

FWP KCC053 Fundamentals of Heterogeneous Catalysis on Surfaces and Nanostructures

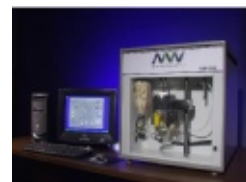
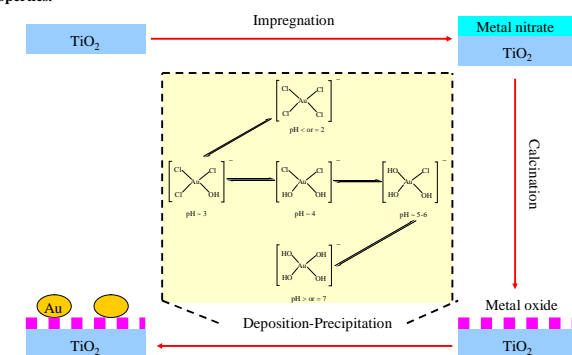
Zhen Ma, Steven H. Overbury, and Sheng Dai
Chemical Sciences Division, Oak Ridge National Laboratory
Oak Ridge, TN 37831



Synopsis: The overarching goal of our research is to investigate the correlations between interfacial structures of nanocatalysts and their catalytic properties. Here we developed three strategies for increasing the structural complexity and catalytic properties of supported gold catalysts via rational design. The first strategy is the pre-modification of TiO₂ or C supported before loading gold nanoparticles. The second strategy is the post-modification of gold catalysts after the gold nanoparticles are already loaded. The third methodology is to load gold nanoparticles on non-conventional supports, such as metal phosphates with new acidic functionalities. These catalysts, with excellent catalytic performance, open up new opportunities for further in-depth research.

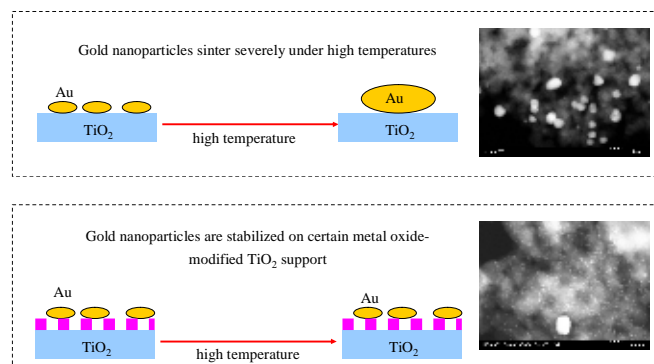
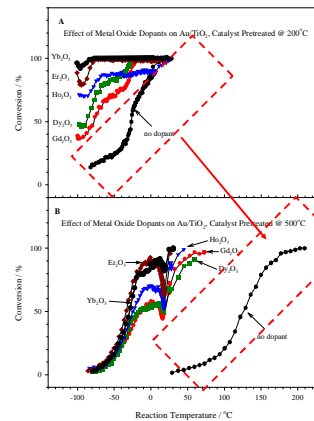
Example #1: The Modification of TiO₂-based Gold Catalysts

Summary: The goal is to study how the sintering of gold nanoparticles can be controlled by rational modification of TiO₂ support and the correlation between structure and catalytic properties.



Altamira AMI-200 reaction facility
1% CO in air, flow rate: 37 ml/min
50 mg catalyst

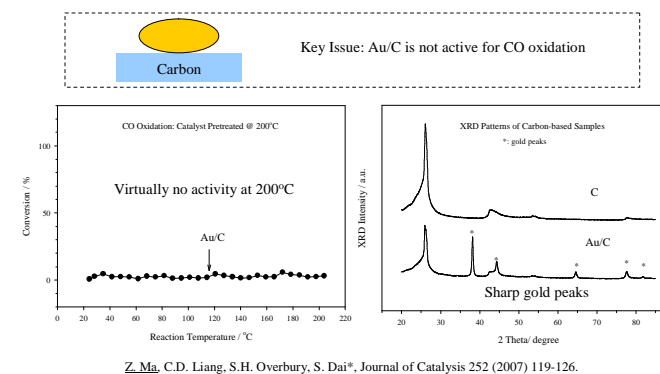
Z. Ma, S.H. Overbury, S. Dai*, Journal of Molecular Catalysis A 273 (2007) 186-197.



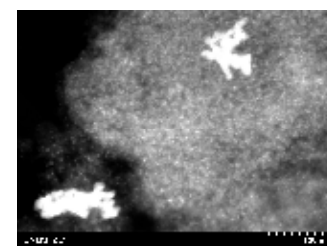
Research sponsored by the Division of Chemical Sciences, Geosciences, and Biosciences, Office of Basic Energy Sciences, U.S. Department of Energy, under contract DE-AC05-00OR22725 with Oak Ridge National Laboratory, managed and operated by UT-Battelle, LLC.

Example #2: Modification of Carbon-based Gold Catalysts

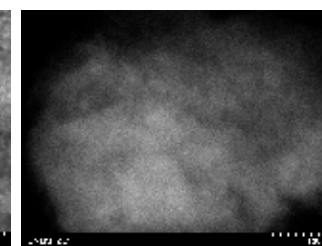
Summary: The goal is to study how inert carbon supports can be activated by MnO₂ additives and used as active supports for loading gold nanoparticles via novel materials synthesis strategies.



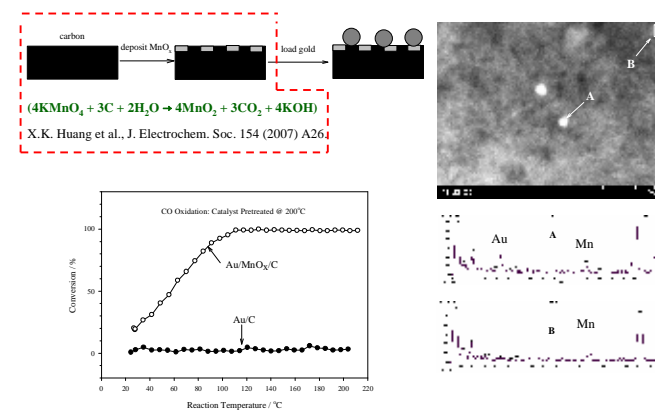
200°C-pretreated Au/C



Carbon support



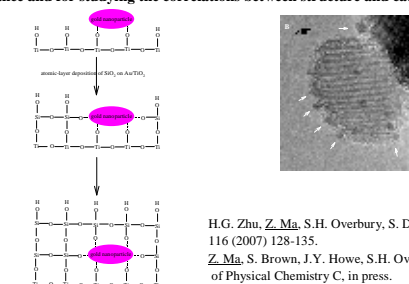
Small gold particles on carbon are not active for gas-phase CO oxidation.
Maybe pure carbon cannot activate molecular oxygen



Dr. Suree Brown, Dr. Hongfeng Yin, Dr. Chengdu Liang, and Dr. Jane Y. Howe contributed to TEM images of our publications. Dr. Suree Brown conducted the ALD.

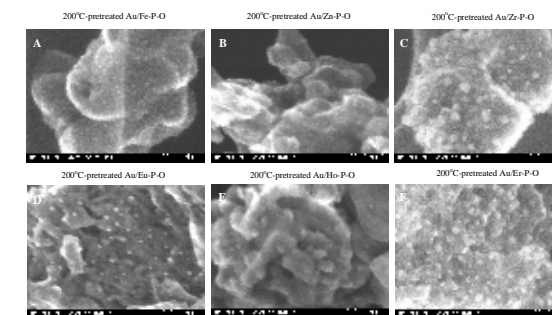
Example #3: The Post-Modification of Au/TiO₂ by SiO₂

Summary: The goal is to rationally modify supported gold catalysts by novel materials synthesis techniques and construct new interfacial structures. These interfacial structures provide new possibilities for tuning the catalytic performance and for studying the correlations between structure and catalytic properties.



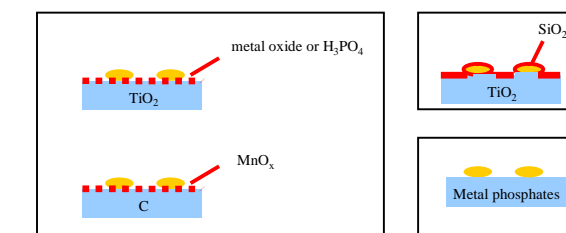
Example #4: Loading Gold Nanoparticles on Metal Phosphates

Summary: The goal is to load gold nanoparticles on acidic metal phosphates as bifunctional catalysts with both metal function and support functions. It is interesting to study the activation mechanism on our newly developed catalysts and explore their applications in organic conversion reactions.



Z. Ma, H.F. Yin, S.H. Overbury, S. Dai*, in preparation.

Conclusions: The Design of Novel Supported Gold Catalysts



Conclusions: Novel catalytic structures have been successfully designed and prepared. These catalysts show tuned catalytic properties and increased structural diversities. More importantly, they provide new opportunities for further in-depth research on the reaction mechanism, kinetics studies, structural characterization, DFT calculation, and other new reactions. Further work is in progress in our research team.

Z. Ma, S. Brown, S.H. Overbury, S. Dai*, Applied Catalysis A 327 (2007) 226-237.
Z. Ma, S.H. Overbury, S. Dai*, Journal of Molecular Catalysis A 273 (2007) 186-197.
Z. Ma, C.D. Liang, S.H. Overbury, S. Dai*, Journal of Catalysis 252 (2007) 119-126.
Z. Ma, S. Brown, J.Y. Howe, S.H. Overbury, S. Dai*, Journal of Physical Chemistry C, in press.
H.G. Zhu, Z. Ma, S.H. Overbury, S. Dai*, Catalysis Letters 116 (2007) 128-135.