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Production of Ethylene by Oxidative Dehydrogenation of Ethane Over Tungsten Bronze Catalysts

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ABSTRACT

Supported and unsupported vanadium-promoted tungsten (hexagonal tungsten bronze) HTB catalysts were synthesized by the reflux method. HTB catalysts studied as references were prepared by the hydrothermal method. The synthesized catalysts were characterized by XRD, Raman, TPD-NH3, TPR, EPR, XPS, UV-vis, BET). The catalytic performances of these materials are tested in the oxidative dehydrogenation reaction of ethane.

Keywords: hexagonal tungsten bronze, vanadium oxide, ODH ethane, ethylene

1. Introduction

Catalysis by metal oxides is widely used, particularly in the partial oxidation of hydrocarbons. These metal oxides can be used in bulk or supported. The presence of a support is of economic interest in order to reduce the quantity of the active phase. These supports can also have an effect on the catalytic performance by modifying the exposure of these sites and modifying their nature by interaction with the support. Mixed metal oxides bronzes have attracted the attention in the last years since they can be effective catalysts in acid and/or oxidation reaction. In general, the physic-chemical properties (acid-base or redox) can be strongly modified by tailoring the catalyst preparation procedure or the catalyst composition, which can also determine the crystal size, the local structure or the electronic/catalytic performance. Metal oxide bronzes which are defined as ternary compounds of the type $A_xX_yO_z$, present a variety of physical and chemical properties. Tungsten-based bronzes have extensively studied as photocatalysts, but they have been less studied in catalysis. Vanadium oxide supported on alumina is active and selective in the ODH of ethane. The oxidative dehydrogenation (ODH) of ethane is carried out on a solid catalyst in which the ethane reacts with an oxidizing species typically oxygen.

2. Experimental

Supported and unsupported vanadium/tungsten bronzes are synthesized by the reflux method. W-V-O materials are synthesized from gels which are prepared by dissolving ammonium para-tungstate in de-ionized water at a temperature of 80°C. Vanadyl sulfate is then added and the mixture is stirred for 10 min. Oxalic acid is also added to some catalysts. The reflux time is 24 hours. The solid obtained is filtered, rinsed and dried at 100° C. for 16 h. The mixed W-V-O oxides supported on alumina are prepared by the reflux method, alumina Al_2O_3 (commercial) is added to the gel. These catalysts are heat treated at 600 °C for 2 h in a flow of N_2.

3. Results and discussion

Results of the catalytic test in oxidative dehydrogenation reaction over the synthesized materials show that W-V oxides bronzes supported on γ-Al_2O_3 are more selective for the oxidative dehydrogenation of ethane to ethylene than the corresponding unsupported ones. It can be observed that the catalytic performance strongly depends on the crystalline structure but also on the nature of V-sites. Catalysts W-V oxide with hexagonal tungsten bronzes have been tested in oxidative dehydrogenation of ethane. This material show a high selectivity in ethylene by comparison with the conventional vanadium oxide supported catalyst. The high dispersion of vanadium sites (V-O-W pair) can lead to a low combustion of ethylene. The catalytic

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and the characterization results observed in the case of the tungsten hexagonal bronzes, show that the catalytic performance depends on the crystalline structure. It was also found that the preparation procedure and the sequence in the incorporation of the precursors appeared to be effective in obtaining solids presenting the h-WO_3 phase.

Table 1: *Catalytic results for the oxidative dehydrogenation of ethane on V-based catalysts.*

Catalyst	Preparation method	Temp. (°C)	Conv. (%)	S-C_2H_4 (%)	S-CO_2 (%)	S-CO (%)
WV-C-1/AL	Reflux	502	13,5	79,6	2,0	18,4
WV-C-2/AL	Reflux	509	24,1	71,4	2,6	26
		518[b]	42	55		
WV-C-3/AL	Reflux	517	45,4	44,9	4,8	50,3
WV-C-2	Reflux	507	1,3	65,9	3,6	30,5
HT-WV-2	Hydrothermal	507	7,4	49,8	4,8	45,4
2V/AL	Impregnation	456	23,4	36	7,9	57
2VOx/AL	Impregnation					
8W/AL	Impregnation	525	77,9	10,4	9,8	79,7
2VOx/8WOx/AL	Impregnation					
8V/W/AL	Impregnation	(505)	70	14,6	8,8	76,6

4. Conclusion

New types of W-V oxide catalysts with hexagonal tungsten bronze structure are tested in ethane ODH. Compared to conventional supported vanadium oxide catalysts, they show high ethylene selectivity at the same ethane conversion. The high dispersion of vanadium sites (like V-O-W peers) promotes low ethylene combustion. Both catalyst preparation and V content greatly influence their catalytic performance in ethane ODH.

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