536c Sintering and Reactivity Studies on Au Catalyst Supported on Aerosol-Derived Spherical Mesoporous Silica Substrates

John P. Gabaldon, Mangesh T. Bore, and Abhaya K. Datye

Spherical mesoporous silica particles prepared by evaporation induced self assembly (EISA) have a unique internal architecture of coiled hexagonally ordered pores, with no obvious termination of the pores on the external surface. The EISA method, when used with aerosols, can produce highly ordered mesostructured silica particles with a total process time of only several seconds or less. This is much shorter than the typical time scales used for the conventional method that relies on the spontaneous selfassembly of amphiphilic molecules from a bulk solution, with concurrent templating of inorganic precursor species. EISA of aerosols or sprays has several other very attractive features compared to traditional bulk solution methods. The process is a continuous scaleable process that can make particles over a fairly wide size range. Particles produced are generally spherical, which frequently has advantages for subsequent powder processing. Perhaps most interesting from the materials synthesis perspective is the flexibility of the method, which provides for uniform incorporation into every particle of any chemical species that can be dissolved or dispersed into a precursor solution or dispersion. The spherical mesoporous silica particles synthesized by the EISA method can be used as templates to form Pt nanowires. It is remarkable that Pt nanowires can be formed within the closed pores inside these spherical silica particles, where conventional mechanisms of pore filling would not be expected to be operative. The results suggest that the silica walls in these mesoporous silica allow transport of volatile Pt complexes during wet reduction in H2. These silica structures have also assisted in studies involving the reactivity of Au catalyst on CO oxidation, and catalyst sintering. Gold nanopartiles show high reactivity for CO oxidation at low temperature, but only when the Au particles are 5 nm or smaller. Other studies have shown that the reactivity for CO oxidation increases for gold when it is supported on a titanium oxide. The increase in CO oxidation reactivity of gold on a titanium oxide may be due to the titanium oxide preventing the sintering of the gold nanoparticles, the titanium oxide affecting the shape of the gold nanoparticle, and/or the titanium oxides participation in the oxidation as a catalyst. The sintering of the gold particles is also prevented by the unique architecture of the aerosol silica. The curved pores provide a unique geometry with the Au nanoparticles protected inside thin silica walls. The curved pores were also found to be more effective in controlling sintering than the straight pores in MCM-41 type mesoporous materials. The silica walls help to control the size of the Au nanopoarticles and allow the transport of gas phase molecules while retaining the catalyst. The permeability to gases makes these spherical silica particles especially suitable for gas phase catalytic reactions, while at the same time confining metallic particles within the silica pores. Utilizing the results from gold catalyst CO oxidation reactivity and the ability to transport volatile metal into these silica structures, titanium oxide is used to coat the pores of the silica structure to increase the reactivity of CO oxidation on gold catalyst while preventing sintering.