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学 位 論 文 内 容 の 要 旨

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学 位 論 文 題 名

Oxidation and Hydrogenation Reactions over Supported Platinum Catalysts.

(担持白金触媒による酸化および水素化反応)

Great issues have been faced by today's fresh keeping of plants in horticulture, transportation and refrigerator. Ethylene released from plant hinders plant fresh keeping because it accelerates plant spoiling and decay. Similarly, the formation of the phenylacetylene as a poisoning impurity in styrene feedstocks degrades polystyrene properties in industrial production. Therefore, the main goal of this thesis is the development of active catalytic systems for the complete oxidation of ethylene at low temperature as well as their reaction mechanism and the study of new catalysts for selective hydrogenation of phenylacetylene into styrene.

Chapter 1 describes the general introduction and the purpose of this work.

Chapter 2 demonstrates oxidation of ethylene performed over metal nanoparticles supported on mesoporous silica. A series of catalysts with various metals were supported on MCM-41. Pt was chosen for further study because of its high performance. Pt was supported on different metal oxides to study the effect of support. The Pt/MCM-41 showed high catalytic activity at low temperature towards ethylene oxidation compared to other catalyst. The conversion of ethylene over 1% Pt/MCM-41 at 0°C was >99.8%, which is higher than that of previous reported results.

In Chapter 3, mechanism of ethylene oxidation over Pt catalysts was investigated. Reaction pathways were proposed by performing *in situ* DRIFT experiment. Similar reaction pathways were found on both Pt/MCM-41 and Pt/SiO₂ where C₂H₄ and O₂ quickly react over Pt surface form HCHO. This was followed by decomposition of HCHO into CO and H species, which react with O species to give CO₂ and H₂O, respectively. C₂H₄ and O₂ adsorbed on Pt/ γ -Al₂O₃, Pt/ZrO₂ or Pt/TiO₂, quickly reacted to form acetaldehyde, which was further oxidized into surface acetate and CO species. These CO species were oxidized partially into CO₂ over Pt/ γ -Al₂O₃. However, it was transformed into formate species on Pt/ZrO₂ and Pt/TiO₂. In case of Pt/CeO₂ the CO species were formed when C₂H₄ and O₂ adsorbed on Pt/CeO₂, which was further oxidized into carbonate species as spectator.

The influence of pore structure, surface silanol groups and incorporation of heteroatoms on mesoporous silica was investigated in Chapter 4. In spite of large differences in pore structure and surface silanol groups among all samples, similar ethylene oxidation performance was obtained. However, the doping of heteroatoms in SBA-15 substantially decreases catalytic activity towards ethylene oxidation. DRIFTS investigation showed that the doping of heteroatoms caused strong adsorption of CO and acetate species on the catalyst surface. The adsorbed species needed relatively higher temperature for its complete removal.

In Chapter 5, Pt catalysts were prepared using zeolite-templated carbon (ZTC) as a support and applied to hydrogenation of phenylacetylene. The Pt catalyst on ZTC prepared from a cationic Pt precursor gave highly dispersed Pt nanoparticles and exhibited the highest conversion in the hydrogenation of phenylacetylene.

Finally, Chapter 6 did the general conclusions.