

Research Project

Photoactive Metal Complexes from Earth-Abundant Elements and Multi-Electron Photochemistry in Donor-Acceptor Compounds

Third-party funded project

Project title Photoactive Metal Complexes from Earth-Abundant Elements and Multi-Electron Photochemistry in Donor-Acceptor Compounds

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Organisation / Research unit

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Department

Project start 01.04.2018

Probable end 31.03.2022

Status Completed

Many of the most widely used photoactive complexes known to date are made from precious elements such as Ru, Ir, Pt or Au. Their replacement by more earth-abundant metals is of long-standing interest, and the first half of this proposed research project aims to explore the possibility of obtaining fundamentally new luminescent and photoredox-active complexes with long-lived excited-states. The plan is to synthesize and investigate a broad range of metal complexes made from earth-abundant metals in various oxidation states that until now have received either extremely limited or no attention at all in photophysical and photochemical contexts. Specifically, it is planned to explore: (i) homoleptic d6 MLCT luminophores and photoredox catalysts made from Cr(0), Mn(I), or Mo(0) with novel chelating isocyanide ligands, (ii) heteroleptic d10 MLCT emitters made from Ni(0) and a combination of chelating diphosphine and diisocyanide ligands, (iii) d-d emitters based on six-coordinate V(II) or Mn(IV) (d3) complexes, (iv) d0 LMCT luminophores based on Ti(IV) or Zr(IV) with chelating ligands made from pyrrolic and phenolic binding units. While the focus of this project is on obtaining fundamental insight into the basic photophysics and photochemistry of these new metal complexes, the development of such compounds made from earth-abundant elements is of significant interest in the contexts of lighting devices, solar cells, sensors, photoredox catalysis in organic chemistry, or for sensitization of reactions leading to the conversion of (solar) light into chemically stored energy, i. e., for so-called solar fuels. The second half of this proposed project aims at gaining fundamental insight into multi-photon, multi-electron transfer reactions going conceptually far beyond traditional work on photoinduced single electron transfer in donor-acceptor compounds. Artificial photosynthesis will have to rely on multi-electron conversions such as water splitting and CO₂ reduction, but currently many studies use sacrificial redox reagents to perform such reactions under irradiation with visible light. This approach will not permit sustainable light-to-chemical energy conversion, and therefore it is highly desirable to explore the basics of multi-photon, multi-electron transfer reactions, as well as the light-driven accumulation of multiple redox equivalents without sacrificial reagents. Toward this end, a series of very carefully designed donor-acceptor compounds will be synthesized and investigated by various photophysical methods. Electrochemical potential inversion, proton-coupled electron transfer (PCET), and redox relays will play an important role in the various sub-projects of this research endeavor.

Financed by

Swiss National Science Foundation (SNSF)

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