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Au/ZnO as catalyst for methanol synthesis: The role of oxygen vacancies

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ABSTRACT

Gold catalysts supported on zinc oxide with Au loadings of 1, 2, and 3 wt% were prepared by the colloidal deposition method and applied in methanol synthesis in CO_2-free ($CO + H_2$) and in CO_2-containing ($CO + CO_2 + H_2$) synthesis gas. The characterization by transmission electron microscopy and X-ray diffraction before and after the catalytic high-pressure tests demonstrated a very narrow and uniform Au particle size distribution and a high stability against sintering. Reactive frontal chromatography (RFC) experiments with N_2O were performed aiming at the titration of oxygen vacancies. With increasing Au loading, the amount of consumed N_2O increased in good correlation with the number of Au perimeter atoms present in the Au/ZnO catalysts suggesting an enhanced formation of oxygen vacancies at the Au/ZnO interface. In both synthesis gas mixtures the presence of the Au particles led to an increased activity compared with pure ZnO. All Au/ZnO samples exhibited higher catalytic activity in the absence of CO_2, as had been observed for pure ZnO with similar apparent activation energy. It is concluded that oxygen vacancies in ZnO are also the active sites in methanol synthesis over Au/ZnO, and that the presence of the Au particles enhances the number of exposed oxygen vacancies in ZnO, presumably located at the interface region.