Center for Molecular Electrocatalysis (CME)
EFRC Director: R. Morris Bullock
Lead Institution: Pacific Northwest National Laboratory
Class: 2009 – 2024

Mission Statement*: To establish cross-cutting principles that facilitate catalyst design for interconversion of electrical and chemical energy, across molecular and heterogeneous catalysis.*

CME will reveal the key mechanistic principles that unite and differentiate molecular and heterogeneous catalysis by providing an integrated thermodynamic framework broadly underpinning catalyst design. Through synergistic parallel efforts, we investigate molecular constructs both in solution and incorporated into electrode interfaces, while also controlling the environment beyond the binding site. (Figure 1).

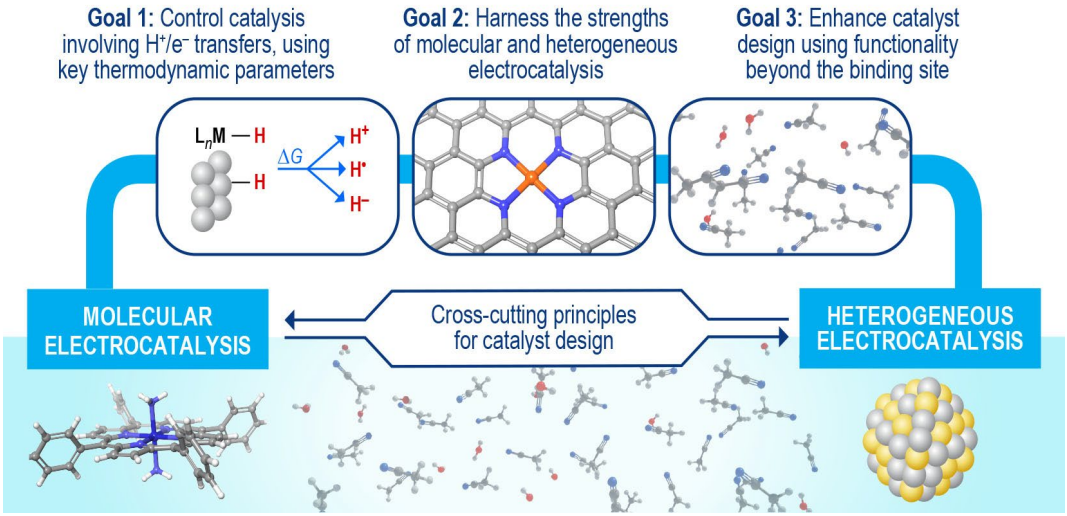

Figure 1. CME is developing cross-cutting design principles spanning molecular and heterogeneous electrocatalysis.

The impact of CME research will enable critical innovations in the design of catalysts. We seek to harness the most attractive features of molecular and heterogeneous catalysts through tailored coordination and solvation environments, such that highly active, efficient, selective, and durable catalysts can be designed for chemical transformations critical to energy conversions. As our research has evolved to include heterogeneous catalysis and interfacial studies, *our core principle has not changed: all catalysis is molecular, involving bond-breaking or bond-forming in molecules at active sites*, whether that site is in a soluble molecular complex, at a nanoparticle, or on a heterogeneous surface. Heterogeneous surfaces offer distinct attributes—including band-mediated charge transport and the ability to organize multiple cooperative active sites—that can be uniquely leveraged by the molecular perspective of CME. Our research is advancing catalyst design by unifying molecular and interfacial reactivity while mastering complex media, building upon our established strength in thermodynamic principles.

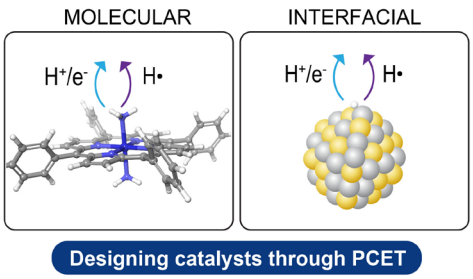

Figure 2. CME is using a proton-coupled electron transfer (PCET) framework as the basis for designing both molecular and interfacial catalysts through precise control of proton and electron transfers.

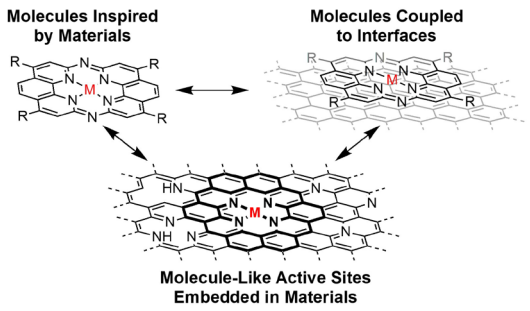

Figure 3. CME aims to bridge and combine principles from molecular and interfacial catalysis to develop new classes of molecules and materials for electrochemical energy conversion.

In the Thrust on **Designing Catalysts through a PCET Framework** (Figure 2), we are developing the fundamentals of energy-related electrocatalysis through a thermochemical PCET lens, combing experiment and theory. We are developing an understanding of the thermochemical landscapes of homogeneous and heterogeneous PCET and oxygen atom transfer processes and the implications of those landscapes for catalysis. Our Thrust on **Bridging Molecular and Interfacial Approaches to Electrocatalysis** seeks to develop unifying principles and predictive models that span and unite the traditionally disparate fields of molecular and heterogeneous electrocatalysis (Figure 3). Our research is exposing how interactions between a molecule and a polarized interface lead to distinct electrocatalytic reaction mechanisms and scaling behaviors. Our research Thrust on **Harnessing the Extended Catalyst Environment** develops the fundamental principles that underlie electrocatalytic processes at heterogeneous interfaces and with molecular catalysts in non-aqueous and low-aqueous media (Figure 4). We focus on how aspects of the catalyst system beyond the active site, including solvents, electrolytes, and redox mediators, affect the mechanisms and kinetics of electrocatalytic reactions to access new regimes of activity, selectivity, and stability.

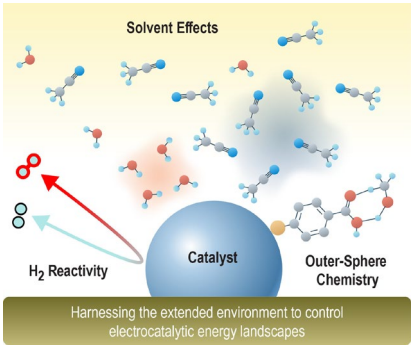

Figure 4. CME focuses beyond the binding site to develop design principles based on the extended environment, including the roles of solvents and outer-sphere interactions.

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