Design, Synthesis, and Characterization of Monometallic and Bimetallic Catalysts

Dissertation Defense by
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Supported metal catalysts for the production of fuels, and pharmaceutical drugs, as well as for energy production and pollution abatement, underpin the global economy and continually upgrade the world’s standard of living. In many applications it is desirable to synthesize the smallest possible metal particles on the catalyst support so as to maximize the number of active sites per mass of metal. We have demonstrated a hybrid method of supported nanoparticle synthesis which combines the advantages of electrostatic adsorption – small particle size and tight size distribution. In the simple incipient wetness impregnation, there is not metal precursor-support interaction, causing extensive sintering and poor size distribution. By inducing electrostatic interaction between metal and support with the strong electrostatic adsorption (SEA) method, small nanoparticles with tight size distribution can be prepared; however, this method requires a large excess amount of the impregnating solution and an additional filtration step, which causes loss of precious metal in excess of monolayer adsorption capacity. Combining the advantages of both methods mentioned, charge enhanced dry impregnation (CEDI) method can be employed.

However, in some cases activity and selectivity improves with increasing particle size and terrace atoms. In the current work, we undertook a broader study of salt addition to investigate the mechanism of size control and to produce larger sized particles with tighter size distributions. By adding halides such as chloride and bromide or other anions like nitrate and citrate during the synthesis of catalysts via the method of charge enhanced dry impregnation (CEDI), metal particles were grown in wide range of 1-7nm. Size control was demonstrated over SiO₂ support. Sizes were verified by consistent XRD, and STEM measurements.

In another work, Ir@Ag bimetallic catalysts with a wide range of Ag coverages on monometallic Ir/γ-A₂O₃ were synthesized in a controlled manner using electroless deposition. Analysis by chemisorption showed unusually high uptakes of hydrogen on Ag-Ir surfaces following thermal treatment at 400 and 600°C in inert gases; computational results suggest that isolated or small ensembles of Ir are capable of multiple hydrogen chemisorption. Two different hydrogenation reactions were used to evaluate catalytic properties of the Ir@Ag catalysts: the hydrogenation of propylene (C₃H₆) which showed very high initial rates and hydrogenolysis of methyl cyclopentane (MCP) to determine the effects of Ag on selectivity to different reaction products. Hydrogenation of C₃H₆ showed very high initial rates indicative of the large reservoir of hydrogen from Ag-Ir.

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