CORE-CM SEMINAR Michigan State University

Charles McCrory University of Michigan

The Electrochemical Conversion of CO2 to CO with Molecular Co Catalysts: Modulating Activity and Selectivity by Controlling the Catalyst's Environment

One of the primary barriers to the large-scale implementation of intermittent renewable energy sources such as solar and wind is effective energy storage. The selective electrochemical reduction of CO_2 in the CO_2 reduction reaction (CO_2RR) is a crucial strategy for storing energy from intermittent energy sources in the form of chemical bonds (e.g. solar fuels). Current state-of-the-art solid-state catalyst materials produce useful products such as methanol or ethylene, but typically do so non-selectively with a variety of gaseous and liquid products including H_2 from competitive water reduction. Alternatively, molecular catalysts show promise for more selective reduction of CO_2 to single products, typically CO or formic acid, but typically show lower overall activity compared to solid-state analogues. My research group is focused on the development of new catalytic systems that reduce CO_2 with the selectivity of molecular catalysts but operate with the activity of solid-state catalytic materials.

In this talk, I will highlight two complementary approaches my group has taken to explore selective CO_2 reduction. First, I will present some of our recent studies exploring the use of polymer encapsulation to control the chemical environment surrounding molecular electrocatalysts in order to increase catalytic activity and selectivity for the CO_2RR . In particular, I will demonstrate that encapsulating cobalt phthalocyanine (CoPc), a mediocre CO_2RR catalyst, within an encapsulating polyvinylpyridine polymer leads to a dramatic increase in the activity and selectivity for the CO_2RR . I will discuss how the encapsulating polymer surrounding the CoPc center modulates the primary, secondary, and outer coordination sphere of the catalysts and how modifying these coordination spheres lead to profound impacts on the catalytic activity, reaction selectivity, and mechanism of CO_2 reduction.

In the second part of the talk, I will discuss our efforts to design new discrete molecular catalysts based on Co complexes with redox-active bis(pyridylmonoimine) ligands (Co(BPMI)). In acetonitrile solutions with low acid concentrations, Co(BPMI) reduces CO₂ selectively to formic but shows poor stability upon catalytic turnover. However, addition of a proton source dramatically enhances the catalyst activity and stability but changes the CO₂RR product from formic acid to CO. I will discuss the mechanism of CO₂RR by Co(BPMI), present some of our efforts to design new related systems to improve activity by modulating ligand flexibility, and show preliminary data from the incorporation of these and related systems into multimetallic assemblies for CO₂ reduction.

Thursday, October 10, 2019 12:00 NOON Room change: Seminar in Room 1300 – Biomedical & Physical Sciences Pizza and coffee/tea in Room 1400A starting at 11:45 am Professor Angela Wilson - Host