Preparation of carbon-supported Pt catalysts covered with microporous silica layers using organosilanes: Sintering resistance and superior catalytic performance for cyclohexane dehydrogenation

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A B S T R A C T
Carbon black-supported Pt metal particles (Pt/CB) were covered with microporous silica layers using different organosilanes like methyltriethoxysilane (MTES)/SiO2 (MTES)/Pt/CB and phenyltriethoxysilane (PhTES)/SiO2 (PhTES)/Pt/CB. Pt metal particles in Pt/CB covered with microporous silica layers were stable at high temperature up to 973 K in a hydrogen atmosphere, because the microporous silica layers that wrapped around the Pt metal particles essentially prevent particle sintering. Methyl or phenyl groups were introduced into the silica layers that covered the Pt metal particles using MTES or PhTES hydrolysis. Micropores were formed in the silica layers effectively after thermal treatment at 973 K. The microporous silica-coated Pt catalysts were used as model catalysts for the dehydrogenation of cyclohexane. The microporous silica-coated Pt catalysts with thermal treatment at 973 K exhibited a higher conversion of cyclohexane compared with Pt/CB. Especially, the SiO2 (PhTES)/Pt/CB catalysts showed relatively higher conversion of cyclohexane compared with the other silica-coated Pt catalysts even when the SiO2 loading was high. Microporous silica layers with a larger micropore volume promoted the diffusion of cyclohexane during the catalytic reaction.

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1. Introduction
Highly dispersed precious metal particles on supports have been shown to be active for various catalytic applications such as automotive catalysts [1], the hydrogenation or dehydrogenation of organic compounds [2,3] and electrocatalysts for fuel cells [4]. The efficiency and lifetime of these supported precious metal catalysts are influenced by the stability of the highly dispersed metal particles. However, it has been reported that supported precious metal catalysts are easily deactivated for reasons like the sintering of metal particles because of high reaction temperatures, the surrounding atmosphere [5,6] and the dissolution of metal particles in the polymer electrolyte fuel cell electrodes under severe conditions [7]. The deactivation of supported metal catalysts has been recognized as a big problem in industrial catalysis. Thus, the development of supported metal catalysts with high resistance toward deactivation is required.

The silica coating technique is a promising method to improve the durability of metal particles toward deactivation. In previous work, we have developed supported metal catalysts covered with silica layers by the hydrolysis and condensation of tetraethoxysilane (TEOS) [8–19]. We have demonstrated that the metal particles in these silica-coated metal catalysts have good resistance to sintering even at high temperatures because the metal nanoparticles were physically covered with silica layers. In addition, the metal particles can react with reactant molecules through the silica layers since the silica layers that are wrapped around the metal particles have a porous structure, therefore, the silica-coated metal catalysts can be applied to catalytic reaction. For example, the coverage of Co, Ni and Pt metal particles with silica layers prevents the sintering of their metal particles during hydrocarbon decomposition, which results in the preferential formation of carbon nanotubes or nanofibers with uniform diameters, while metal catalysts that are not covered with silica layers form carbon nanotubes or nanofibers with various diameters because these metal particles are severely aggregated during hydrocarbon decomposition.

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