Effect of zeolite acidity on pre-treated Fe-based catalysts for e-fuel production via CO_2-modified Fischer-Tropsch Synthesis

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Significance and Relevance

This work is significant in advancing CO_2-modified Fischer-Tropsch synthesis (FTS) by exploring the role of zeolite acidity in influencing Na-Fe_3O_4 catalyst selectivity and product distribution. By combining extensive characterization techniques, including Detailed Hydrocarbon Analysis (DHA) and Mössbauer spectroscopy, with chemometric tools like Principal Component Analysis (PCA), the study provides a comprehensive understanding of how catalyst phase composition and zeolite choice impact hydrocarbon profiles. These insights are essential for developing tailored catalysts that optimize CO_2-to-hydrocarbon conversion, promoting sustainable fuel production.

Preferred and 2nd choice for the topic: CO_2 utilization and recycling / Sustainable and clean energy production and transport
Preferred presentation: Oral preferred or Short Oral

Introduction and Motivations

The increasing demand for sustainable fuels has accelerated research in CO_2 conversion technologies, particularly in the Fischer-Tropsch synthesis (FTS) process for generating liquid hydrocarbons. This study investigates the impact of zeolites with different acidities on a Na-Fe_3O_4 catalyst in CO_2-modified FTS. Specifically, the research explores how different zeolites can enhance the conversion efficiency and selectivity of the catalyst, offering new insights into optimizing the CO_2-to-hydrocarbon pathway.

Materials and Methods

The Na-Fe_3O_4 catalyst was synthesized and combined with commercial zeolites, then subjected to optimized pre-treatment conditions based on prior work [1]. Catalyst and product characterizations were performed using XRD, Mössbauer spectroscopy, and Detailed Hydrocarbon Analysis (DHA), to examine bulk composition, surface properties, and hydrocarbon distributions. Principal Component Analysis (PCA) was applied to the DHA data to reveal correlations between molecular compositions and test conditions, highlighting how different zeolites influence hydrocarbon profiles.

Results and Discussion

Two pretreatment methods were evaluated: H_2 activation (pretreatment A) and H_2 activation followed by carburization (CO/H_2) at 400°C (pretreatment AC400). The H_2 activation facilitated the formation of Hägg carbide (χ-Fe_5C_2), producing hydrocarbons mainly in the gasoline range. In contrast, the AC400 pretreatment promoted the formation of ε'-$Fe_{2.2}C$, a more active phase selective for longer-chain hydrocarbons[2]. Both pretreatments were combined with four MFI zeolites with increasing acidity (SiO_2/Al_2O_3 ratios of 23, 38, 80, and 117). The addition of zeolites shifted the oil product distribution from olefin/naphthene-rich to iso-paraffin/aromatic-dominant compositions (Fig. 1) and increased the C6+ yield by at least 25%[3]. DHA identified and quantified over 150 hydrocarbons, clarifying how zeolite acidity influences reaction mechanisms. Chemometric analysis, particularly PCA, identified recurring patterns in hydrocarbon formation associated with different SiO_2/Al_2O_3 ratios. Mössbauer spectroscopy further clarified the zeolites' influence, showing changes in both product distribution and phase transformations during FTS. This study demonstrates the importance of integrating multiple characterization techniques, including detailed analysis of both liquid products and catalyst structures, to provide a comprehensive understanding of CO_2-modified FTS. The synergistic use of DHA, PCA, and Mössbauer spectroscopy offers a novel and thorough approach for optimizing catalysts and reaction pathways in sustainable fuel production.

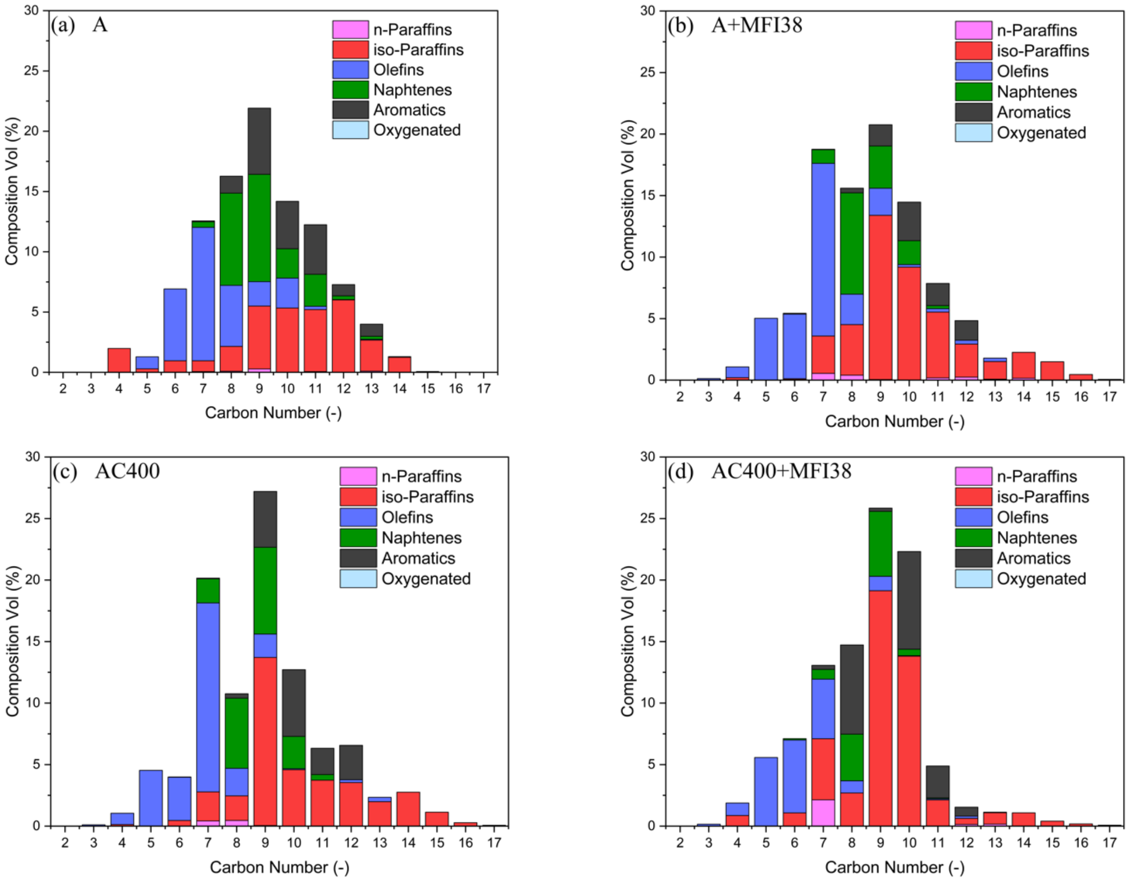

Figure 1 Detailed Hydrocarbon Analysis profiles for samples pretreated with H_2 activation alone (a), H_2 activation coupled with MFI38 (b), H_2 activation followed by 400°C carburization alone (c), and H_2 activation with 400°C carburization coupled with MFI38 (d).

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