Strategy for Improved Photoconversion Efficiency in Thin Photoelectrode Films by Controlling π-Spacer Dihedral Angle

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Benzo[c][1,2,5]thiadiazole (BT) has been used in dye-sensitized solar cells (DSCs) for its light-harvesting abilities. However, as a strongly electron deficient unit, BT causes rapid back electron transfer (BET), which in turn lowers the photoconversion efficiency (PCE) of devices. Herein, we report a powerful strategy for retarding BET by controlling both the photoelectrode thickness and π-spacer dihedral angle. To achieve this, we introduced planar (**BT-T**) or twisted π-spacers (**BT-P**, **BT-MP**, and **BT-HT**) between BT units and anchoring groups and used different photoelectrode thicknesses between 1.8 and 10 μm (Figure 1). Computational and experimental results show that twisted π-spacers were more efficient at retarding BET than the planar π-spacer. However, BET was found to be less important than expected, and light harvesting efficiency (LHE) played a critical role as the thickness of the photoelectrode decreased because charge collection efficiency was enhanced. The planar dye **BT-T** obtained the highest LHE, this value remained unusually high even in 1.8 μm photoelectrodes. As a result, **BT-T** gave a PCE of 6.5% (J_{sc} = 13.56 mA/cm^2, V_{oc} = 0.67 V, and FF = 0.72) in thin 1.8 μm photoelectrodes with 3.5 μm scattering layers, which represented a roughly 40% enhancement compared to the PCE in 10 μm photoelectrodes (4.76%). Overall, these results provide a novel approach to achieving ultrathin and highly efficient flexible DSCs.

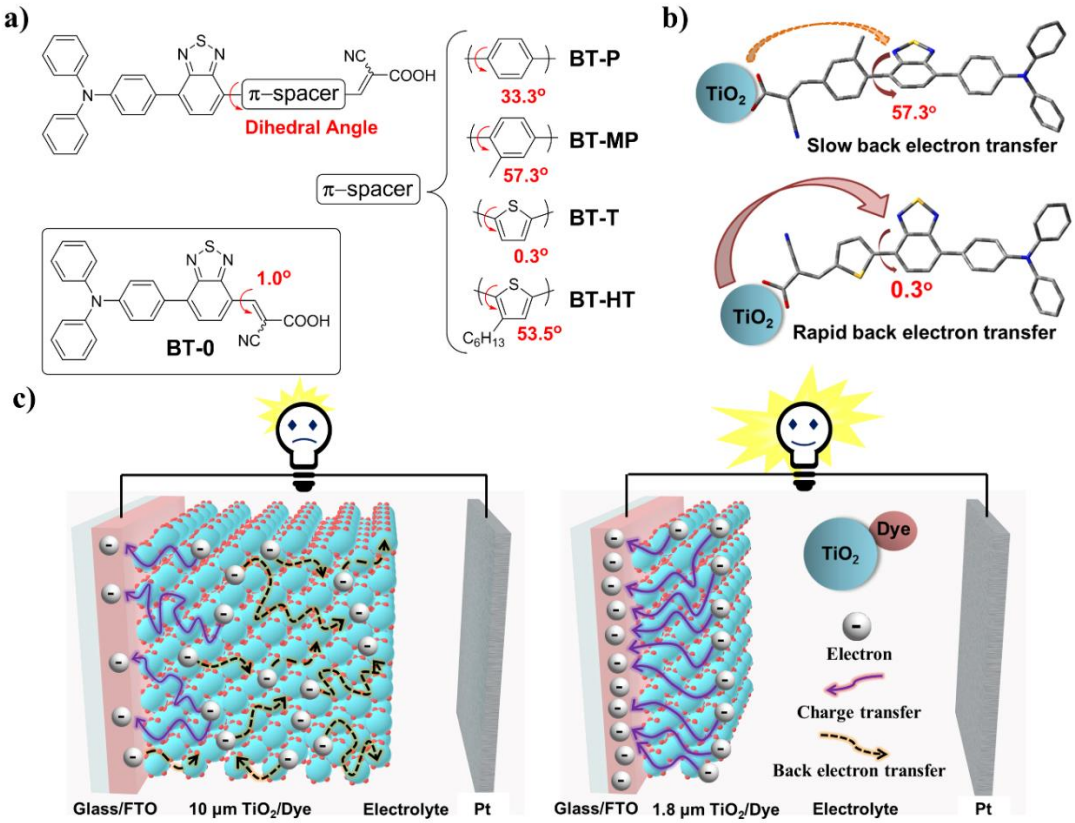

Figure 1. a) Chemical structures of BT-series showing dihedral angles between BT and π-spacers. Schematic figures: b) back electron transfer with different π-spacers, and c) enhancement of charge collection efficiency in a thin 1.8 μm photoelectrode compared with a thicker 10 μm photoelectrode.