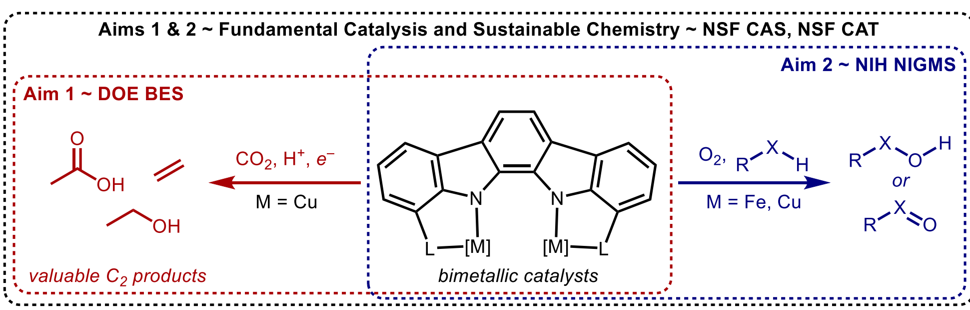
Overview/Abstract – Bimetallic Catalysts for CO_2 Upcycling and Oxygenase Mimicry

The use of catalysts in modern chemical industries plays a pivotal role in the production of everything from Styrofoam® and Nylon® (i.e. commodity chemicals, >23 million tons/yr combined) to pharmaceuticals like ibuprofen (i.e. specialty chemicals, <0.05 million tons/yr). However, while these materials and chemicals play a vital role in manufacturing, medicine, and agriculture, their production and value to modern society is often heavily decoupled from their environmental impact. Most commodity chemicals are produced directly from fossil fuels in energy-intensive processes that release significant amounts of CO_2 that is exacerbated by their large scale. Relatedly, specialty chemicals rely on increasingly scarce precious metal catalysts to drive transformations with a high degree of selectivity and yield, but these metals are increasingly difficult to source and current sources are rapidly depleting. Unfortunately, the underlying *fundamental chemistry needed to address these problems in sustainability remains underdeveloped*.

Therefore, herein we propose to develop a new modular catalyst platform to solve divergent problems in the production of both commodity and specialty chemicals. To achieve this goal, we will develop a new ligand platform that uses an indolocarbazole backbone to support two metal centers in a host of configurations. We believe this will allow us to mimic the reactivity of heterogeneous and enzymatic catalysts, while affording control over the bimetallic active site. In turn, this high degree of molecular precision will allow us to drive reactions with high yield and selectivity, while also linking catalyst structure and function. In turn, our detailed understanding of how our catalysts work, and why, will help us to develop improved second-generation catalysts *and* providing insight into optimal catalyst features in the enzymatic and heterogeneous catalysts that served as initial inspiration. To demonstrate the power of our approach, we will use the proposed bimetallic catalysts to address two key challenges in sustainable chemistry:

Aim 1. Bimetallic Electrocatalysts for the Upcycling of CO_2 to C_2 Products. In this aim we will synthesize a suite of $(P_2IC)Cu_2$ complex and study their ability to electrocatalytically convert CO_2 into valuable C_2 products. Notably, this approach would recycle CO_2 waste into carbon-neutral products, all while providing an alternative route to existing fossil fuel-derived commodity chemicals.

Aim 2. Bimetallic Oxygenase Mimics for Late-Stage Functionalization. In this aim we will use nature as our inspiration for developing new catalyst technologies using cheap and abundant metals such as Cu and Fe to replace increasingly scarce precious metal catalysts in the late-stage functionalization of specialty chemicals such as pharmaceuticals.



These two aims will not only serve to establish the indolocarbazole backbone as a valuable approach for addressing key challenges in the sustainable production of specialty and commodity chemicals, but they also directly address key funding targets of the DOE, NIH, and NSF. As such, the work carried out as part of this seed grant will be used as preliminary data in the submission of *at least 3 federal grant proposals*: (i) a DOE Early Career Award, (ii) an NIH R35, and (iii) an NIH R01 as well as potential (iv) an NSF CAREER. In addition, these results will also be used as the core of several early-career fellowship applications including the Sloan Research Fellowship, Beckman Young Investigator Program, and the Packard Fellowship Program.