

Highly Selective Partial Oxidation Reactions on Size-Selected Nanocatalysts: Towards the Understanding of Size/Shape & Function Relationship in Catalysis

S. Vajda[#], L.A. Curtiss[#], M.J. Pellin[#], R.E. Winans[#], C.L. Marshall[#], J.P. Greeley[#], G. E. Ballentine[#],
J.W. Elam[#], S. Lee[#], B. Lee[#], Y. Lei^{*#}, C. Lo[#], R.J. Meyer^{*}, S. Mucherie[#], P.A. Redfern[#], S. Seifert[#],
[#]Argonne National Laboratory, Argonne, USA, ^{*}University of Illinois at Chicago, Chicago, USA,

K. Sell, A. Kleibert, V. von Oyenhausen, K.-H. Meiwes-Broer
Institut für Physik, Universität Rostock, Rostock, Germany,

A. Fraile-Rodríguez
Swiss Light Source & Paul Scherrer Institut, Villigen, Switzerland
E-mail: vajda@anl.gov

The focus of this presentation is on achieving high catalytic activity and selectivity in oxidative reactions by using highly monodisperse sub-nm size atomic metal clusters and few nm size particles. The applied techniques allow for ultimate control of both: surface composition, as well as catalytic particle size and composition – prerequisites in producing highly uniform active sites on technologically relevant supports for basic catalysis studies¹.

The catalytic systems were synthesized by production of sub-nanometer size-selected nanocatalysts in a laser ablation cluster source² and few nm large nanoparticles in an arc cluster ion source (ACIS)³ and their subsequent deposition on a chemically uniform thin alumina film prepared by atomic layer deposition technique on naturally oxidized silicon wafer⁴.

Epoxidation of Propylene. The samples of size-selected silver nanocatalysts were first imaged with scanning electron microscopy (SEM) for uniformity (see Fig.1) and their catalytic properties studied in partial oxidation of propylene under realistic reaction conditions using a unique setup which allows for *in situ* real time monitoring of changes in catalyst size and shape by synchrotron X-ray scattering with simultaneous monitoring of the products formed⁵. Changes in the catalytic particle shape and the evolution of catalyst's reactivity as a function of size and reaction temperature will be discussed and compared to the performance of sub-nm size gold catalysts.

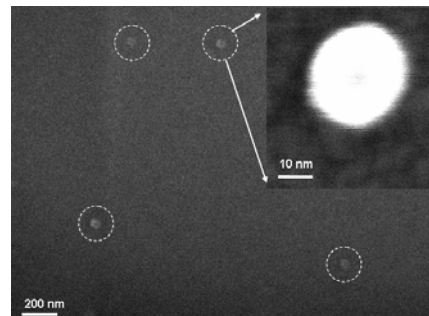


Figure 1. SEM images of the Ag catalyst captured at different magnification.

Oxidative Dehydrogenation of Propane (ODHP). As another example, the use of sub-nm size Pt catalyst supported on mesoporous anodized alumina membranes (AAO) in highly selective and efficient production of propylene from propane will be discussed^{5,6}.

This work was supported by the U.S. Department of Energy (DOE), Office of Basic Energy Sciences, Chemical Science Division operated by the University of Chicago LLC; the Molecular Dynamics and Theoretical Chemistry Office of Air Force Office of Scientific Research (AFOSR); the German Science Foundation (DFG) and the Swiss National Science Foundation.

[1] Bell, A. T., *Science* 299, 1688 (2003).

[2] Vajda, S., Winans, R.E., Elam, J., Pellin, M.J., Seifert, S., Tikhonov, G.Y., and Tomczyk, N.A. *Top. Catal.* 39, 161 (2006)

[3] Methling, R.-P., Senz, V., Klinkenberg, E.-D., Diederich, Th., Tiggesbäumker, J., Holzhüter, Bansmann, J., and Meiwes-Broer, K.H., *Europ.Phys. J D* 16, 173 (2001)

[4] Elam, J. W.; George, S. M., *Chem. Mat.* 15, 1020 (2003).

[5] Vajda, S.; Ballentine, G. E.; Mucherie, S.; Marshall, C. L.; Elam, J. W.; Pellin, M. J.; Lee, B.; Lo, C.-T.; Seifert, S.; Winans, R. E.; Calo, J. M., In *Am. Chem. Soc., Div. Fuel Chem.*, (2007) in press

[6] S. Vajda, M.J. Pellin, L.A. Curtiss, C.L. Marshall, J.W. Elam, J.P. Greeley, S. Mucherie, P.A. Redfern, *to be submitted*