Proton-Coupled Electron Transfer (PCET) with Bimetallic Systems

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Electron-transfer (ET) is one of the simplest chemical events, yet, it is a vital process for chemical reactivity. Proton-coupled electron transfer (PCET) is the concerted transfer of protons and electrons which is fundamental in chemical and biological systems and processes in electrochemistry, photosynthesis, and enzymatic activity. Similarly, mixed-valence (MV) systems are pervasive in nature and provide a platform for multiple electron transfers systems in nature. While both fields have been studied extensively, little work has been done on the intersection of the two. This project explores the elegant use of MV bimetallic systems through hydrogen bonded networks. A variety of compounds have been synthesized using Mo$_2$(DAniF)$_3$(O$_2$CCH$_3$) (where DAniF is the anion of N,N'-di-p-anisylformamidine) as the starting material, and further reacted with four different bisbidentate ligands (2,5-dihydroxy-1,4-benzoquinone, 3,4-dihydroxycyclobut-3-ene-1,2-dione, N,N'-dimethyloxamide, ethanedioic acid). Electrochemical and spectroscopic data show that the rate of ET in the hydrogen-bonded system is several orders of magnitude lower compared to the covalently bridged dimer, as predicted.

Figure 1 – PCET through multiple-bonded bimetallic systems