-near-infrared wavelength region than those of the other devices suggests that the bilayer layer collect electrons more efficiently at the perovskite/TiO<sub>2</sub> edge because it successfully lowers the interfacial energy barrier. The outstanding performance of the bilayer ETL based device is attributed to the enhanced the content of photogenerated charge carrier sites and reduced the agglomeration and trapping at the SP-TiO<sub>2</sub> interface, as evident by PL analysis.



**Fig.** (a) *J-V* characteristics of devices with and without bilayer and (b) IPCE spectra of devices with and without bilayer. *Keywords*: PHJ PSCs, Interface Engineering; SP-TiO<sub>2</sub>/BK SC-TiO<sub>2</sub> NPs, SP-TiO<sub>2</sub>/AT SC-TiO<sub>2</sub> NPs. Acknowledgement: This study was supported in part by Research and Study Project of Tokai University General Research Organization.