Catalytic Role of Mesoporous Silica in the Preferential Oxidation of Monoxide in Excess Hydrogen

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The hydrogen-driven polymer fuel cells are regarded as one of the promising power source due to the high energy efficiency and low CO2 emission. Since the Pt-based anodes are extremely susceptible to the poisoning by CO, one of the key processes for the application is the production of “clean” H2 free of CO (< 10 ppm). Preferential oxidation of carbon monoxide (CO + 1/2 O2 + H2 → CO2 + H2, PROX) in excess H2 is designed to decrease the CO concentration to ppm level.

Well dispersed Pt nano particles with ca. 2.5 nm particle size can be prepared on FSM-16 mesoporous silica by the impregnation and oxidation-reduction methods. The Pt/FSM-16 mesoporous silica (5 wt% Pt loading) gives 95% CO conversion and selectivity at room temperature, and ca. 100% conversion and selectivity at temperature higher than 333 K (O2/CO = 1/2).1) In sharp contrast, the Pt particles supported on amorphous silica counterpart (5 wt% Pt loading) just give 66% CO conversion even at 423 K. This demonstrates a promotional effect of mesoporous silica. However, such high activity cannot be achieved over 5 wt% Pt/FSM-16 in the absence of H2. This suggests a different and non-competitive Langmuir-Hinshelwood reaction mechanism.

The isotope-trace technique is used to clarify the origin of promotional effect. In the adsorption of 13CO or a mixture of 12CO and 13CO, gaseous 12CO2 or 13CO2 peak can be observed without the introduction of gas-phase O2. On the other hand, the C16O18O peak cannot be observed during the sequential adsorption of CO, 18O2 and D2.1) As a result, it can be concluded that the surface silanols groups of FSM-16 trigger the oxidation reaction and was incorporated into the CO2 product. The gas-phase O2 is not directly involved in the oxidation reaction.

Mesoporous silicas are widely applied in catalysis due to their high specific surface area (~ 1000 m2/g) and ordered large pore (2~10 nm). In general, the functions of mesoporous silicas are to provide high dispersion for supported active species or overcome the diffusion limitation for bulky substrate. In this work, we reveal that the promotional effect of FSM-16 is not related with structural parameters, but related with the high reactivity of surface silanols. Further study will reveal more understanding about the catalytic role of mesoporous silica.

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Recent Publications:

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Dr. Huang started his research work on hydrocarbon process during his master course and doctoral student course from 2000 in Laboratory of Applied Catalysis, Dalian Institute of Chemical Physics (DICP), Chinese Academy of Sciences (CAS). After getting Ph. D. degree in DICP in 2006, he came to Catalytic Research Center, Hokkaido University as a postdoc researcher in 2007. His present research interests are related with the synthesis, applications, and mechanistic study of mesoporous material in heterogeneous catalysis.