

Toward Computational Design of Iron-Based Chromophores for Solar Energy Conversion

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Photoactive transition metal complexes anchored to semiconductor surfaces play an important role as chromophores in artificial systems for solar energy conversion, such as dye-sensitized solar cells (DSSCs). Fe(II)-polypyridines share many properties with Ru(II)-polypyridines, which have been successfully used as photosensitizers in DSSCs. Visible light excitation in both types of compounds results in the population of photoactive metal-to-ligand charge transfer states (MLCT). The main obstacle to the utilization of Fe(II)-based compounds as photosensitizers is the short lifetime of the initially populated MLCT states due to their deactivation by ultrafast intersystem crossing events into photo-inactive metal centered (MC) ligand-field states. We employ density functional theory and quantum dynamics simulations to investigate how various modifications to the polypyridine ligands as well as semiconductor anchor groups influence the relative energies of the MC and MLCT states and relative rates of the intersystem crossing and interfacial electron transfer events. The results obtained lead to a better understanding of structure-property relationships in these complexes and have implications for development of photosensitizers based on first-row transition metals.