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PREPARATION AND CHARACTERIZATION OF γ-Al2O3 DOPED WITH SELECTED ELEMENTS AND CORRELATING SURFACE PROPERTIES WITH THE CATALYTIC ACTIVITY IN METHANOL DEHYDRATION TO DIMETHYL ETHER

by

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Abstract

The increasing demand for energy is associated with challenges that include environmental concerns and limited reserves. Dimethyl ether, DME, which can be obtained from different feedstocks, including natural gas and biomass, has recently been recognized as an ultraclean environmentally friendly fuel due to the fact that it possesses unique characteristics that make it an efficient alternative fuel for diesel fuel engines. In addition, DME is an industrially important intermediate for a variety of chemicals. A promising potential route for dimethyl ether production is catalytic dehydration of methanol over solid acid catalysts. Therefore, exploring new solid acid catalytic materials and understanding the mechanistic steps of methanol adsorption on their surfaces is of great importance for developing modified efficient catalysts for this process. In the present work, solid acid catalysts based on modified γ-Al_2O_3 were prepared by sol-gel method and were studied as catalysts for methanol to dimethyl ether conversion. The main focus of the present thesis is to investigate the effect of selected metal dopants on the surface chemical properties of γ-Al_2O_3, especially acid-base characteristics, and to correlate these effects with their catalytic activity in dehydration of methanol to DME. The selected dopants include transition metal ions with different *d*-configurations and different oxidation states, such as Ti(IV), V(III) and Ni(II) to elucidate any possible electronic effect on the alumina surface chemical behavior. Other dopants that were anticipated to affect the acid-base properties of alumina were also tested include Si and Mg. The prepared catalysts were characterized by various physical and chemical techniques including adsorption of probe molecules, namely ammonia and methanol. The study showed very promising results where doping γ-Al_2O_3 resulted in significant textural and chemical modifications including an enhanced overall surface acidity. The catalytic activity study showed that the incorporation of certain concentrations of Ti(IV) and Ni(II) ions in the γ-Al_2O_3 matrix resulted in an enhanced catalytic activity. The catalytic activity of the catalysts was correlated with their textural, chemical, and structural modifications resulting from the presence of the dopant ions. In addition, comparison between the studied alumina-based solids and selected ZSM5 zeolites showed that the acidic character of the OH groups on their surfaces vary and therefore, different routes of methanol adsorption and dehydration were proposed for the two types of materials. Methanol adsorption and dehydration was proposed to be associative on the surface of ZSM5 zeolites, where Brønsted acid sites played a key role in adsorption and dehydration reaction. On the other hand, dissociative adsorption on Lewis acid-base pairs dominates the interactions with γ-Al_2O_3-based solids.

Keywords: methanol dehydration, methanol adsorption, dimethyl ether, alternative fuel, acid catalyst.