PNNL/IIC R&D Activities in Environmental Catalysis

Chuck Peden

Laboratory Fellow & Associate Director

Institute for Interfacial Catalysis

chuck.peden@pnl.gov

- Diesel vehicle emission control
- Photocatalysis
- Past history in methane production via hydrogenation of industrial waste streams (led, in part, to activity in catalysis for upgrading biomass)

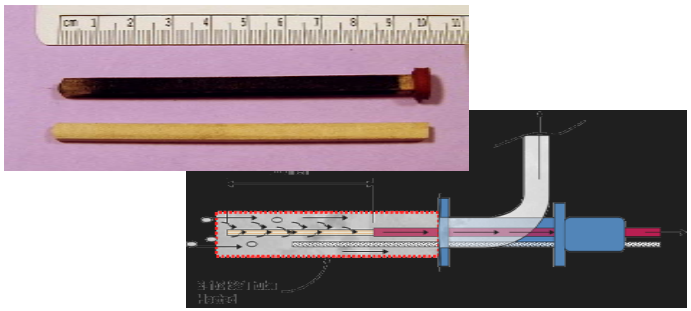
DOE/EE-funded Vehicle Emission Control Work in the IIC at PNNL

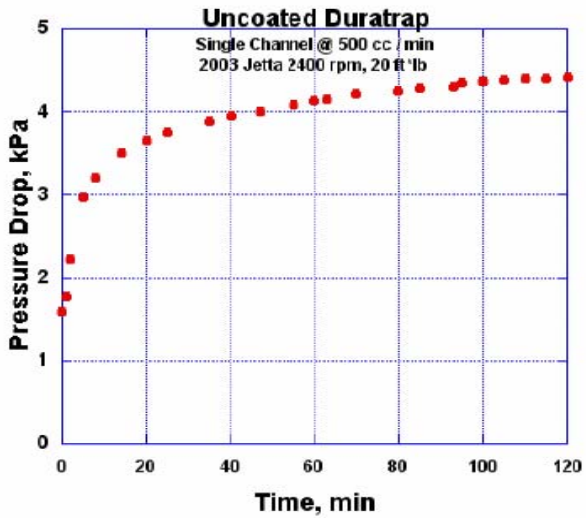
- R&D on all catalyst technologies for reducing emissions from diesel-powered vehicles
  - NOx Reduction (hydrocarbon and urea SCR, and lean-NOx traps)
  - Particulate (Soot) Reduction
  - Sulfur Removal
  - Low Temperature CO and Hydrocarbon Oxidation (for low temperature combustion engines now in development)
- Strong industry participation in all programs via CRADAs (past and current partners with Caterpiller, Cummins, DaimlerChrysler, Delphi, Dow, Ford, and GM)
- More than 25 staff scientists and engineers with backgrounds in chemical and mechanical engineering, physical chemistry, and materials science.
- Programs utilize EMSL science facilities and exhaust emissions lab.

Diesel Particulate Filter Characterization and Modeling

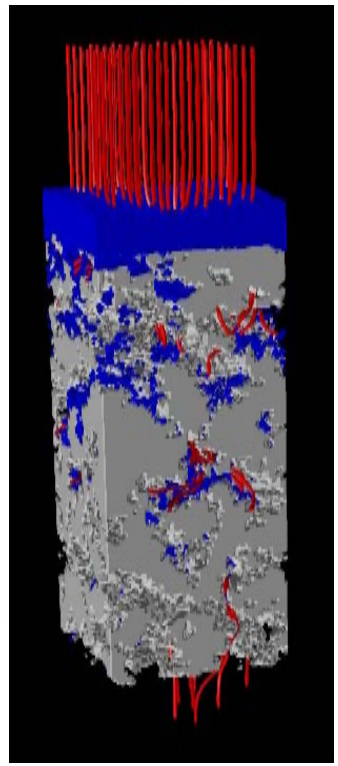
Heather Dillon , Tom Gallant, Gary Maupin, David Rector, George Muntean, Mark Stewart

Catalyst Monolith Single Channel Loading Experiments





3D Flow Field Solution Using Lattice-Boltzmann Methods



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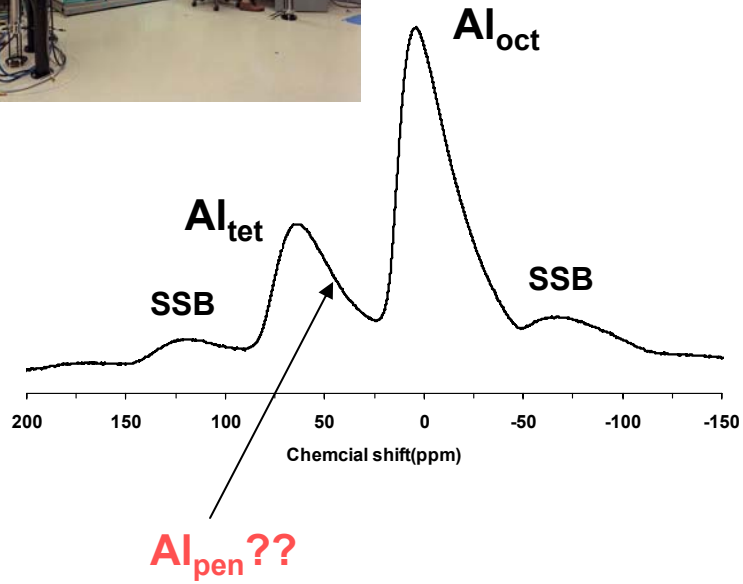
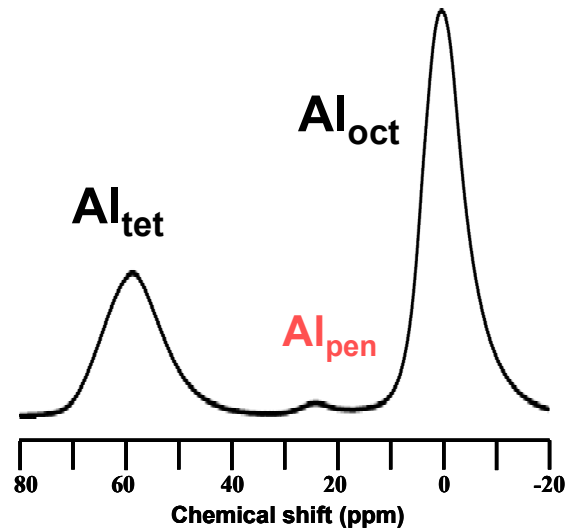
Use of one-of-a kind Ultra-High Field NMR in EMSL for Catalysis Studies



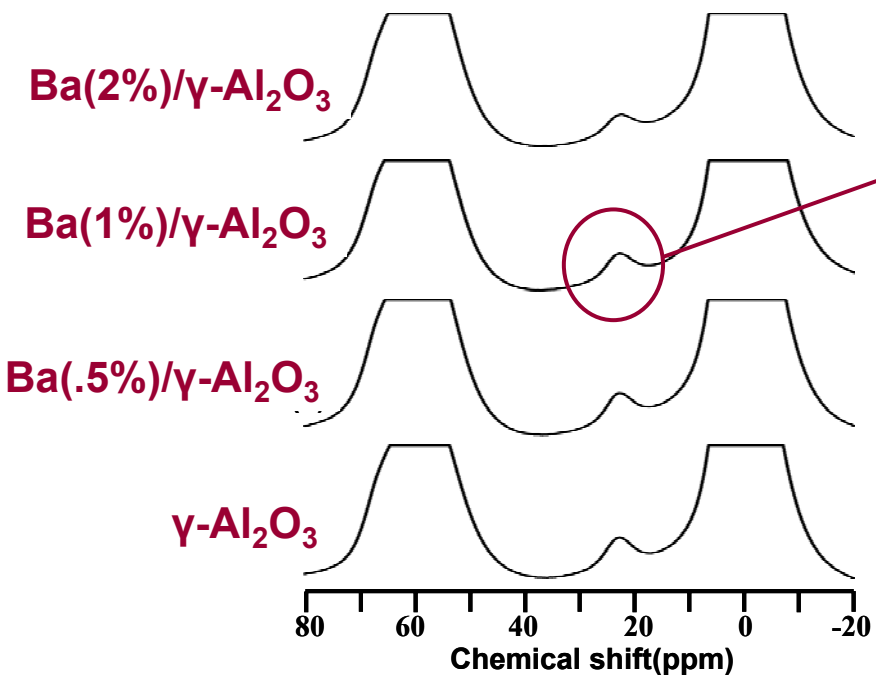


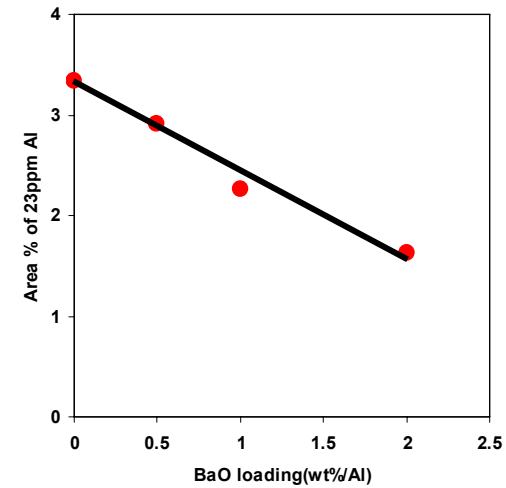
Penta-coordinate Al^{+3} ions readily observable in γ-Al_2O_3.

JH Kwak, JZ Hu, DH Kim, J Szanyi, CHF Peden, Journal of Catalysis, submitted.

Lewis acidic 5-fold Al sites on γ-Al_2O_3 surfaces are nucleation sites for catalytic phases!

Addition of a catalytic phase, BaO, quantitatively 'titrates' 5-fold Al sites.



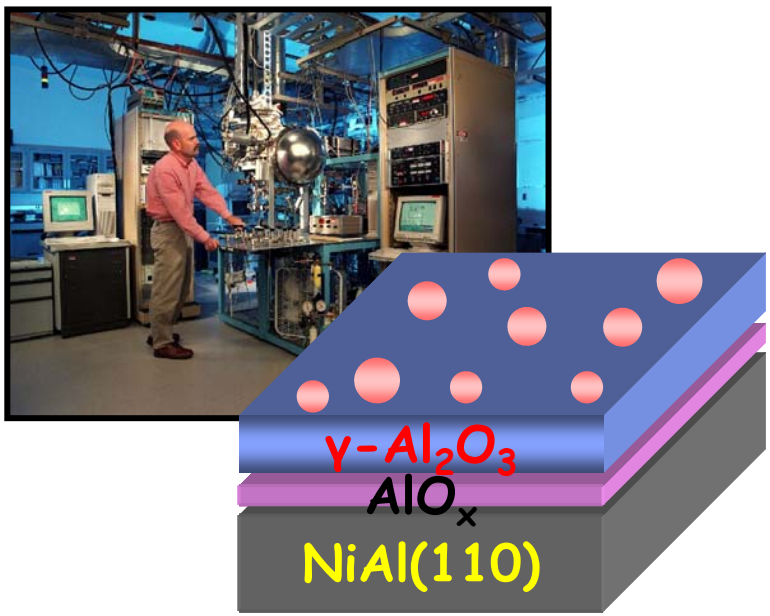


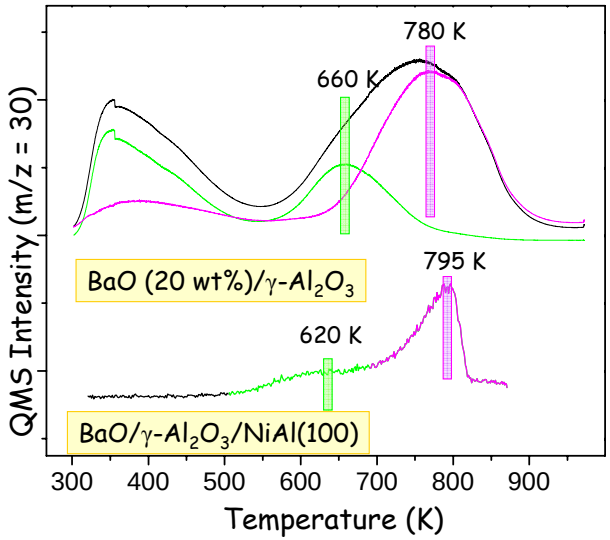
Impregnation with aqueous $Ba(NO_3)_2$ followed by calcination in dry air at 500 °C.

JH Kwak, JZ Hu, DH Kim, J Szanyi, CHF Peden, Journal of Catalysis, submitted.

Applied NOx Emission Control R&D Supported by DOE/Office of Science/BES-funded work

Fundamental studies of nitrogen oxide surface chemistry: A model system approach – Provide the conceptual foundations to describe the surface chemistry (particularly NOx) of base metal oxide nanostructures on oxide supports.



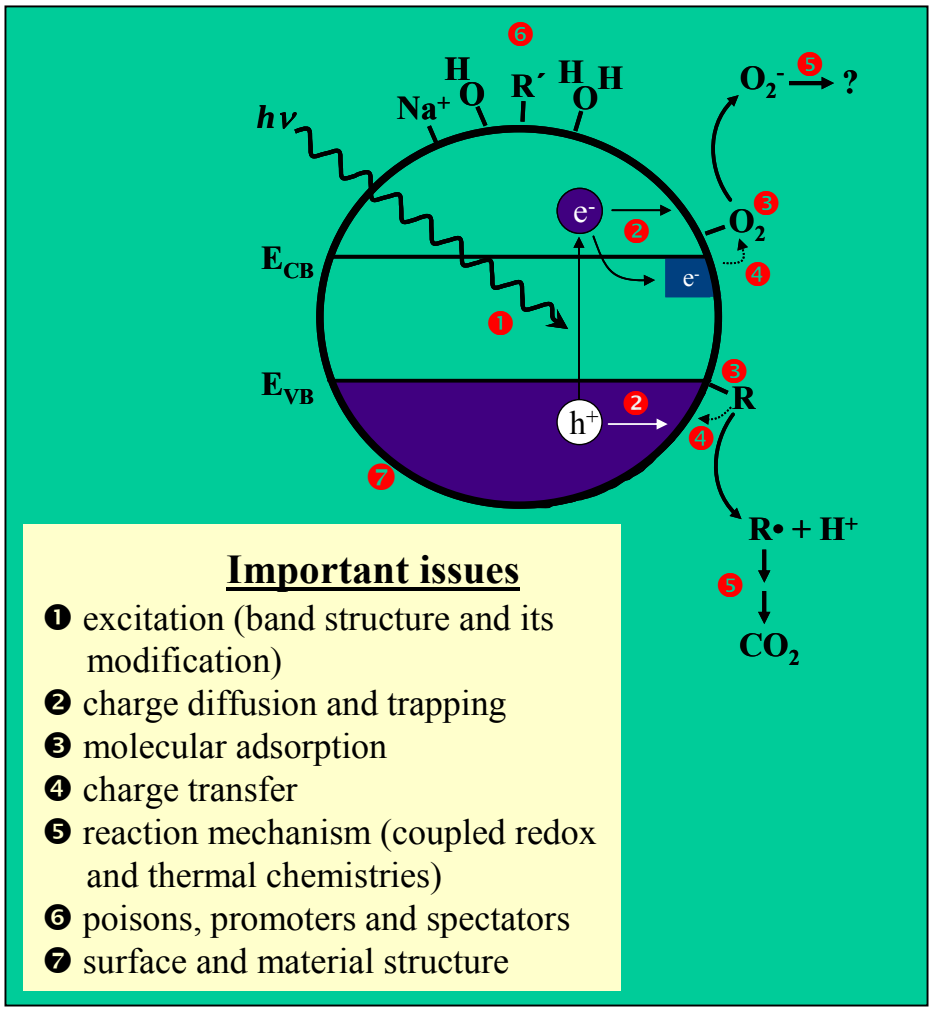


UHV TPD from the model system is consistent with the high surface area BaO on alumina

Szanyi, J.; Kwak, J.H.; Hanson, J.; Wang, C.M.; Szailer, T.; Peden, C.H.F., J. Phys. Chem. B **109** (2005) 7339

Ozensoy, E.; Peden, C.H.F.; Szanyi, J., J. Catal. **243** (2006) 149

Solar Photocatalysis



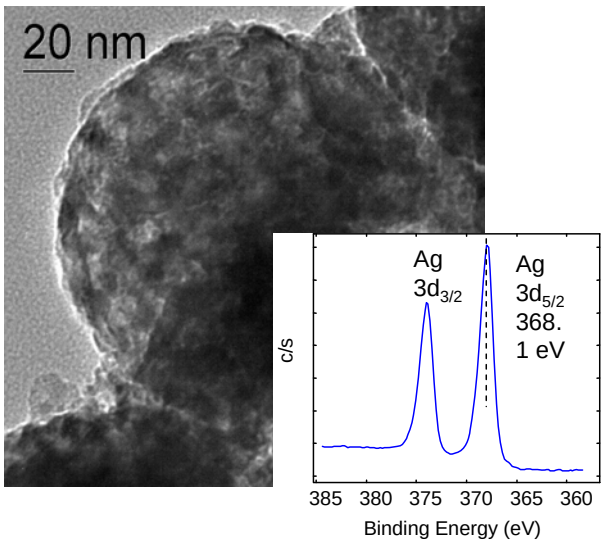
- Focus of applications:
  - organic pollutant oxidation
  - H_2 generation from water
  - CO_2 Utilization
  - More complete use of the solar spectrum
- Strong activities in fundamental science funded by DOE/Basic Energy Sciences
- New applied work currently internally funded

Synthesis and characterization of nano-structured heterogeneous photocatalysts

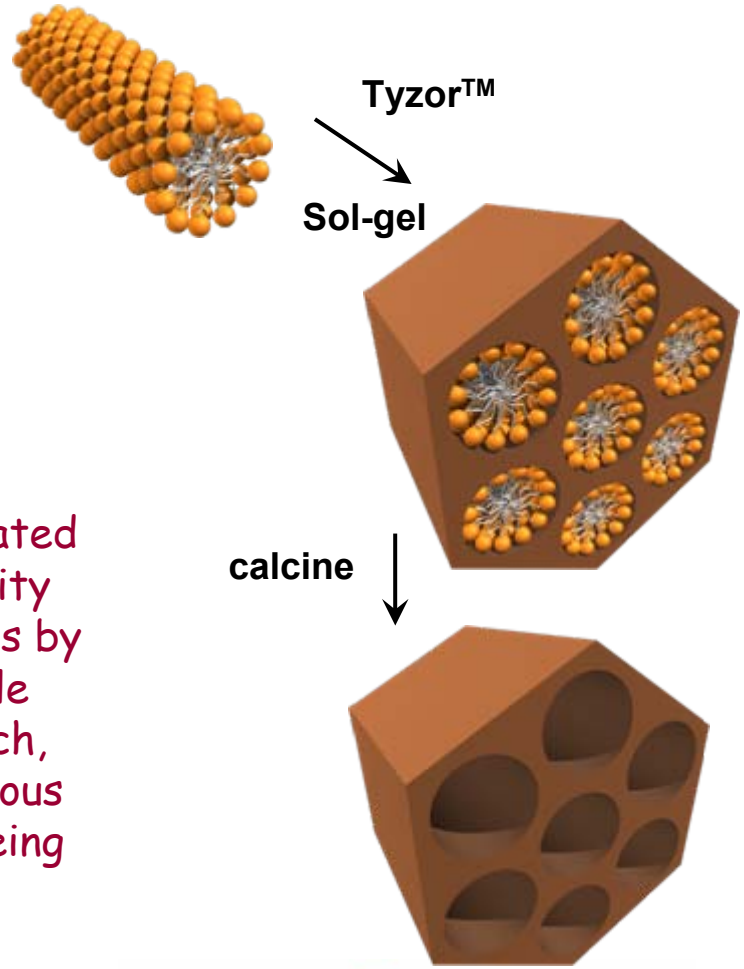
This project is focused on the design and synthesis of nanostructured, high surface area TiO_2 phases with controlled dopant concentrations.

Surfactant templated mesoporous TiO_2 powders and thin films (features on 2-3 nm scale).

Photocatalytic partial hydrocarbon oxidation is used to test activity and selectivity of these new photocatalyst compositions.



Ag(I) has been implicated in enhancing the activity of TiO_2 photocatalysts by inhibiting electron-hole recombination. As such, its effect on mesoporous titania structure is being explored.



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