Theoretical Evidence of Solvent-Mediated Excited-State Dynamics in a Functionalized Iron Sensitizer

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The solvent-mediated excited-state dynamics of the COOH-functionalized Fe-carbene photosensitizer \([\text{Fe(bmicp)}_2]^{2+}\) (bmicp= 2,6-bis(3-methyl-imidazole-1-ylidine)-4-carboxy-pyridine) is studied by time-dependent density functional theory, as well as classical and quantum dynamics simulations. We demonstrate the crucial role of the polar acetonitrile solvent in stabilizing the metal-to-ligand charge transfer (MLCT) states of the investigated molecule using the conductor polarizable continuum model. This leads to dynamics that avoid sub-ps back electron transfer to the metal and an exceptionally long-lived 1MLCT state that does not undergo sub-ps 1MLCT → 3MLCT intersystem crossing as it is energetically isolated. We identify two components of the excited-state solvent reorganization processes: an initial rotation (∼300 fs) and diffusional dynamics within the local cage surrounding the rotated solvent molecule (∼2 ps). Finally, it is found that the relaxation of the solvent only slightly affects the excited-state population dynamics of \([\text{Fe(bmicp)}_2]^{2+}\).

General information
State: Accepted/In press
Organisations: Department of Chemistry, Neutrons and X-rays for Materials Physics, Department of Physics
Number of pages: 10
Publication date: 2019
Peer-reviewed: Yes

Publication Information
Journal: Journal of Physical Chemistry C
ISSN (Print): 1932-7447
Ratings:
BFI (2019): BFI-level 1
Web of Science (2019): Indexed yes
BFI (2018): BFI-level 1
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 1
Scopus rating (2017): CiteScore 4.58 SJR 2.135 SNIP 1.147
Web of Science (2017): Impact factor 4.484
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 4.48 SJR 1.964 SNIP 1.195
Web of Science (2016): Impact factor 4.536
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 1
Scopus rating (2015): CiteScore 4.68 SJR 1.886 SNIP 1.26
Web of Science (2015): Impact factor 4.509
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 1
Scopus rating (2014): CiteScore 5.08 SJR 2.032 SNIP 1.447
Web of Science (2014): Impact factor 4.772
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 1
Scopus rating (2013): CiteScore 5.14 SJR 2.143 SNIP 1.445
Web of Science (2013): Impact factor 4.835
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 1
Scopus rating (2012): CiteScore 4.98 SJR 2.529 SNIP 1.461
Web of Science (2012): Impact factor 4.814
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 1
Scopus rating (2011): CiteScore 4.92 SJR 2.339 SNIP 1.465