

Busaya Chamnankid 2012: Reduction of NO over Copper Containing Silica-Aluminosilicate Catalysts: Effect of Chitosan Addition, Core-Shell Structure and Metal Incorporation Techniques. Doctor of Engineering (Chemical Engineering), Major Field: Chemical Engineering, Department of Chemical Engineering. Thesis Advisor: Associate Professor Paisan Kongkachuichay, Ph.D. 114 pages.

This research focuses on the synthesis, characterization and activity of porous materials. The purpose of these works is to investigate the novel method to synthesize the mesoporous silica/aluminosilicate composites within one step via a sol-gel process. The pH condition of the hydrolysis-condensation process was found to be the main factor which affected the formation of core-shell structure or infiltration structure of silica and aluminosilicate materials. The mesoporous silica matrix was first developed in a strong acidic condition after that the pH of the solution was adjusted to 6.5 or 11.5. Since the silica solubility was significantly increased with the increasing pH therefore in a strong basic condition (pH 11.5), silica matrix was dissolved back into silicate form and reacted with the tetrahedral aluminates to form an aluminosilicate. Under this condition, the composite of aluminosilicate uniformly infiltrated in silica framework was established. On the other hand, in a slightly acidic condition of pH 6.5, the dense silica matrix could be dissolved only at the edge of the cluster. Therefore the aluminates could be formed and subsequently corner-linked with the unbound silicate starting from the outer layer of the dense silica matrix. As a result, a core-shell structure of silica and aluminosilicate shell was formed.

The synthesized materials were used as catalytic supporters composed of copper metal for NO_x removal process using H₂ as a reducing agent. Furthermore, two different techniques to introduce Cu species into the support including an incipient wetness impregnation method and a substitution technique were also studied. The amount of metal used in this work was also studied, and it was found that the maximum amount of copper was 1.5 wt% that could be incorporated inside the silica/aluminosilicate framework. By using the substitution technique, the copper species were expected to replace aluminum atoms of aluminosilicate. As a result, the tetrahedral coordination of Cu²⁺ atoms were surrounded with four framework oxygen atoms, at this position the Cu²⁺ atoms were being in the most stable site and were difficult to be reduced. On the contrary, in case of impregnation method was used, the Cu species were mainly existed at the outer surface of clusters and were easily reduced into Cu⁺¹. The Cu⁺¹ species were considered to be the active sites for converting NO to N₂. When the effect of chitosan was investigated, it was found that the addition of chitosan created macropores and small domains which provided a higher capillary force between clusters of silica-aluminosilicate composites leading to a higher amount of Cu atoms which were trapped inside the pores. However, the excess Cu atoms and the macropores seem not to be necessary for the enhancing of NO conversion.

Student's signature

Thesis Advisor's signature