

Optimization of C₆₀-exTTF Compositions; Control Over Short- and Long-range Processes

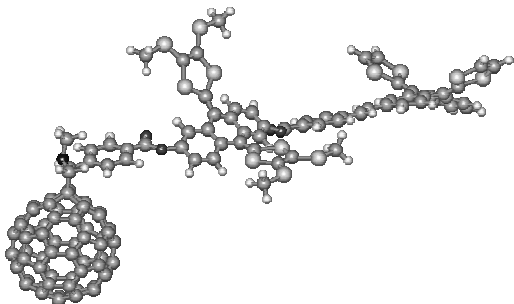
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The continuing quest for new donor-acceptor ensembles is driven by several goals: *i*) faster and more efficient charge separation, *ii*) slower charge recombination, *iii*) minimizing the loss of excited state energy and *iv*) exploring simpler systems with fewer components.

Recent reports indicate that aromatization of an oxidized donor moiety can also exert a notable impact on the improvement of light-induced charge-separation.¹ An illustration is given in a case where a tetrathiafulvalene (TTF) donor has been attached in close proximity to C₆₀.² In the resulting C₆₀-TTF dyads, charge-separated radical ion pairs are formed, whose lifetimes are in the range of nanoseconds.

Further advances in the stabilization concept were based on the use of π -extended tetrathiafulvalenes (exTTF).³ We have previously reported the synthesis of different C₆₀-exTTF dyads.⁴ Pico- and nanosecond transient absorption measurements reveal that the instantaneously formed fullerene singlet excited state transforms rapidly into the charge-separated radical pair. Remarkably, the lifetimes of the charge-separated states in C₆₀-exTTF are in the range of several hundreds of nanoseconds.

In this communication we apply the concept of gain of aromaticity and planarity upon oxidation of the exTTF donor to a newly designed series of different exTTF-containing dyads and triads. Details on the synthetic, electrochemical and photophysical work will be presented, highlighting the noteworthy impact that the chemical spacer [C₆₀-exTTF], or a second exTTF unit [C₆₀-exTTF₁-exTTF₂], exert on the improvement of light-induced charge separation. The lifetimes for intramolecular charge separation, determined in triads [C₆₀-exTTF₁-exTTF₂], are by far the longest values ever reported in molecular triads.



References

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