Center for Atomic-level Catalyst Design (CALCD)
EFRC Director: James Spivey
Lead Institution: Louisiana State University

***Mission Statement**: To build effective catalysts from first principles via computational catalysis and atomic-level characterization.*

***Rationale**.* Recent advances in computational catalysis, synthesis of solid catalytic surfaces at the nanoscale, and in our ability to unambiguously characterize these materials, may allow us to control and direct energy-related reactions in ways not now possible. Despite rapid progress in these areas, we do not yet have the ability (a) to computationally design an ideal catalyst for a reaction of reasonable complexity at conditions of practical interest, (b) to prepare the catalyst with a degree of atomic-level precision that mimics the surfaces that we can simulate by computation, nor (c) to characterize the catalyst surface with atomic-level resolution, especially at working conditions.

This gap between simulated (computational) and real world catalysis can be visualized in a number of ways, e.g., by comparing the degree of difficulty in (a) calculating the interactions of even simple molecules with extremely small metal clusters and (b) in synthesizing these same clusters atom-by-atom (Fig. 1). Assuming other factors are constant (such as the complexity of the reaction), as the size of the active catalyst surface increases, the level of control over the atomic-level structure of the surface decreases, for example because defects are inevitably introduced in the preparation of the catalyst. However, a computational description of the working catalyst based on quantum mechanics becomes less realistic as the size of the catalyst surface increases (especially under reaction conditions), typically because we must make simplifying assumptions that cannot be easily verified. Although the length scales representing the limits of computation or synthesis may vary for different metals and reactions, there is a gap in our ability to identify an ideal catalyst by computation and to then prepare and characterize it unambiguously with atomic-level precision (Fig. 2).

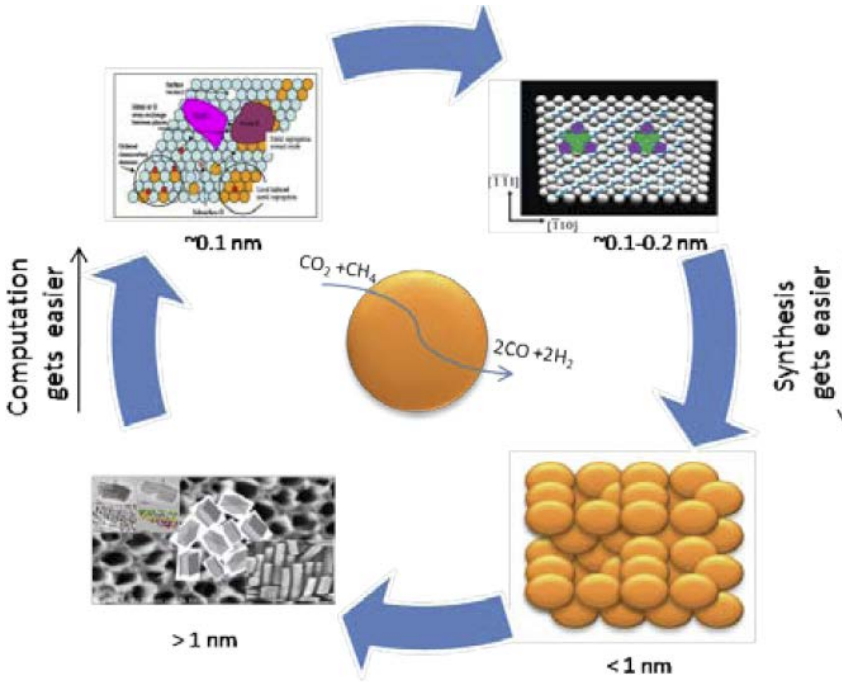

Fig. 1. Schematic representation of the degree of difficulty in computation and synthesis of catalysts.

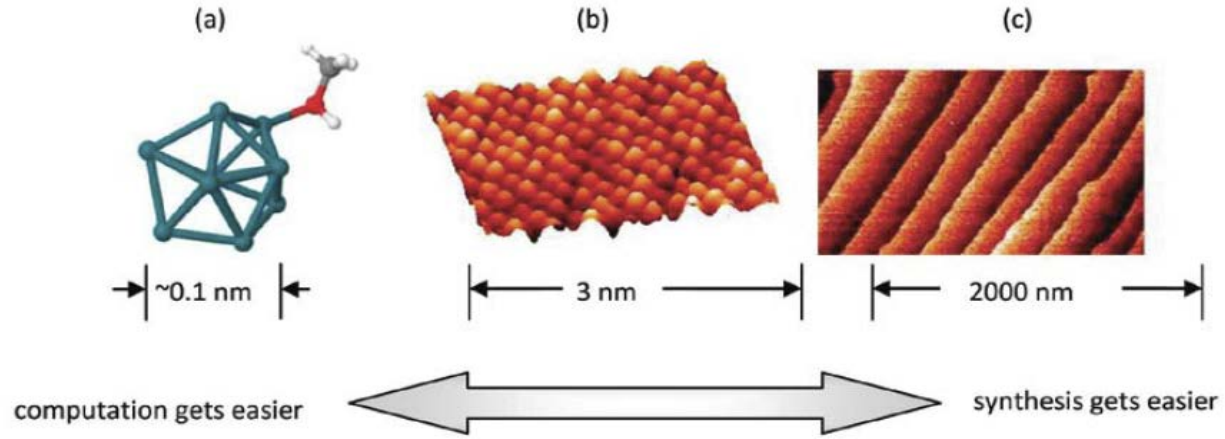

Fig. 2. Difficulty of computational and synthesis/characterization research vary in opposite directions with length scale; (a) CH_3OH on Rh_7 cluster (DFT model by D. Bruce, Clemson Univ.); (b) $Pt_{25}Rh_{75}$(100) Alloy 3-d topography (www.omicron.de); (c) Mono-atomic steps on SiO_2 layer - in UHV at T = 800K in AFM mode.

***Approach*.** Our approach is two-fold; (a) extend the capabilities of current computational and synthesis/characterization tools and (b) use these tools to synthesize and characterize computationally designed catalysts. The computational effort focuses on developing first-principle-based multi-scale models to predict catalytic behavior by following the dynamic evolution in both composition and structure over experimentally relevant time and length scales. The morphological changes and reactivity of the catalyst under various realistic conditions are being explored. Modeling predictions are validated by experiment. While Density Functional Theory (DFT) has been a very useful complement to surface science experiments, our multi-scale modeling is designed to be a step towards a more predictive role for computational simulations.

The synthesis effort is intended to extend (to ever-smaller dimensions) the length scales at which precise, computationally-specified structures of supported metals can be prepared. The approach is an iterative one that is anticipated to challenge the current limits of both synthesis tools and computational techniques.

Center for Atomic-Level Catalyst Design	
Louisiana State University	James Spivey (Director), C. Kumar, W. Plummer, R. Kurtz, P. Sprunger, K. Dooley, G. Griffin, J. Flake, A. Roy, Y. Xu
Clemson University	D. Bruce
Texas A&M University	L. Liu
Georgia Tech	D. Sholl
University of Utrecht, Netherlands	K. P. de Jong, P.E. de Jongh
University of Florida	S. Sinnott
Ohio State University	A. Asthagiri
Technical Univ. of Vienna	U.Diebold
Pennsylvania State University	M. Janik
Grambling State University	N. Seetala

Contact: *Ellen Stevens, Business Manager; 225-578-0058; esteve2@lsu.edu; www.efrc.lsu.edu*