Mixed ionic-electronic transport in crystalline P3HT with explicit dopants and additives: *ab-initio* & machine learning force fields molecular dynamics

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Here, we present *ab-initio* molecular dynamics (AIMD) and Machine Learning Force Fields molecular dynamics (MLFFMD) calculations on the mixed ionic-electronic transport for crystalline poly(3-hexylthiophene) (P3HT) using explicit lithium bis(trifluoromethanesulfonyl) imide (LiTFSI) dopant and dimethoxyethane (DME) additive. Up to now, very few works have been reported using AIMD calculations dealing with mixed ionic-electronic transport in crystalline P3HT, including explicit dopants and additives[1, 2]. Our ionic and electronic conductivities estimations show good correlation with experimental reports of similar mixed ionic-electronic conductors. The explicit atomistic treatment of all components in the calculations allowed us to reveal some interesting behavior due to the presence of dopants on the most relevant parameters associated to mixed ionic-electronic transport in a wide temperature range. For the electronic transport, our transfer integral (J) and reorientation energies (λ) values showed an increment respect to typical unexplicit-doped calculations. Furthermore, we also introduce the role of the explicit dopant on the inter-chain, intra-chain, "effective" doping and charge-transfer complex bonding distances and their associated static and dynamic disorder effects on the electronic transport. All these properties have been properly recovered with the use of the state-of-the-art tools, such as MLFFMD as implemented in VASP 6.3[3-6], in order to increase the number of explicit atoms, thus reducing the memory requirements and speeding up the molecular dynamics simulations.

References

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