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Evaluation of Ga/HZSM-5 catalysts prepared by atomic layer deposition on the ethane dehydroaromatization reaction

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Introduction

Ethane is one of the main components of natural gas liquids and, with the increasing exploration of shale gas, represents an abundant source of raw material [1]. Ethane dehydroaromatization (EDA), a catalytic process for converting ethane into aromatics, provides an alternative to traditional naphtha cracking to produce benzene, toluene, and xylenes (BTX). It is known that ZSM-5 catalysts modified with active metals are effective for the EDA reaction due to the three-dimensional micropore channel and strong Brønsted acid sites (BAS) presented in the MFY-type zeolites [2].

Gallium catalysts supported on zeolites have been investigated for alkane dehydrogenation and dehydroaromatization and have shown promising results [3]. These catalysts are typically prepared using traditional synthesis methods, such as impregnation, ion exchange and precipitation. However, the nonuniformity of active sites generated by these methods complicates the balance of key properties like activity, selectivity, and stability. Atomic layer deposition (ALD) has proven effective in controlling the deposition of metal and metal oxide sites, thereby enhancing these catalytic properties [4].

Therefore, the present study investigates the catalytic activity of Ga/HZSM-5 catalysts prepared by ALD for the EDA reaction. Additionally, the catalysts were characterized to identify the Ga species present and compared with those prepared by traditional incipient wetness impregnation (IWI) to evaluate the influence of the synthesis method.

Materials and Methods

The catalysts were synthesized by atomic layer deposition (ALD) using a GEMSTAR ALD System (Arradiance). Trimethylgallium was employed as the Ga precursor, and O₃ was used as an oxygen source. A varying number of ALD cycles were performed to obtain catalysts with different metal loadings. For the catalysts prepared by incipient wetness impregnation (IWI), gallium nitrate hydrate was used as the precursor. All catalysts were supported on commercial ZSM-5 zeolite (SiO₂/Al₂O₃ = 50) from Zeolyst. The catalytic tests were conducted in a fixed-bed tubular reactor with online analysis via GC chromatography at 600°C and atmospheric pressure.

Results and Discussion

The ethane conversion and BTX selectivity of the catalysts prepared by ALD with 1, 3, 5, and 10 cycles are presented in Figure 1. It can be observed that as the number of ALD cycles increases, resulting in higher Ga content, the ethane conversion also increases. This indicates that ethane dehydrogenation to ethylene, the first step in the EDA reaction, occurs on metallic sites. On the other hand, the stability of the BTX selectivity decreases with increasing Ga loading. All catalysts show similar selectivity at the beginning of the reaction; however, it decreases rapidly, especially for catalysts with higher Ga loading. The main cause of this deactivation is coke deposition, which blocks the entrance to the zeolite channels and prevents ethylene molecules from accessing the Brønsted acid sites, where BTX formation takes place, leading to a rapid decline in BTX selectivity. Therefore, there is a trade-off between Ga loading to achieve higher ethane conversion and maintaining catalyst stability.

Catalysts prepared by IWI were also tested to evaluate the effect of the synthesis method. Although both methods lead to the majority of Ga being deposited as gallium oxide, ALD-prepared catalysts seem to exhibit better stability at low metal loadings, suggesting that a more homogeneous Ga distribution could be key to developing a more selective and stable catalyst for this reaction.

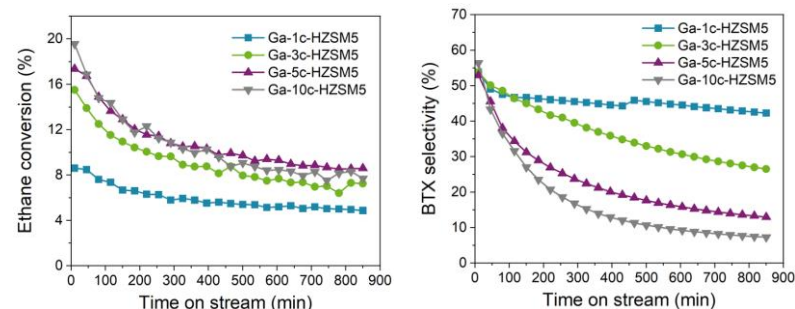


Figure 1. Ethane conversion (left) and BTX selectivity (right) of Ga/HZSM-5 catalysts prepared by ALD with 1, 3, 5 and 10 cycles of Ga deposition on the ethane dehydroaromatization reaction.

Significance

A direct process to obtain BTX from ethane is highly desired, the investigation of catalysts throughout new synthesis methods may be the key to making the process viable. Gallium deposited by ALD on zeolites has been little reported in literature and presents potential of exploration.

References

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