Molecular and Microstructural Factors Influencing Charge Carrier Recombination at Donor-acceptor Interfaces for Organic Photovoltaics

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Electronic processes at the organic hetero-interface between electron donating and electron accepting molecules determine the photocurrent, photovoltage and ultimately, the power conversion efficiency of organic solar cells. While devices with incident-photon-to-extracted-charge conversion yields of over 85%, and absorbed photon-to-extracted-charge conversion yields of 90-100% have been achieved, the difference between the optical gap of main absorber and open-circuit voltage (V_{oc}) is much larger than for inorganic and perovskite based solar cells. The main improvements of the V_{oc} of organic solar cells have so far been made by tailoring donor-acceptor interfacial energetics, taking advantage of well-known principles of molecular design. Nevertheless, for most material systems we consistently find a large, but virtually constant difference (~0.6 eV) between eV_{oc} and the energy of the intermolecular charge transfer (CT) state, E_{CT}. Added to this, electron transfer losses are very often larger than 0.1 eV, resulting in overall voltage losses larger than 0.7 eV. We present experimental evidence that the E_{CT}-eV_{oc} difference can be reduced by reducing the physical interfacial area available for free charge carrier recombination. We quantify this by analyzing the strength of the interfacial CT state absorption and emission signal at photon energies below the optical gap of the neat materials. We further discuss the influence of measurable molecular properties, such as the electronic coupling, molecular reorganization as well as non-radiative recombination pathways on free carrier recombination and V_{oc}. By controlling molecular and microstructural factors, we achieve a high V_{oc} of 1.18 V for a device with an incident-photon-to-extracted-charge conversion yield of 79% at 1.77 eV (700 nm), corresponding to the lowest voltage losses for organic solar cells achieved to date.