Surface Modified Catalyst Supports

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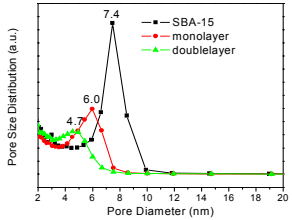
Hydrolytic Surface Sol-Gel Process

- Non-aqueous condensation of metal-alkoxide precursor molecules with surface hydroxyl groups
- Aqueous hydrolysis of adsorbed metal-alkoxide species to regenerate surface hydroxyls

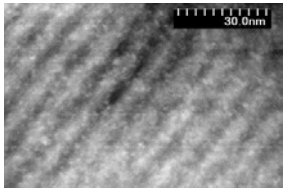
The iteration of the sequential condensation and hydrolysis reactions allows the layer-by-layer coating of a selected metal oxide on a hydroxyl-terminated surface.

Surface Modification of Mesoporous Silica with TiO_2



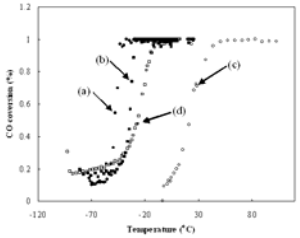
Pore size distribution as function of the number of TiO_2 layer

Preparation of Ultra-Small Gold Nanoparticles



Z-contrast TEM image of ultrasmall gold nanoparticles on ordered mesoporous materials. The bright spots (0.8~1.0 nm) correspond to gold nanoparticles.

Catalytic Activity for Low Temperature CO Oxidation



Comparison of catalytic light-off curves for CO oxidation by gold catalysts on Degussa P-25 and titania-modified SBA-15:
a) as-synthesized Au-P25,
b) as-synthesized Au-TiO_2/SBA-15,
c) 300°C, 8% O_2-He treated Au-P25, and
d) 300°C, 8% O_2-He treated Au-TiO_2/SBA-15.

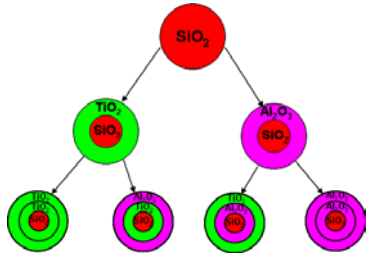
Deposition of Gold on Un-Modified Mesoporous Silica

Dark-field TEM image of aggregated large gold particles on mesoporous silica SBA-15. The resulting catalyst showed no detectable activity for CO oxidation even at temperatures as high as 160°C.

Main conclusions:

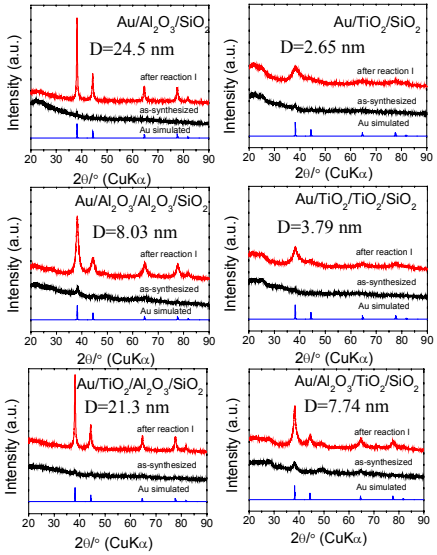
- Surface sol-gel process is an effective method for the modification of silica mesopore surfaces and the tuning of mesopore diameters.
- The ultra-small gold nanoparticles were deposited on the surface-modified mesoporous silica without the constraint of surface isoelectric points.
- The Au nanocatalyst on TiO_2-modified SBA-15 is highly active for CO oxidation.
- This general method can be used to modify the surfaces of mesopores with other metal-oxide monolayer or multilayer species.

Modification of Amorphous Fumed Silica



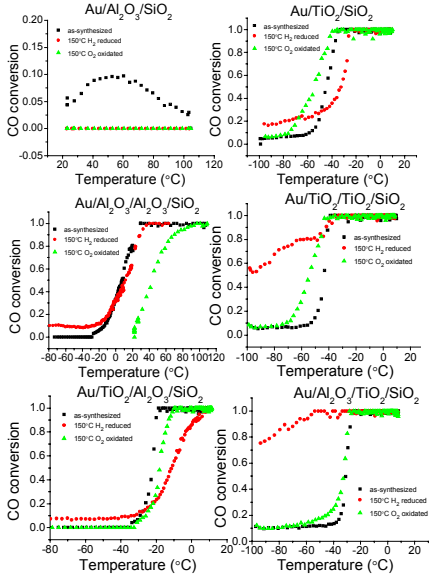
The schematic diagram for the basic coating protocol for ultra-thin TiO_2 and/or Al_2O_3 layers on fumed SiO_2.

Deposition of Gold Nanoparticles on Surface-Modified Fumed Silica



The simulated XRD pattern of Au and experimental XRD patterns for both as-synthesized and that after running the lightoff curve of the as-synthesized catalysts without pretreatment (reaction I). The inset is mean Au particle size after reaction I.

Catalytic Activity for Low Temperature CO Oxidation



The light-off curves of resulting Au nanocatalysts. These light-off curves were measured from the samples both as-synthesized and pretreated under the following conditions: 150°C in H_2 (50% H_2/He) and 150°C in O_2 (8% O_2/He).

Main conclusions:

- The surfaces of amorphous silica materials were tailored by a surface sol-gel process.
- The surface-functionalization sequence is an important factor in determining the catalytic activities of gold nanoparticles deposited.
- The surface functionalization of silica by TiO_2 for the immobilization and the stabilization of gold nanoparticles is very important.

ACKNOWLEDGMENT

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