Models Across Multiple Length Scales to Advance Biomass Upgrading

by

Michael J. Orella

Submitted to the Department of Chemical Engineering on 27 March 2020, in partial fulfillment of the requirements for the degree of
Doctor of Philosophy in Chemical Engineering

Abstract:

The development of efficient electrochemical processes, which can utilize electrons from renewable energy sources and incorporate sustainable sources of carbon present in biomass, could enable the decarbonization of the industrial sector and spur technological and scientific innovation. Moreover, electrochemical processing, specifically hydrogenation and hydrodeoxygenation, may allow new molecular transformations at previously unachievable conditions, unlocking what had been inaccessible or unimaginable chemical processing. Accordingly, there is significant room for exploration in organic electrochemistry to identify opportunities within the chemical industry to both replace crude-oil derived feedstocks with biomass and to shift from traditional thermally-driven reactions to those that use electrical energy. Advancing the science and engineering of these nascent process concepts requires an interdisciplinary approach with key knowledge gaps that traverse distinct research communities and apply to the problem at multiple scales.

My thesis work developed modeling toolkits that will be useful across the spectrum of biomass generation *in planta* to electrochemical processing of liquefied feedstocks, all of which are available open source to reduce the barrier to entry for new researchers interested in the potential for this interdisciplinary topic. Specifically, I developed Lignin-KMC, a model based on kinetic Monte Carlo methods that utilize first-principle-derived kinetic parameters to predict the structure of native lignin biopolymers, as having an accurate molecular model of reactor feeds is necessary to understand any reactivity trends that may be observed. Next, I created DropPy, a Python-based toolkit for automating the analysis of contact angle goniometry data, as the performance of many electrochemical cells can be anticipated from the wettability of the electrode surface. Finally, I established a generalized techno-economic framework which could be used to evaluate the overall cost to the consumer of electrochemically-derived products, and could be used by researchers with various electrolysis interests to better understand the most critical areas of improvement for their devices. Through these three developments, the efficiency of research carried out in this space will be improved, hopefully speeding the eventual development of electrochemical upgrading devices at a lower total research cost and final system price.

Thesis Supervisor: Fikile R. Brushett
Title: Cecil and Ida Green Career Development Chair, Associate Professor of Chemical Engineering

Thesis Supervisor: Yuriy Román-Leshkov
Title: Professor of Chemical Engineering