

Light-Harvesting and Charge Separation in a π -Conjugated Antenna Polymer Bound to TiO_2 Meyer

Scientific Achievement

Multifunctional polymer-based molecular assembly capable of light-harvesting, charge-separation, and charge storage

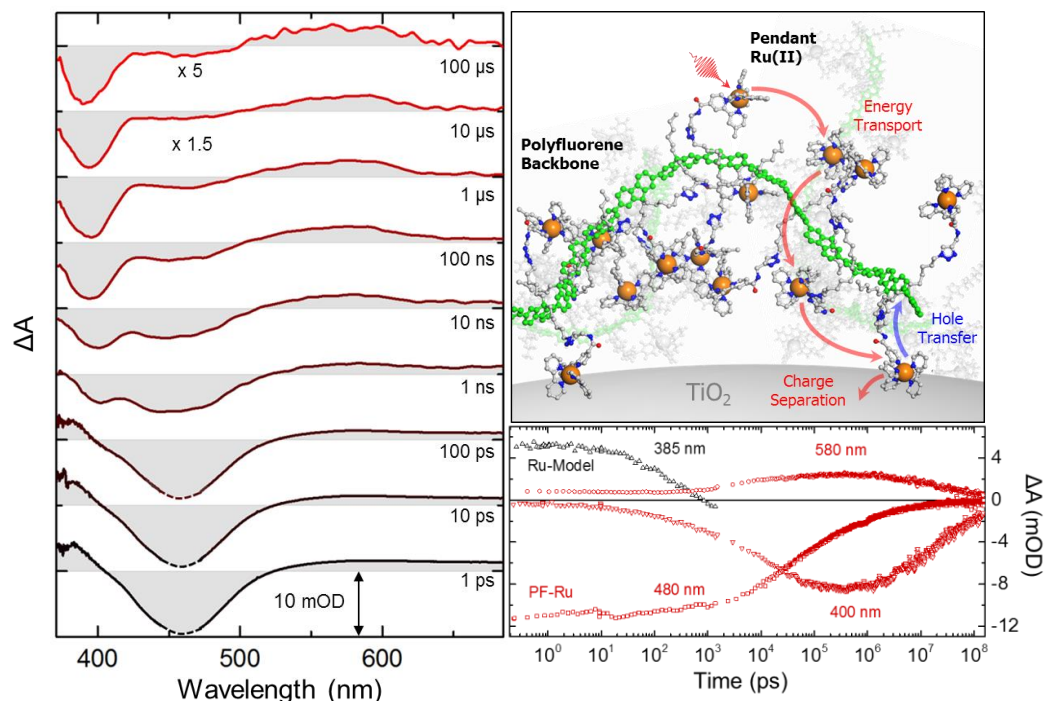
Significance and Impact

The design of molecular assemblies to enable effective transfer of excitation energy to charge-separation sites, while hole transfer away from the surface for use in multielectron catalysis for solar fuels production.

Research Details

- Functionalization of ~30% of the pendant Ru(II) sites enables site binding to metal oxide surface.
- Excitation of surface-bound sites results in prompt (ps) electron injection into TiO_2 nanoparticle, followed by ultrafast hole transfer to the backbone.
- Energy transport from unbound sites to surface sites for charge separation occurs on longer time scales.
- Charge separated state persists for hundreds of microseconds

Leem, G.; Morseth, Z.A.; Puodziukynaite, E.; Jiang, J.; Zhen, F.; Reynolds, J.R.; Papanikolas, J.M.; Schanze, K.S. "Light-harvesting and charge separation in a π -conjugated antenna polymer bound to TiO_2 ," J. Phys. Chem. C. 2014, ASAP. DOI: 10.1021/jp5113558



(Left) Transient absorption spectra of a PF-Ru-loaded TiO_2 film following excitation at 450 nm from 1 ps to 100 μs. The 10 and 100 μs spectra have been scaled for clarity, while regions containing scatter from the 450 nm pump have been marked with a dashed line.

(Upper right) Illustration of the initial dynamic processes occurring following the excitation of the Ru(II) pendants within the PF-Ru assembly attached to the surface of a TiO_2 nanoparticle.

(Lower right) Combined kinetics traces from 200 fs to 150 μs for the PF-Ru assembly (red) at 400, 480 and 580 nm and the Ru-Model (black) at 385 nm.

Work was performed at The University of North Carolina at Chapel Hill and The University of Florida