Novel catalyst with layered structure: [Al]- and [V]- magadiite

Gizeuda L. Paz[1](PG), Heloise O. Pastore[1] (PQ)*

*[1]Institute of Chemistry, University of Campinas, Monteiro Lobato St., 270, 13084-971, Campinas, SP, Brazil (gpmmm@iqm.unicamp.br)**

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Abstract

[M]-magadiites were used in catalytic ethanol conversion monitored by in situ FTIR: ethylene and acetaldehyde were the products obtained.

Introduction

Al and V-modified magadiite (Si/Al=15 and Si/V=33 molar ratios) were obtained by Seed Induced Crystallization (SIC)[1] method and the catalytic performances of these materials were investigated on ethanol conversion reactions. This reaction can take place by two competitive paths accompanied by a side reaction (reactions 1-3): $C_2H_5OH \rightarrow C_2H_4 + H_2O$ (1), $2C_2H_5OH \rightarrow C_2H_5OC_2H_5 + H_2O$ (2) and $C_2H_5OH \rightarrow CH_3CHO + H_2$ (3). Reaction (1) is dominant at high temperatures while reaction (2) is observed at low temperatures. Also at high temperatures acetaldehyde can be obtained as the product of a side reaction (3). In this work, the reactions were monitored using in situ FTIR of surface and gas phase at various temperatures.

Results and discussion

The samples were dehydrated and calcined before ethanol adsorption. After ethanol adsorption, the samples were treated at 100, 150, 200, 250, 300 and 350 °C and remained at each of these temperatures for 1 h. Fig. 1 shows the spectra of the gas phase of the reaction performed on H^+-[Al]-magadiite (A) and H^+-[V]-magadiite (B). Hardly any difference can be observed in the gas phase of the cell from room temperature up to 250 °C in both samples: the bands observed are typical of ethanol (1241 δ(CH), 1085-1048 and 880 cm^{-1} ν(C-O/C-C)). When the temperature arrives at 300 °C, and after at 350 °C, new bands appear at 949 cm^{-1} (curves (f) and (g)) assigned to CH_2 wagging vibrations of ethylene[2] and 1760-1730 cm^{-1} assigned to νC=O (curves (f) and (g)) of acetaldehyde[3]. The presence of water becomes evident at 1640 cm^{-1} (Figure 1A, curve (g)) due to its production in reaction (1). It is necessary to point out that liquid water and ethanol (at 1085, 1048 and 880 cm^{-1}, Figure 1A, curves (f) and (g)) were observed after the reaction at 300 °C and 350 °C due to a film formed in the cell windows.

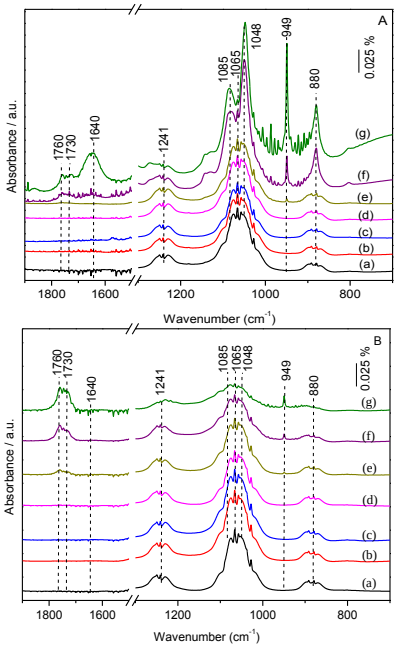

Figure 1. FTIR spectra of gas phase after ethanol was adsorbed on H^+-[Al]-magadiite (A) and H^+-[V]-magadiite (B) at (a) room temperature, (b) 100 °C, (c) 150 °C, (d) 200 °C, (e) 250 °C, (f) 300 °C, and (g) 350 °C.

Conclusions

The activity of these solids in ethanol conversion is connected to the framework heteroelement: ethylene is largely produced on H^+-[Al]-magadiite while acetaldehyde can come either from ethanol oxidation catalyzed by siloxane bridges with production of H_2 or by V-oxidizing in H^+-[Al]-magadiite and H^+-[V]-magadiite, respectively.

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