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Catalytic Oxidation of n-Butanol over Mg₄Ce₂ Hydrothalcite based System: Effect of Water Vapor and Volatile Organic Compounds Mixture

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ABSTRACT

This study investigates Mg₄Ce₂ layered double hydroxides (LDH), focusing on its structural and catalytic properties. The sample was characterized by XRD, BET, Raman, XPS, and DTA/TGA techniques. Its performance in the oxidation of n-butanol, including the influence of water vapor and the presence of ethyl acetate was studied. Results show promising catalytic activity of Mg₄Ce₂ hydrotalcite, with total butanol conversion achieved at 250°C. However, the presence of water vapor and the coexistence of ethyl acetate and butanol lead to slight decreases in catalytic efficiency, with a more pronounced inhibitory effect observed when both compounds are present together.

Keywords: Oxidation, n-Butanol, Volatile organic compounds, Cerium, Hydrotalcite.

1 Introduction

Industrial processes release a multitude of volatile organic compounds (VOCs) into the atmosphere. These emissions have detrimental effects on human health and contribute to air pollution. To simulate realistic VOC treatment conditions, water must be introduced into the initial VOC mixture due to the typical humidity of the surrounding air. Water generally acts as an inhibitor of VOC oxidation on noble metal catalysts supported on metal oxides, although the extent of this inhibition depends on the specific VOC characteristics [1-3]. Various catalysts based on transition metals are employed for this reaction. However, the oxides of these metals possess certain disadvantages, like poor reducibility, inadequate distribution and dispersion of the active phase, leading to decreased catalyst stability and activity. The use of hydrotalcite as a precursor has proven highly effective in obtaining materials with excellent catalytic behavior and improved dispersion of the active phase. Furthermore, cerium, known for its substantial oxygen storage capacity, is suggested as a high-performance metal catalyst for VOC elimination.

2 Experimental

Mg₄Ce₂ hydrotalcite (Mg₄Ce₂- HT) was synthesized as precursor through co-precipitation. It was characterized by several techniques (BET, XRD, Raman and XPS) and evaluated in the total oxidation of n-butanol at 130°- 280°C. The impact of water vapor and the ethyl acetate/n-butanol mixture on enhancing butanol conversion into oxidation products was also investigated.

3 Results and Discussion

Characterization results confirmed the presence of characteristic XRD patterns for the hydrotalcite phase in the Mg₄Ce₂-HT system, which aligned with the Raman spectroscopy findings. The hydrotalcite phase remains conserved even after calcination at 280°C. Analysis of textural properties using BET and BJH method reveals that the studied system has a good specific surface and the nitrogen adsorption isotherm obtained is type IV, it is characteristic of a mesoporous solid. Catalytic results revealed excellent activity of Mg₄Ce₂-HT in the complete oxidation of butanol. It achieved total butanol conversion with over 82% CO₂



yield at 250°C. The presence of water vapor in the reaction mixture negatively impacted the catalytic activity of Mg₄Ce₂-HT in butanol combustion, causing a slight shift in the butanol conversion profile towards higher temperatures, requiring an additional 20°C for equivalent conversion without water vapor. The oxidation of the n-butanol/ethyl acetate mixture using Mg₄Ce₂-HT was evaluated at a 1:1 molar ratio, with the feed composition maintaining a 1% volume of total VOCs. A mutual inhibiting effect between the two compounds was observed, with butanol combustion exhibiting a more pronounced effect. Specifically, the presence of ethyl acetate significantly hindered the rate of butanol oxidation, leading to higher conversion temperatures of 50-60°C compared to the oxidation of butanol alone. Conversely, the oxidation of ethyl acetate was only slightly affected by the presence of butanol.

4 Conclusions

In this study, Mg₄Ce₂-HT was successfully synthesized by co-precipitation method. It exhibited interesting properties for the total oxidation reaction of n-butanol. Adding water vapor to the reaction mixture slightly decreased the butanol oxidation, while the presence of ethyl acetate and butanol together exhibited a mutual inhibitory effect, with butanol demonstrating a more prominent influence.

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