Catalytic Gasification of Carbon

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Research Scope and Objectives

The primary aim of our work is to understand the catalytic effect of metals on the gasification of carbon by various reactants (H_2, O_2, H_2O, CO_2, NO).

Description of Research Effort

Largely as a result of the doctoral work of W. L. Holstein, a working hypothesis has been formulated. With the possible exception of O_2, it says that the role of the metallic catalyst is to break carbon-carbon bonds at the metal-carbon interface with subsequent attack by the gasifying reactant on the reactive carbidic carbon at the metal surface. This work has led to the following publications:

"Platinum and Palladium-Catalyzed Gasification of Carbon by Hydrogen, Water, and Carbon Dioxide", W. L. Holstein, Ph.D. Dissertation, Stanford University, 1981.

"Hydrogenolysis of Carbon and Its Catalysis by Platinum", W. L. Holstein and M. Boudart, Journal of Catalysis 72, 328-337 (1981).

"The Palladium Catalyzed Conversion of Amorphous to Graphitic Carbon", W. L. Holstein, R. D. Moorhead, H. Poppa, and M. Boudart, in Chemistry and Physics of Carbon, P. Thrower, Ed., Marcel Dekker, Inc., New York and Basel; Vol. 18, pp. 139-171, 1982.

"Transition Metal and Metal Oxide Catalyzed Gasification of Carbon by Oxygen, Water, and Carbon Dioxide", W. L. Holstein and M. Boudart, Fuel, 62, 162-165 (1983).

"The Temperature Difference Between a Supported Catalyst Particle and Its Support During Exothermic and Endothermic Catalytic Reactions", W. L. Holstein and M. Boudart, Latin American Journal of Chemical Engineering and Applied Chemistry, in press.

"In Situ Electron Microscopy Study of the Palladium-Amorphous Carbon Interaction in Carbon Dioxide and Oxygen Atmospheres", M. Boudart, W. L. Holstein, R. D. Moorhead, and H. Poppa, submitted to Journal of Applied Catalysis.

"Platinum-Catalyzed Gasification of Carbon by Hydrogen and Water as a Function of Carbon Conversion", J. E. Yie and W. L. Holstein, in preparation.

"Platinum-Catalyzed Gasification of Carbon by Nitric Oxide", K. Lim and M. Boudart, in preparation.

Future Research

In the work described above, it became important to know vibrational frequencies of reactive groups at carbon surfaces, to interpret measured kinetic isotope effects with hydrogen and deuterium. To obtain the data on IR opaque samples, a controlled atmosphere Photo-Acoustic Fourier Transform Infra-Red (PAFTIR) Spectroscopy cell was built and has been checked out in the case of carbon monoxide adsorbed on highly dispersed palladium on silicagel. It was found that CO disproportionates to CO_2 and carbon on very small palladium clusters. This surprising result has been substantiated fully in the doctoral work of Shinichi Ichikawa, sponsored by NASA. In the PAFTIR cell, disproportionation takes place as a result of heating of the sample by the infra-red beam.

At any rate, to interpret PAFTIR data on carbon surfaces, it is necessary to obtain first IR spectra on transparent samples. A study is now underway to accumulate IR data on carbonaceous residues ("coke") produced during hydrogenolysis of n-hexane on Pt/Al_2O_3 catalysts. A collaborative effort has been initiated with Professor G. Froment at the University of Gent, who is investigating systematically the kinetics of coke formation on Pt/Al_2O_3. At Stanford, we have already established by in situ IR spectroscopy that profound changes in IR spectra of residues occur upon evacuation of the sample in the spectrophotometer at the end of a hydrogenolysis run.

This work constitutes the main part of the doctoral work of Francisco Rivera-Latas. When completed, it will provide the supporting data to return to PAFTIR studies of coked samples and carbon surfaces with the ultimate aim to return to catalytic gasification.