

fs-recombination in Fe-based solar cells limits the performance

Wednesday 15 June 2022 16:40 (10 minutes)

It has been widely recognized in the scientific community that scarce elements such as ruthenium are not ideal for sustainable technology. During the past 10 years, progress has been made in exploring first-row transition metals as replacements for scarce metals in many solar cell and photocatalysis applications.[1] Iron analogues to well-performing ruthenium-complexes were early found to not yield nearly the same solar cell performance, despite Ru and Fe being congeners.[2] Prior to our efforts, by means of ultrafast spectroscopy it was found that the relevant excited state deactivates in less than a ps, a timescale not accessible for most electron-transfer reactions.[3] Nowadays many examples of Fe-based complexes with ps-ns lifetimes have been demonstrated, mainly realized by the carbene-ligand.[4]

In the work described here, a set of push-pull Fe-carbene complexes have been characterized by means of time-resolved spectroscopy both in solution and after sensitization of titania nanoparticles.[5] In this way, parts of the dye-sensitized solar cell responsible for interfacial electron-transfer between the dye and the semiconductor are recreated, making it possible to study these processes. Creating rod-like push-pull complexes is a key strategy to facilitate charge separation in solar cells, and to mitigate charge recombination. Our results show that indeed electron injection into titania happen within 100 fs after excitation[5] and is highly efficient.[6] However, recombination leaves only ~10 % of the initially injected electrons for the timescale accessible to contribute to solar cell performance. Moreover, the recombination was found to take place mainly with a time constant of 100 fs and return the dye molecules to their excited state.[5] The discovery of the ultrafast recombination reaction identifies a key bottleneck that limits the development of Fe-based solar cells, something that would never have been found without employing ultrafast characterisation techniques.

[1] O.S. Wenger, Photoactive Complexes with Earth-Abundant Metals, *J. Am. Chem. Soc.* 140 (2018) 13522–13533. <https://doi.org/10.1021/jacs.8b08822>.

[2] S. Ferrere, B.A. Gregg, Photosensitization of TiO₂ by [FeII(2,2'-bipyridine-4,4'-dicarboxylic acid)₂(CN)₂]: Band Selective Electron Injection from Ultra-Short-Lived Excited States, *J. Am. Chem. Soc.* 120 (1998) 843–844. <https://doi.org/10.1021/ja973504e>

[3] J. E. Monat, J. K. McCusker, Femtosecond Excited-State Dynamics of an Iron(II) Polypyridyl Solar Cell Sensitizer Model, *J. Am. Chem. Soc.* 122 (2000) 4092–4097. <https://doi.org/10.1021/ja992436o>

[4] L. Lindh et al., Photophysics and Photochemistry of Iron Carbene Complexes for Solar Energy Conversion and Photocatalysis, *Catalysts* 10 (2020) 315. <https://doi.org/10.3390/catal10030315>

[5] L. Lindh et al., Dye-Sensitized Solar Cells based on Fe N-heterocyclic Carbene Photosensitizers with Improved Rod-like Push-Pull Functionality, *Chemical Science* 12 (2021) 16035–16053. <https://doi.org/10.1039/d1sc02963k>

[6] T. Harlang et al., Iron sensitizer converts light to electrons with 92% yield, *Nature Chemistry* 7 (2015) 883–889. <https://doi.org/10.1038/nchem.2365>

Authors: LINDH, Linnea (Chemical Physics/Theoretical Chemistry Lund University); Dr ROSEMANN, Nils W. (Chemical Physics Lund University); Prof. WÄRNMARK, Kenneth (CAS Lund University); Dr CHÁBERA, Pavel (Chemical Physics Lund University); Prof. PERSSON, Petter (Theoretical Chemistry Lund University); Prof. YARTSEV, Arkady (Chemical Physics Lund University)

Presenter: LINDH, Linnea (Chemical Physics/Theoretical Chemistry Lund University)

Session Classification: Sektionen för atom-, molekyl- och optisk fysik

Track Classification: Parallel session: atom-, molekyl- och optisk fysik