Fragment-Based Quantum Mechanical/Molecular Mechanical Simulations of Thermodynamic and Kinetic Process of the Ru2+-Ru3+ Self-Exchange Electron Transfer

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Abstract

A fragment-based fractional number of electrons (FNE) approach is developed to study entire electron transfer (ET) processes from the electron donor region to the acceptor region in the condensed phase. Both regions are described by the density-fragment interaction (DFI) method, while FNE as an efficient ET order parameter is applied to simulate the electron transfer process. In association with the QM/MM energy expression, the DFI-FNE method is demonstrated to describe ET processes robustly with the Ru2+-Ru3+ self-exchange ET as a proof-of-concept example. This method allows for systematic calculations of redox free energies, reorganization energies, electronic couplings and the absolute ET rate constants within the Marcus regime.

Keywords

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