Excited-State Calculation for large size C60/Pentacene interface Based on the Fragment Molecular Orbital Method

(P) Md. Khorshed Alam[1], Takatoshi Fujita[1], Takeo Hoshi[2]
[1]Institute for Molecular Science, Okazaki, Aichi 444-0865, Japan
[2]Department of Applied Mathematics and Physics, Tottori University, Japan
E-mail: khorshed@ims.ac.jp

The electronic structure at organic-organic interfaces is of great importance in the performance of various organic electronic devices. In organic photovoltaic (OPV) devices charge-transfer (CT) states at the interface between electron donor (D) and electron acceptor (A) materials play a major role in both exciton-dissociation and charge-recombination processes. Pentacene and C_{60} are archetypal donor and acceptor materials, respectively, due to their high mobilities for both charge and exciton [1]. Zheng et al. reported that increasing the number of molecules in pentacene/C_{60} complexes until $3C_{60}$/7 pentacene [2] results in some lowering of CT state energy. Thus, a large system of D/A clusters is expected to lead to more reliable explanation of the electronic processes at the pentacene-C_{60} interface. However, forecasting electronic excited states of large systems with rational accuracy is still a challenging concern in quantum chemistry technique.

We applied the fragment molecular orbital (FMO) method to calculate excited states in large molecular systems. In this work, we investigate the CT states in large C_{60}/pentacene complexes containing twenty pentacene molecules and twenty C_{60} molecules. These complexes are extracted from a bilayer architecture modeled by molecular dynamics simulation. We have analyzed CT states in terms of electron(e)-hole(h) separation and delocalization of electron or hole wave function. We have found that in the energy region lower than pentacene absorption, CT states are localized with their small e-h separations, while CT states are delocalized with larger e-h separation in higher-energy region. The calculated energy (1.1 eV) of the lowest CT states derived from our large complexes are in very good agreement with the experimental reported values [3]. We will discuss detail about the charge recombination rate k and its parameters in the Marcus equation, i.e., the reorganization energy and the transfer integral H_{ab} in the conference.

References:

1. Qin et al. *Phys. Status Solidi A*, 208 (2011) 1967.
2. Zheng et al. *ACS Appl. Mater. Interfaces* 9 (2017)18095.
3. Brigeman et al. *Adv. Energy Mater.* 6 (2016) 1601001.