

FEBRUARY 28, 2025 @ 9:00 AM | 102 COLBURN LAB

YA-HUEI (CATHY)  
CHIN

University of Toronto

Professor in the Department of Chemical Engineering &amp; Applied Chemistry

**CATALYTIC DANCES OF  
MOLECULES ON FREE  
ENERGY SURFACES:  
DEVELOPING KNOWLEDGE  
FRAMEWORK EXPLOITING  
EXCESS POTENTIAL IN C-O  
AND C-H BOND SCISSION  
CATALYSIS**

Chemical transformation of oxygenates derived from biomass feedstock to value-added products and liquid energy carriers involves multiple reactions, each containing its own catalytic cycle with sequences of elementary reaction steps. Within these intertwined catalytic cycles, C-O and C-H bond scissions, together with hydrogen attack steps are ubiquitous catalytic events. The relative rates of these steps, within the context of transition state theory, relate directly to the free energy landscape of the reactions; these rates, together, govern the surface coverages of the reactive intermediates or spectators, the selectivity, and also the catalyst life.

In this lecture, I will introduce the thermodynamic and kinetic frameworks to rationalize how reaction microenvironment, defined as the immediate surrounding of the active sites and reacting species, would alter the fate of the reacting species, illustrated with selected examples of C-O and C-H bond scission catalysis of alcohols and phenolic species. By analyzing the common mechanistic traits of uni-molecular C-O bond scission catalyzed by Brønsted acid sites, pinpointing the key descriptor of solvent molecules leading to the promotional effects, and putting the observed reactivity trends within the context of thermochemistry (of the transition states vs. that of the reactant state), we are able to capture and rationalize the catalytic consequences of local environments on rates, covering a wide range of solvents and reactants. We put forward a multi-dimensional kinetic phase diagram, connecting the thermochemical properties of the catalyst and reactants to their turnover rates. Using our approach, we are able to rationalize and then to tune the reaction microenvironment, thus moderating the turnovers for several classes of reactions, including the hydrogen insertion chemistry in hydrogenation/hydrodeoxygenation reactions of substituted phenols, electrophilic addition reactions of phenols and carbonyls, as well as C-O scission reactions in alcohols. This platform knowledge serves as a guide towards designing reaction microenvironment that would expedite catalytic turnovers.