XAS Structural Study on Novel Copper-Containing Hierarchical Zeolites

Ko-Kai Chang (張格愷), Ting-Tse Tseng (曾定澤), Albert Chang (張鈞彥), Chia-Min Yang (楊家銘)*

Department of Chemistry, National Tsing Hua University, Hsinchu 30013, Taiwan

Abstract

Hierarchical silicalite-1 materials with high surface areas, high pore volume ratios, high accessibilities, and facile mass transport properties exhibited better catalytic performance than bulk silicalite-1. However, the most common strategy to prepare metal-containing silicalite-1 was impregnation which could not easily control the position and dispersity of metal. Here, we successfully synthesized a new type triblock structure-directing agent (SDA) with metal chelating group in the middle part. The triblock SDA was C_6H_{13}-$N^+(CH_3)_2$-$(CH_2)_6$-$N^+(CH_3)_2$-$(CH_2)_6$-NH-$(CH_2)_2$-NH-$(CH_2)_6$-$N^+(CH_3)_2$-$(CH_2)_6$-$N^+(CH_3)_2$-C_6H_{13}($4Br^-$), abbreviated to N_2-En-N_2. By using this new type SDA, we successfully synthesized hierarchical silicalite-1 comprising highly-branched, self-pillared, orthogonally-stacked and metal chelating agent at the same time. The highly disperse copper complexes between two MFI nanosheets were prepared by aid of chelating part. Average valence state of Cu was between 0 and +1 by X-ray absorption spectroscopy after hydrogen reduction and copper particles were estimated about 0.9 nm by EXAFS analysis. The small copper particles were prepared and partial copper particles bonded to the zeolite surface according to XAS analysis. We applied this catalyst to selective oxidation of propylene catalytic reaction, and high acrolein selectivity at 180 ℃ was obtained.

Keywords – Hierarchical zeolites, Structure-directing agent, Highly disperse copper particles