

# Electrochemical Conversion of Greenhouse Gases and Air Pollutants: Carbon Dioxide, Carbon Monoxide, and Nitrogen Oxides

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Motivated by the need to mitigate greenhouse gas and air pollutant emissions and the decreasing price of renewable electricity, electrochemical conversion technologies have emerged as promising alternatives to existing technologies which typically rely on fossil fuels. Various molecules, including carbon dioxide (CO<sub>2</sub>), carbon monoxide (CO), and nitrogen oxides (NO<sub>x</sub>), can be electrochemically converted to value-added fuels and chemicals or harmless molecules. In this thesis, I present remaining challenges, opportunities, and our effort to further advance the electrochemical technologies to convert three sets of gases (i.e., CO<sub>2</sub>, CO, and NO<sub>x</sub>) via catalyst development, reactor engineering, reaction environment modification, and fundamental studies.

I first examine the effect of NO<sub>x</sub> (i.e., nitric oxide, nitrogen dioxide, and nitrous oxide) in electrochemical CO<sub>2</sub> reduction reaction on three model electrocatalysts (i.e., copper, silver, and tin). Because industrial CO<sub>2</sub> point sources often contain gaseous impurities, understanding the effect of these impurities in electrochemical CO<sub>2</sub> reduction reaction is crucial for practical application.

Next, I develop a non-equilibrium synthesis method, in collaboration with Liangbing Hu group at University of Maryland, to synthesize a wide range of homogeneously mixed copper-based bimetallic catalysts regardless of the miscibility of the two metals and evaluate them in electrochemical CO reduction reaction towards multi-carbon (C<sub>2+</sub>) products formation. The non-equilibrium method gives access to novel catalysts that cannot be synthesized using conventional methods and provides an ideal platform to study the effect of secondary metals on copper catalyst in electrochemical CO reduction reaction.

Then I develop the two-step electrochemical CO<sub>2</sub> conversion process towards selective acetate and ethylene formation, in which CO<sub>2</sub> is first converted to CO in the first electrolyzer, and CO is further converted to acetate and ethylene in the second electrolyzer. The flexibility to optimize two electrolyzers individually allows for the more effective C<sub>2+</sub> products formation compared to direct CO<sub>2</sub> electroreduction in one step.

Lastly, I show an electrochemical route to convert NO<sub>x</sub> at high reaction rates (400 mA cm<sup>-2</sup>) under ambient conditions. Activity and selectivity of various transition metal catalysts are examined and strategies to steer selectivity via reaction environment modification (i.e., NO coverages and electrolyte pH) are described. This work offers an alternative method to electrochemically abate NO<sub>x</sub> emissions using renewable electricity.