

Mesoporous Au Catalysts: Synthesis, Characterization and Catalytic Oxidation

S.H. Overbury, Sheng Dai, Haoguo Zhu, Byunghwan Lee, Zongtao Zhang
Oak Ridge National Laboratory
M.A. Amiridis, Lorna Soto
University of South Carolina

Recently, Au nanoparticles have been demonstrated to be highly effective catalysts for oxidation of CO if particles are in the proper size range. We have explored various synthetic approaches for preparation of Au catalysts in mesoporous silica and titania supports to achieve uniform Au nanoparticle size distribution. Synthesis methods include wet impregnation, co-assembly synthesis with functional ligands (e.g. monodentate and polydentate amines), ion exchange, and dendrimer assisted assembly in mesoporous silica, titania and zirconia. Most syntheses were done using AuCl_4^- anion precursor. The mesopores are created using various surfactant template molecules, e.g. Pluronic and CTAB. Both size and location of Au nanoparticles in the mesoporous supports are highly dependent on the synthesis protocols. For example, the direct reduction of the monolithic, mesoscopic silica host, uniformly doped by AuCl_4^- gives rise to the aggregation of Au nanoparticles on external surfaces. Because of lack of space confinement on the external surfaces, the growth of gold nanoparticles is uncontrolled, resulting in large gold nanoparticles with broad size distribution. This diffusion of AuCl_4^- can be significantly decreased by post synthesis treatment through the replacement of Cl^- with other ligands (e.g. amines). TEM characterization is consistent with this assertion. These materials have been characterized structurally by BET, TEM, and small angle x-ray scattering. Neutron scattering measurements to determine Au nanoparticle size distributions are planned. FTIR has been used to observe the growth of Au particles as inferred by changes in the CO uptake occurring as a function of reduction temperature. Results of catalytic measurements of the oxidation of toluene and CO will be presented along with characterization by temperature programmed desorption and temperature programmed reduction/oxidation.

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.