



Characterization of photo-induced electron and hole transfer in a porphyrin based ambipolar organic molecule with cascade energy levels

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ABSTRACT

A porphyrin based ambipolar organic molecule consisting of styrene based triphenylamine derivative (**MTPA**) as electron donor, s-triazine group (**TRC**) as electron acceptor 1 and metal-free tetraphenyl porphyrin (**HTPP**) as electron acceptor 2 was synthesized and characterized using computational methods and electrochemical and spectroscopic measurements. The kinetics analysis indicates that the photo-induced charge-separated states, **MTPA**⁺-**TRC-HTPP**⁻, were generated by sequential electron transfers from **MTPA** to **TRC** then to **HTPP** and/or a direct hole transfer from **HTPP** to **MTPA**. In toluene, the charge-separated states were formed with 54% through electron transfer and 46% through hole transfer once **MTPA** moiety was excited. However, in dichloromethane, they were formed with 75% through electron transfer and 25% through hole transfer. Furthermore, more charge-separated species were generated in dichloromethane than in toluene.

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1. Introduction

Understanding the photophysical processes and rational design of organic materials are imperative for any technological applications associated with organic sensitizers. For instance, organic solar cells have been developed rapidly as an inexpensive alternative to large-area Si-based solar modules because of their potential for low-cost and high-throughput production [1–5]. In these solar

cells, light is absorbed by organic sensitizers and generates electron-hole pairs denoted as excitons. Then, these excitons dissociate to generate charge-separated species. For a solar cell to be efficient, the photophysical process from exciton generation to charge separation must be efficient and the reverse process, i.e. charge recombination, must be limited.

It has been shown that photo-induced electron (hole) transfer processes can convert light into charges and form charge-separated states effectively in donor-acceptor (D-A) systems [6]. Particularly, the electron excited to the lowest unoccupied molecular orbital (LUMO) level of the donor moiety (L_D) can be transferred to the lower-lying LUMO level of the acceptor moiety (L_A), with the hole remaining in the highest occupied molecular orbital (HOMO) level of the donor moiety. Alternatively, the same charge-separated state can be achieved when the electron in acceptor moiety is photo-excited, following a hole transfer from the HOMO level of the acceptor moiety (H_A) to the HOMO level of the donor moiety (H_D). Not surprisingly, researchers turn their attentions to the

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development of ambipolar organic molecules, in which both electron and hole transfers can take place and therefore the charge-separated states can be formed more effectively [7–13].

Porphyrim derivatives with structural similarity to natural light-harvesting chromophores in the photosynthetic reaction centers have been developed [14–31]. An efficiency up to 13.0% has been shown recently with a D-A structured diphenylamine modified porphyrin dye in dye-sensitized solar cells (DSSC) by Grätzel and coworkers [14]. As the electronic donor, metalloporphyrins have lower oxidation potentials than those of the corresponding metal-free porphyrins, which are used extensively in photo-induced

electron transfer of organic solar cells or DSSC and obtained high efficiency (>10%) [19–25]. On the other hand, protonated porphyrins and metalloporphyrins with electron-withdrawing substituents can act as electron acceptors in some photo-induced electron transfer systems [25–31]. Though the electronic transport properties of porphyrin derivatives that acted as either electron donor or acceptor have been widely reported, characterization of contributions from photo-induced electron and hole transfers in porphyrin based ambipolar molecules has not been discussed much.

Our previous work has shown that triazine was a typical

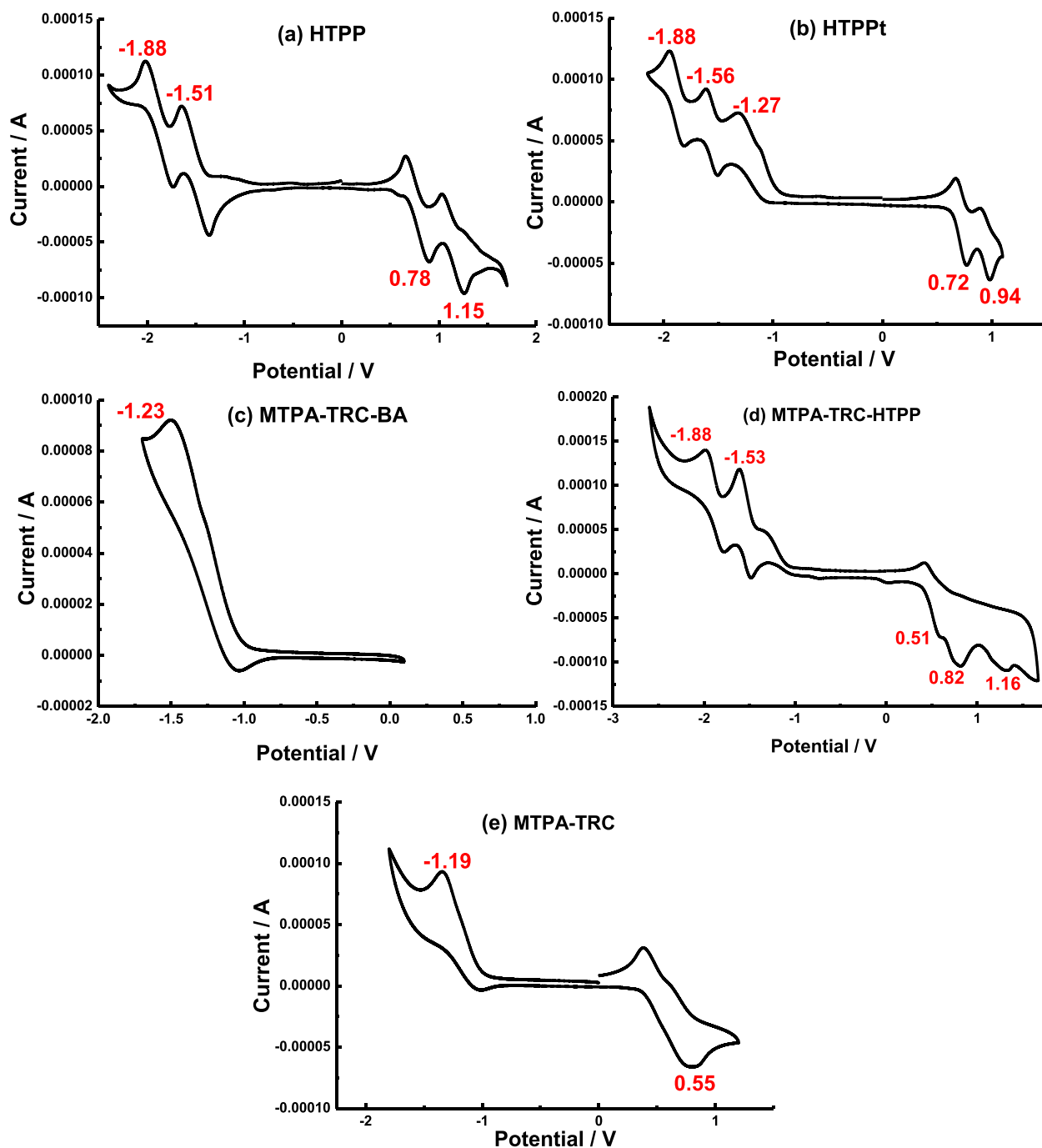


Fig. 1. Electrochemical curves of HTPP (a), HTPPt (b), MTPA-TRC-BA (c) and MTPA-TRC-HTPP (d) in dichloromethane vs Ag/Ag⁺. The concentration is 5×10^{-3} mol L⁻¹. The data of MTPA-TRC-BA (c) and MTPA-TRC (e) were taken from our previous work [34,36].

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