Integrated Mesoscale Architectures for Sustainable Catalysis (IMASC)
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Mission Statement*: Improve catalytic selectivity by quantitatively scaling from model studies to catalytic conditions using advanced experiment and theory with a strategic objective to develop overarching design principles.*

The vision of IMASC is to advance the fundamental science necessary to reduce the carbon footprint of the chemical industries sector. Specifically, the mission is to develop the ability to increase energy efficiency through improvement of catalytic selectivity using dilute alloy catalysts, by quantitatively scaling from model studies to catalytic conditions using advanced experiment and theory. The principle governing the use of dilute alloy catalysts is that the reactive minority metal, e.g. Pd, Ni, Pt, and Ag, initiates the catalytic cycle, whereas the abundant host, Au or Ag, imparts selectivity. A strategic objective is to develop overarching design principles and to develop a general methodology for understanding catalytic processes.

Background, knowledge gaps, Mission and Goals
Chemical production, which relies heavily on heterogeneous catalysis, now accounts for nearly 25% of energy use worldwide. Forecasts for global energy demand project this number will rise to 45% by 2040. The Center's mission and operational imperative is rooted in a singular reality: current trends in energy supply and use are unsustainable—economically, environmentally, and socially. A fully integrated theoretical-computational-experimental approach to the design of selective catalysts is needed to boost energy efficiency in the industry's production processes. IMASC 2.0 answers this call, with significant progress already made at the intersection of surface chemistry and physics to transform how catalysts are designed.

Heterogeneous catalytic processes are extremely complex, requiring optimization of factors across multiple scales of length, time, pressure, and temperature. To develop such catalyst processes mandates team science and an interdisciplinary approach, inclusive of materials synthesis, mechanistic surface chemistry, reaction kinetics and *in-situ* and *operando* characterization. Numerous studies show that complex metal/oxide interfaces, generally present in catalysts, appear to play an important synergistic role in determining reactivity. Further, since materials are often affected by the reaction environment, pre- and on-stream activation and optimization of performance is necessary.

Historically, heterogeneous catalytic processes have been devised empirically, with broad guidelines informed by prior experience in organic and organometallic chemistry. Recent advances in theory and experiments provide tools with the potential to move beyond the traditional "trial-and-error" approach to design principles that predict and develop highly efficient heterogeneous catalysis materials systems. IMASC bridges this divide and address key knowledge gaps motivated by key basic research needs identified by DOE.

Four scientific goals support this mission

 **Design and synthesize robust nanoporous dilute alloy catalysts.**

IMASC has developed a toolbox of dilute, alloy catalyst materials designed to provide insight into catalyst function. Two classes, a free-standing nanoporous material and a raspberry colloid templated nanoparticle material, each demonstrate excellent properties. They exhibit remarkably stable catalytic performance and impart a high degree of reaction selectivity.

 **Predict catalytic selectivity through understanding of kinetics and mechanism.**

A central goal of IMASC is to predict reaction selectivity under catalytic conditions based on reaction mechanisms and kinetics determined from fundamental studies. In IMASC remarkable progress has been made on the research objective to advance the paradigm for control of selective oxidation and hydrogenation on dilute alloy catalysts and to quantitatively model their kinetics.

 **Exploit rearrangement at interfaces to enhance selectivity and activity.**

IMASC is focusing on a critical challenge in heterogeneous catalysis—the development of catalytic processes that achieve and maintain high activity and high selectivity through optimization of the composition and structure of dilute alloy catalysts. A key principle underlying our approach is to devise an approach whereby the minority more reactive metal is at or near the surface so it can initiate the catalytic cycle; e.g. O_2 activation for oxidation or H_2 dissociation for hydrogenation

 **Advance methodology for catalytic design.**

Understanding the evolution of structure and composition of catalysts and their relationship to catalytic function is a central goal of IMASC that is enabled by the development and implementation of advanced characterization and theoretical modeling. Experimental tools that have been advanced in IMASC include *in operando* atomic-scale imaging using electron microscopy, structural determination using EXAFS in combination with machine learning, and atom probe tomography for 3-D imaging of composition using atom probe tomography. Theoretical tools have also been advanced in IMASC research that model catalyst restructuring using accelerated molecular dynamics, and kinetic modeling of reactive steps by including a more accurate treatment of pre-exponential factors.

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