

109 - Investigation of axial ligand exchange as related to the immobilization of cobaloxime catalysts on a carbon surface

*Katarina Hauser, hauserkj@gmail.com, Christopher Eubanks, Matthew Freer, Dale Wheeler, Michael Hambourger. A.R. Smith
Department of Chemistry, Appalachian State University, Boone, NC 28608, United States*

The widespread implementation of renewable energy systems will require a means of energy storage. Towards this end, there is great interest in the development of active, cost-effective catalysts for water splitting via electrolysis or photolysis. Bis(glyoximate) cobalt complexes, cobaloximes, contain only Earth-abundant elements and are promising catalysts for the hydrogen evolution half-reaction. Surface immobilization of these complexes is likely to improve their stability while increasing catalyst retention in a flow through device. Axial pyridine ligation is well known in this class of compounds, and has received attention as a method of coupling cobaloxime catalysts to chromophores and nanoparticle surfaces. However, recent literature suggests that ligand exchange may interfere with the stable coupling of cobaloximes in photochemical systems. Herein we report on the stability of a cobaloxime-pyridine adduct under different solvent conditions. Cyclic voltammetry, UV-vis spectroscopy, elemental analysis, and steady-state fluorescence experiments are used to investigate ligand exchange in cobaloxime systems. Our results are consistent with non-photochemical exchange of the pyridyl ligand in the presence of acid or a coordinating solvent. Since these solvent conditions emulate those commonly used during catalytic turnover, the results suggest that a more robust method of cobaloxime immobilization is required.

Sunday, September 8, 2013 06:00 PM

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