

Abstract

In the thesis the activity of the Cu/ZnO composite materials was investigated with various microscopic and spectroscopic methods. The investigated Cu/ZnO composites were further used as catalysts for CH₃OH synthesis at pressure 1 bar and temperature 250 °C and the obtained results revealed, that the synthesized catalysts exhibited superior activity and stability under synthesis conditions in comparison with the Cu/ZnO/Al₂O₃ commercial catalysts. Novel catalysts were prepared in a way, where Cu crystallites were deposited on the surface of prior synthesized ZnO support.

Using EXAFS analysis it was shown, that during the catalysts' reduction and subsequent CH₃OH catalytic synthesis a certain amount of ZnO was reduced into metallic Zn, which further diffused into Cu crystal lattice, thereby forming an alloy. The EXAFS revealed that the alloy Cu-Zn present in the investigated catalysts can be described as Cu_{0.2}Zn₇ with hexagonal close packed crystal structure. In the prepared catalysts around 18% Cu is bonded into alloy Cu-Zn, while the amount of the alloy in the commercial catalysts is significantly lower (around 11% of Cu is bonded with Zn). Therefore, the relative high activity for the CH₃OH synthesis of the prepared catalysts compared to the activity of commercial catalysts was assigned to rather high concentration of the alloy present in the prepared catalysts. In such catalysts the spillover mechanism is distinct, since the Cu and ZnO components are in intimate contact.

Moreover, it was shown with the microscopy TEM that the Cu crystallites in the prepared catalysts are rich with planar defects such as twin boundaries or stacking faults, running throughout the particles. It was further revealed with the microscopy TEM, that crystal planes reorient where the twin boundary disturbs the ideal Cu lattice. A shear-strain map across a defect was constructed in order to confirm that the strain is localized around the defects. As known from the literature, surface defects are much more active for the adsorption of reaction molecules in comparison with ideal terraces. Consequently, relative high activity of the prepared catalysts can be assigned both to increases amount of the alloy Cu-Zn, as well as to high abundant concentration of defects present in lattice Cu.