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Thermally reduced gold nanoparticles confined by ordered mesoporous carbon as an efficient catalyst for selective hydrogenation

Gold nanocatalysts represent a new generation of catalysts for the selective oxidation and reduction using molecular O_2 and H_2, showing great potentials for green chemistry. Activated carbons are one of the most frequently used supports in industry. However, activated carbon has been seldom used for gold deposition. Here a coordination-assisted self-assembly approach is adopted for the intercalation of thermally reduced gold nanoparticles inside ordered mesoporous carbon frameworks. An almost complete conversion of benzyl alcohol to benzoic acid is achieved within 60 min over the Au/C catalyst with gold nanoparticles approximately 9.0 nm under 90°C and 1 MPa, using potassium hydroxide as a base. A reduction of gold particle size from 9.0 to 3.4 nm in the catalyst leads to a high activity toward the selective oxidation of benzyl alcohol to benzyl acid and toward the reduction of p-nitrophenol to p-aminophenol at low temperatures such as 0°C. The electronic modification of the d-orbitals of small particles is extremely important for chemisorption of O_2 at atmosphere pressure and low temperatures. Interstingly, thermally reduced Au/C nanocatalyst with gold nanoparticles approximately 2.8 nm is highly active and selective to convert p-chloronitrobenzene and 4-nitrophenol to corresponding amines using H_2 as a reducing agent, reaching an initial reaction rate of 12.7 and 6.5 min^{-1}, respectively. By comparison, the commercial Au/C catalyst is inert under the same reaction conditions. Trapping by the SH-functionalized SBA-15 solids confirms the negligible gold leaching and the heterogeneous active centers for thermally reduced Au/C. Obvious changes are undetected for catalytic performance after five runs. These results indicate that the gold-containing mesoporous carbon catalyst is stable and can be reused. The simultaneous thermal reduction of gold nanoparticles and pyrolysis of the matrix may facilitate the involvement of gold inside the carbon matrix, the modification of carbon atoms on the gold surface, and the reconstruction of the surface induced by CO adsorption. The generation of low-coordinated gold atoms possibly reduces the H_2 dissociation barrier, and can therefore significantly improve the hydrogenation activity.

Biography

Ying Wan received her PhD degree in Industrial Catalysis from the East China University of Science and Technology in 2002. Then, she joined Shanghai Normal University where she was promoted to a full professor in 2006. In 2005-2007, she carried out her postdoctoral research at Fudan University working with Professor Dongyuan Zhao. Currently, Ying Wan is the leader of the Program for Innovative Research Team in University, China. Her research focuses on sintering-, and poisoning-resistance metal nanocatalysts supported on mesoporous carbons, and their applications in green organic synthesis and energy chemistry. She has contributed to about 70 peer-reviewed scientific publications with more than 7000-times citations and 3 books. She has been an associate editor of *Journal of Porous Materials* since 2013.

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