

Photo-induced Charge Transfer of Fullerene and Non-fullerene Conjugated Polymer Blends via *ab initio* Excited-state Dynamics

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ABSTRACT

Organic conjugated polymers (CPs) are promising candidates for organic photovoltaic (OPV) devices due to their unique tunable mechanical and optoelectronic performance. Over the last decade, optoelectronic properties of narrow bandgap CPs as a blend with acceptors units are largely optimized which leads to noticeable progress in OPV technology. However, despite the active research which is being taken place to enhance OPV's, their power conversion efficiency is still lower than their organic counterparts (i.e., silicon). One of the most important parameters in the PV devices is the rate of photo-induced charge transfer (CT) at the interface of the CP and acceptor unit. It would be largely beneficial to computationally examine the ability of a different molecular configuration serving as an efficient CT interface before any synthesis process to narrow down the search list of possible high-performance bulk heterojunctions (BHJ). In this study, we employ *ab initio* molecular dynamics, to explore photo-induced CT of diketopyrrolopyrrole (DPP) based polymer as a blend with non-fullerene (i.e., ITIC) and fullerene (i.e., PCBM) acceptor units. The results of charge carrier dynamics induced by selected photoexcitation show that hole density redistribution in space is much faster than electron relaxation. We track the relaxation rates of charge carriers over time, where the derivative of the difference between the rate of electron and hole can qualitatively represents the current density at zero voltage. This can be utilized to characterize the CT performance of CPs blended with different acceptor units. Relaxation rate results indicate that CPs blend with ITIC offers a better PV performance, illustrating the potential of our approach to explore BHJ electronic performance for OPV devices and narrowing down the list of potential candidates.

Keywords: Conjugated polymers; Density functional theory; *ab initio* excited-state; Organic photovoltaics.