UNIVERSITY OF

COLLEGE OF ARTS & SCIENCES Department of Chemistry Graduate Student Literature Seminar November 18, 2024 @3:00 pm CBLL16

Controlling selectivity in electrocatalytic CO2 reduction with an external magnetic field

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The electrochemical reduction of CO_2 under ambient conditions is an important and attractive process to mitigate rising environmental ramifications of the rising atmospheric concentration of CO_2 and to produce valuable chemical feedstocks. A preeminent challenge in this process is controlling reaction selectivity. In this seminar, I will present our approach of using an external magnetic field to improve the selectivity of driving the CO_2 reduction against the competing H_2O reduction reaction in neutral, aqueous electrolytic solutions. While magnetic fields can affect electrochemical reactions in numerous ways, we focus on the influence of the Lorentz force in electrocatalytic selectivity, as this is the most prevalent mechanism that persist across all electrochemical systems and magnetic properties of electrocatalysts.¹ In the first part of the talk, I will describe how the presence of an external magnetic field affects the competition between the H_2O and CO_2 reduction reactions by increasing mass transport via the Lorentz force.² With electronalytical chemistry and gas chromatography, we find that both an increase in magnetic field strength and an increase in current density leads to an increase in the ratio of C0 to H_2 products from the respective reactions. Finite-element simulations and voltammetry reveal that the increase in CO_2 reduction selectivity in a magnetic field is attributable to a decrease in interfacial pH, an increase in pH gradient at the electrode–electrolyte interface, and increase in interfacial CO_2 concentration. In the second half of the talk, I will share how we use small quantities of ionic liquid additives in the electrolyte, which form charged complexes with CO_2 , to improve the transport of CO_2 to the electrode surface in a magnetic field, yielding greater enhancements in reaction selectivity.³ I will also discuss how the mass of the ionic liquid affects the mass transport enhancement and selectivity between CO_2 and H_2O electroreduction.

1. Karki, N.; Mufoyongo, F. L.; Wilson, A. J. Utilizing the Magnetic Properties of Electrodes and Magnetic Fields in Electrocatalysis. Inorg. Chem. Front. 2024, 11 (17), 5414–5434. https://doi.org/10.1039/D4QI01296H.

2. Karki, N.; Marquina, I. G.; Hemmer, J. V.; Yu, Y.; Wilson, A. J. Suppressing Competing Solvent Reduction in CO₂ Electroreduction with a Magnetic Field. J. Phys. Chem. Lett. 2024, 7045–7054. https://doi.org/10.1021/acs.jpclett.4c01672.

3. Karki, N.; Wilson, A. J. Enhancing Electrocatalytic CO₂ Reduction Using Imidazolium-Based Ionic Liquids in the Presence of an External Magnetic Field. 2024, in preparation.