Multiscale Simulations on Charge Transport in Covalent Organic Frameworks

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Covalent organic frameworks (COFs) are potential candidates for applications in optoelectronic devices and solar cells due to their ability to transport charge through their aromatic molecular units. This study[1] investigated the charge transport (CT) in COFs with a multiscale simulations technique, which combines classical molecular dynamics simulations, quantum chemical calculations, and carrier dynamics simulations. To efficiently estimate the charge transfer integrals from quantum chemical calculations, we developed the FMO-DFTB/LCMO approach by combining the fragment molecular orbital (FMO), density-functional tight-binding (DFTB), and linear-combination of molecular orbitals (LCMO) methods. We observed that the thermal motions of COFs cause substantial fluctuations of the transfer integrals. We evaluated the charge carrier diffusion using Ehrenfest dynamics and kinetic Monte Carlo simulations, including the fluctuations of the transfer integrals. Both simulation approaches provided similar high carrier mobilities of ca. 2 $cm^2 V^{-1} s^{-1}$ without using adjustable CT parameters.

[1] Kitoh-Nishioka, H. et al. J. Phys. Chem. C **2017**, *121*, 17712-17726.