

Science.

Technology.

Innovation.

The grand challenge for catalysis science in the 21st century is to understand how to design catalyst structures to control activity and selectivity, and use this understanding in addressing a secure energy future.

Pacific Northwest National Laboratory Institute for Interfacial Catalysis Basic Research

Advances in capabilities to control, measure, and compute chemical and physical properties of materials accurately and with exquisite spatial and temporal resolution are reinvigorating fundamental catalysis research. This research is critical to developing innovative catalyst materials and technologies to efficiently and effectively use energy sources to sustain our standard of living without compromising our environment. The Pacific Northwest National Laboratory's (PNNL's) Institute for Interfacial Catalysis is answering some of the most critical unsolved problems in catalysis.

Studying Active and Selective Catalysts From the Atom Up

To answer the grand challenge questions in catalysis science, we are unlocking the structure and behavior of materials at the atomic and molecular levels.

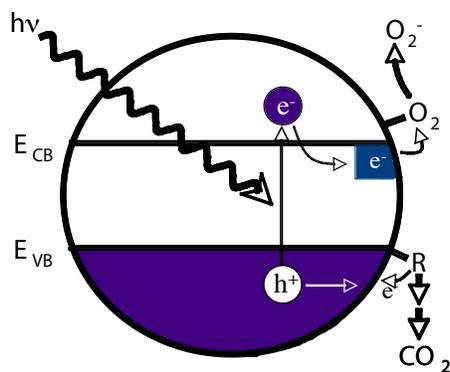
Here are a few examples:

To answer questions important to building solar-initiated processes, we are determining molecular-level interactions of the heterogeneous photocatalyst TiO_2 . Specifically, we are studying

- Organic photooxidation reaction pathways
- Rates, mechanisms, and energy redistributions associated with charge carrier generation, separation, diffusion, trapping, and reactivity
- Dependence of photocatalytic reactions on the phase and molecular structure of the TiO_2 surface.

To investigate the fundamental aspects of NO_x surface chemistry on base metal oxide nanostructures, we are

- Preparing and characterizing base-metal oxide nanostructures formed on well-ordered oxide substrates
- Characterizing the surface chemistry of small molecules containing unpaired electron density (NO_x) on the model oxide systems.



TiO_2 nanoparticle with its band structure superimposed. A photon excites an electron across the band gap, and both charge carriers (electron and hole) diffuse to the surface where they perform redox chemistry on adsorbed species. This process is catalytic for multiple such events when both halves of the redox reaction coexist and compensate each other.

Pacific Northwest
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Characterizing the base metal oxide nanostructures, and studying the reaction chemistry of NO_x molecules on the base metal oxide relies heavily on an array of traditional and cutting-edge resources, including variable temperature ultrahigh vacuum (UHV) scanning tunneling microscopy; Auger, X-ray and Fourier Transform Infrared (FTIR) spectroscopies; and temperature-programmed desorption.

To understand and control the energies of catalytic intermediates involved in molecular catalytic processes, we are investigating

- New thermodynamic and kinetic methods for measuring heterolytic and homolytic bond dissociation free energies and enthalpies for M-H, S-H, M-C, and S-C bonds
- Role of structural features of the first- and second-coordination spheres in determining these bond energies
- Empirical models and theoretical methods (density functional theory calculations) for predicting both heterolytic and homolytic bond energies, and the relative energies of catalytic intermediates
- The use of this information and models in the rational design of catalysts for a variety of processes, including electrocatalytic H_2 oxidation and production, H_2 storage, dehydrosulfurization, CO reduction, and other catalytic reactions of importance to the nation's energy future.



Pushing the Frontiers of Fundamental Research

The Institute for Interfacial Catalysis leads research projects that expand the frontiers of catalysis science, including

- Early transition metal oxides as catalysts: Crossing scales from clusters to single crystals to functioning materials led by Charles Peden
 - Fundamental studies of photocatalysis on TiO_2 led by Michael Henderson
 - Multiscale synthesis of transition metal oxide catalysts led by Jun Liu
 - An energy-based approach to molecular and interfacial catalyst development: Thermochemistry, kinetics, and theory of catalytic transformations led by James Franz
 - Stimulus-controlled catalysts as bio-inspired mimics led by Wendy Shaw
- Fundamental investigations of water splitting on model TiO_2 photocatalysts doped for visible light absorption led by Michael Henderson
 - Control of hydrogen release and uptake in condensed phases led by Thomas Autrey
 - Nanostructured catalysts for hydrogen generation from renewable feedstock led by Yong Wang
 - Fundamental studies of nitrogen oxide surface chemistry: A model system approach led by János Szanyi
 - Cationic ionic hydrogenations: Developing concepts and new catalytic processes that substitute inexpensive metals for precious metals led by Morris Bullock.

Fundamental catalysis science at PNNL is possible because of the Institute for Interfacial Catalysis's expertise in material synthesis, surface chemistry, scanning microscopy, photodynamics, and theoretical modeling.

About PNNL

Pacific Northwest National Laboratory, a U.S. Department of Energy Office of Science laboratory, solves complex problems in energy, the environment, and national security by advancing the understanding of science. PNNL employs more than 4000 staff, has a business volume of \$750 million, and has been managed by Ohio-based Battelle since the lab's inception in 1965.

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