

Transporting and transferring electrons across perovskite-acceptor interfaces

(¹Graduate School of Environmental Science, Hokkaido University, ²University of Rostock, ³University of Hyogo, ⁴Kyoto University) ○Sachith Bhagyashree Mahesha,¹ Takuya Okamoto,¹ Ghimire Sushant,² Tomokazu Umeyama,³ Yuta Takano,¹ Hiroshi Imahori,⁴ Vasudevanpillai Biju^{1*}

Keywords: halide perovskite; nanocrystals; interfacial electron transfer; fullerene

Halide perovskite nanocrystals (PNCs) become attractive due to their high photoluminescence (PL) quantum yields, tunable bandgap, and straightforward synthesis.¹ As a result, these materials become superior light absorbers in perovskite solar cells (PSCs) and the certified power conversion efficiency of a PSC exceeds 25%. Recently, photoinduced electron transfer (PET) studies in PNC-based electron donor-acceptor (D-A) systems receive great momentum, which is owing to the general interest to optimize the perovskite solar cell technology.² Radiative and nonradiative carrier losses adversely affect the energy conversion efficiency in thick perovskite films.³ Both the PL lifetime and transient absorption measurements in the presence of electron- and hole-accepting materials reveal the high charge mobility in perovskite films. However, the distance- or thickness-dependent recombination dynamics of photogenerated excitons/charge carriers at the perovskite-electron acceptor interfaces are yet to be completely understood.

We reveal the extent of diffusion, the degree of radiative loss, and the rate of diffusion-controlled interfacial electron transfer in heterojunction films of cesium or formamidinium lead bromide (Cs/FAPbBr₃) PNCs and C₆₀ or TiO₂. Self-assembled PNC films prepared from FAPbBr₃ show a longer PL lifetime than a solution, showing a long-range carrier migration or efficient carrier-trapping de-trapping cycles. To examine the nature of PET from PNCs, we perform PL studies of PNC solutions in the absence or presence of C₆₀. The acceptor quenches the PL intensity but not the lifetime in a solution, revealing a static electron transfer. Conversely, the distance-dependent electron transfer in films changes from dynamic to static by moving the pump-probe towards the donor-acceptor interface.⁴ While radiative recombination dominates the electron transfer at 800 μm or farther, the acceptors quantitatively scavenge the photogenerated carriers within 100 μm.

In summary, the adsorption of C₆₀ molecules onto the hydrophobic-caped surface of a PNC leads to static and quantitative PL quenching by electron transfer. Also, the electron transfer efficiency is appreciably high for excitons generated within 800 μm of a D-A interface. The migrating or trapped photogenerated charge carriers in the D-A films enable diffusion-controlled interfacial electron transfer.

1) S. Ghimire, et al. *ACS Energy Lett.* 4 (2019) 133. 2) L. Chouhan, et al. *Chem. Soc. Rev.* 49 (2020) 2869. 3) V. C. Nair, et al. *Angew. Chem. Int. Ed.* 56 (2017) 1214. 4) B. M. Sachith et al. *J. Phys. Chem. Lett.* 12 (2021), 8644.