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Towards Machine Learning-driven Catalyst Design and Optimization of Operating Conditions for the Production of Jet Fuel Via Fischer-Tropsch Synthesis

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Fischer-Tropsch synthesis (FTS) offers a promising route for producing sustainable jet fuels from syngas. However, optimizing the catalyst design and operating conditions to maximize the desired C_8-C_{16} jet fuel range is a challenging task. This study introduces the application of a machine learning (ML) framework to guide the design of Co/Fe-supported FTS catalysts and operating conditions for enhanced fuel selectivity. A comprehensive dataset was constructed with 21 input features spanning catalyst structure, preparation method, activation procedure, and FTS operating parameters. The random forest ML algorithm was evaluated for predicting CO conversion and C_8-C_{16} selectivity using this dataset. Feature engineering identified the most significant descriptors influencing performance. A principal component analysis reduced the dataset dimensionality prior to ML modelling. The random forest algorithm achieved high prediction accuracy for the conversion of CO (R^2 = 0.92) and C_8-C_{16} selectivity (R^2 = 0.90). In addition to confirming the known effects of operating conditions, key roles of Co/Fe-supported properties were elucidated. This ML framework provides a powerful tool for the rational design of FTS catalysts and operating windows to maximize jet fuel productivity.

1. Introduction

The growing global energy demand and concerns over climate change have spurred intense research into developing renewable and sustainable energy resources. Fischer-Tropsch synthesis (FTS), a catalytic process that converts syngas – a mixture of hydrogen and carbon monoxide (CO + H_2) into light olefins, liquid fuels, and value-added chemicals, has received renewed attention in recent decades (Grainca et al., 2023). Syngas can be derived from sustainable processes such as biomass gasification, making FTS an attractive route for producing clean transportation fuels and chemicals (Sineva et al., 2023). The product distribution in the FTS process is heavily influenced by the catalyst composition and formulation, as well as the reaction conditions employed. Cobalt and iron are two of the most widely utilized active metal components for catalyzing the hydrogenation of syngas in FTS catalysts (Kriventceva et al., 2013).

The design of an FTS catalyst involves several critical factors, such as the catalyst preparation method, the choice of support material, the addition of promoters, and the calcination and reduction treatments applied. Due to the complex nature of FTS, which involves multiple interconnected reaction steps, accurately predicting the product selectivity poses a significant challenge. In this context, modeling techniques offer an alternative approach to tackle these difficulties by enabling the prediction of optimal catalyst structures and operating conditions tailored to the desired product distribution (Alayat et al., 2019). Machine learning (ML) methodologies have found widespread application in deciphering the intricacies of many industrial applications (Dorneanu et al., 2022), including catalyst screening (Tomacruz et al., 2022) or catalytic reactions characterized by complex reaction mechanisms, such as the FTS process. However, the majority of prior studies in this domain have concentrated either on specific catalyst formulations or on operating conditions, failing to adopt a holistic approach that simultaneously considers the effects of both catalyst structure and process parameters. Notably, the influence of catalyst support materials, particularly zeolites, Al_2O_3, and SiO_2, on the FTS reaction has not been extensively investigated using ML techniques.

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One of the challenges in investigating the influence of catalyst support materials on the FTS process towards jet fuel production using ML techniques is limited availability of high-quality experimental data. ML models require large datasets for training, but there is a scarcity of comprehensive experimental data on the performance of various support materials for jet fuel production via the FTS process. Most studies focus on specific support materials or operating conditions, making it difficult to develop generalizable models. However, with the increasing availability of experimental data, advancements in computational resources, and interdisciplinary collaborations, machine learning is expected to play a more significant role in optimizing catalyst support materials for the FTS process towards jet fuel production in the future. This represents a significant gap in the current understanding of the process, as these support materials can potentially play a crucial role in modulating the reaction mechanism and product selectivity (Teimouri et al., 2021). Zeolites, Al_2O_3, and SiO_2 possess unique structural and textural properties that can significantly influence the FTS reaction mechanism and product selectivity towards jet fuels. These support materials can modulate metal-support interactions, alter the electronic properties of the active phase, and introduce acidity or basicity, thereby affecting the product distribution. The choice of support material and its characteristics, such as specific surface area, pore size, and surface acidity, play a crucial role in tuning the selectivity toward jet fuels in FTS.

Previous studies have explored using ML approaches to investigate product distribution in the FTS process. However, a comprehensive model that simultaneously considers the effects of operational conditions and catalyst structure, including catalyst synthesis, reduction, and the FTS process itself, specifically focusing on zeolites, Al_2O_3, and SiO_2-based supports towards jet fuels, has been lacking (Sun et al., 2011). Teimouri et al. (2024) developed an ML framework to predict the performance of carbon-supported Fe/Co catalysts for FTS of clean liquid fuels. Their research offers a strategic plan for engineering highly effective catalysts derived from carbon materials. Essentially, their study provides guidelines for the systematic development of carbon-based catalytic systems geared towards producing renewable or non-petroleum-based fuels for various modes of transportation. Thus, this study aims to develop a comprehensive ML framework to predict the FTS performance of zeolite, alumina, and silica-supported catalysts towards jet fuel production by considering the simultaneous effects of catalyst structure, preparation method, activation procedure, and operating conditions. Ultimately, this work will provide valuable insights for the rational design of efficient zeolite, alumina, and silica-based catalysts tailored for specific FTS product streams towards jet fuel.

2. Methodology

Data curation is a critical step in developing an effective ML algorithm, as the quality of the input data significantly affects the model's performance. This work curated a dataset from 10 recent publications (comprising 43 datasets) that investigated the use of zeolite, alumina, and silica supports for Fischer-Tropsch synthesis (FTS) catalysts toward jet fuel production. The compiled dataset was first statistically analyzed to ensure its robustness and then mechanistically evaluated to understand the underlying relationships between the input features and the desired output. Furthermore, 21 relevant descriptors related to catalyst formulation and operational conditions were identified as input features for the ML models. Given the importance of producing sustainable jet fuels via FTS, the C_8-C_{16} selectivity (jet fuel range) and CO conversion were set as the target responses. Subsequently, these 21 input features were classified into two main categories: catalyst formulation and operational conditions. The catalyst formulation descriptors included the support material (zeolite, alumina, or silica), active metal (Co or Fe), promoter type and loading, catalyst preparation method, calcination temperature, and textural properties (surface area and pore size). The operational condition descriptors encompassed parameters related to catalyst reduction (temperature, pressure, and gas composition), and FTS reaction conditions (temperature, pressure, H_2/CO ratio, and space velocity).

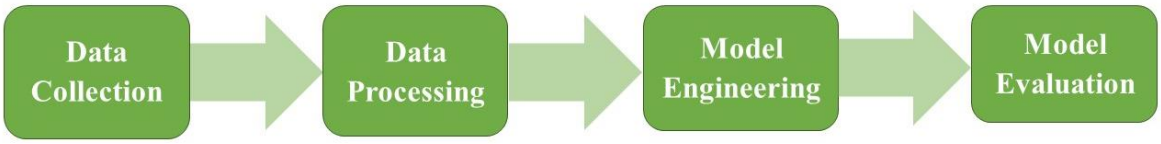

Figure 1: The stages of the ML framework

To ensure the quality and completeness of the dataset, rigorous data cleaning and preprocessing steps were performed, as illustrated in Figure 1. These steps included handling missing values through appropriate

imputation techniques, identifying and addressing outliers, and applying feature scaling and encoding to ensure consistent data representation for the ML algorithms.

3. Results

3.1 Analysis of input data

The relationships between input features (catalyst descriptors and operational conditions) and output variables (C_8-C_{16} selectivity, CO conversion) were analyzed using Spearman rank correlation and principal component analysis (PCA) (Figure 2). Positive correlations were found between reduction atmosphere and active metal type (Fe requires H_2/CO, Co requires H_2), as well as between larger catalyst pore size and improved C_8-C_{16} selectivity due to better diffusion. A negative correlation was seen between promoter loading and C_8-C_{16} selectivity, suggesting that over-promotion can block active sites. PCA revealed the first 6 principal components accounted for over 80 % of variance, with active metal/promoter in PC1, reduction conditions in PC2, catalyst synthesis conditions in PC4, and FTS reaction conditions in PC5, highlighting the importance of both catalyst formulation and operational parameters (Figure 3). An examination of the experimental data projected onto the first two principal components revealed some key insights (Figure 4). Most data points clustered along a linear plane, reflecting variations from changing operational conditions. However, some distinct outlier clusters were identified, corresponding to unique reaction conditions differing significantly from the rest of the dataset. Visualizing the principal component projections allowed for the identification of patterns, groupings, and anomalies within the data. A scatter plot of PCA1 vs PCA2, color-coded by active metal type, showed distinct clusters for Fe vs. Co catalysts, indicating that Fe has a significant influence on the variance captured by PCA1. The projections also revealed unexplored regions in the parameter space, highlighting gaps in the existing literature and need for further research to optimize jet fuel production by investigating these unexplored areas.

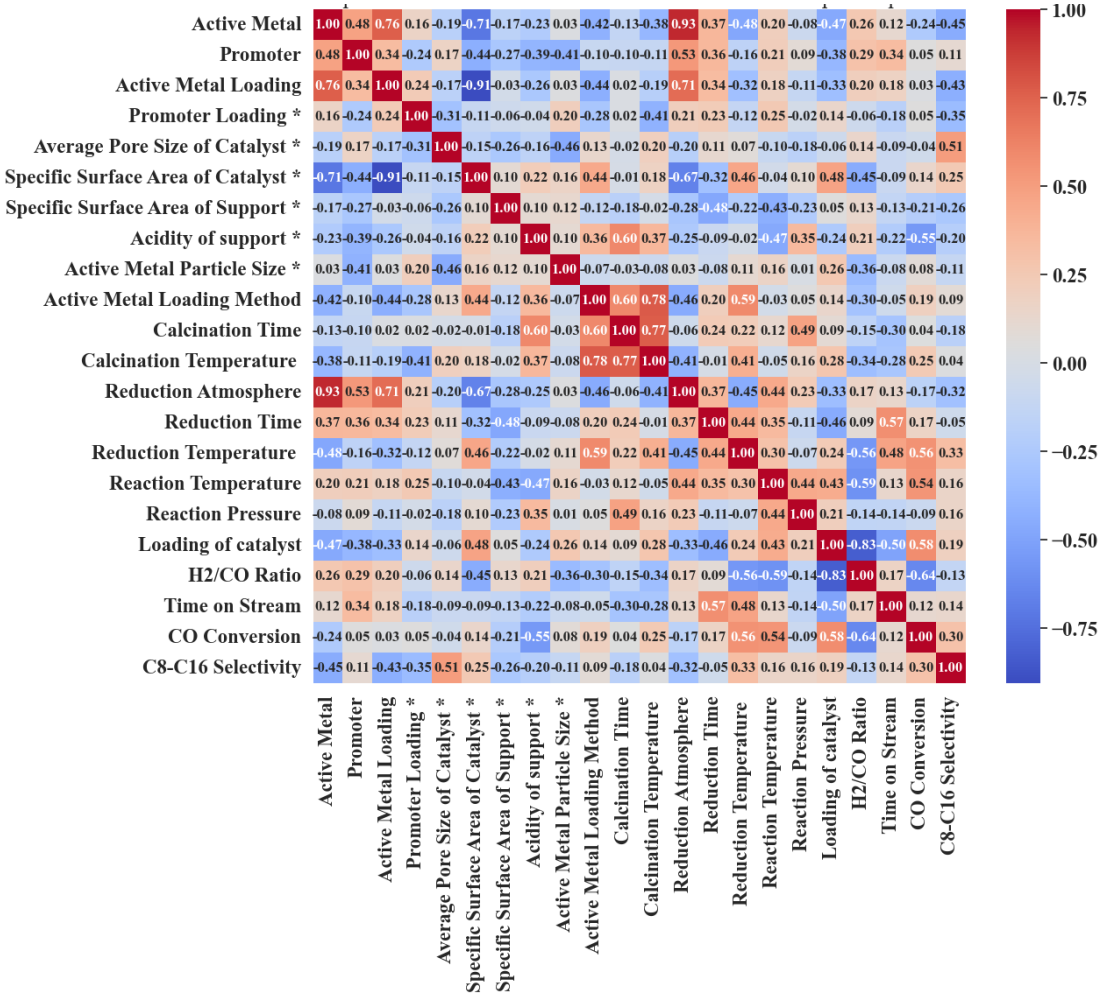

Figure 2: Spearman correlation matrix showing the relationships between input features and output responses

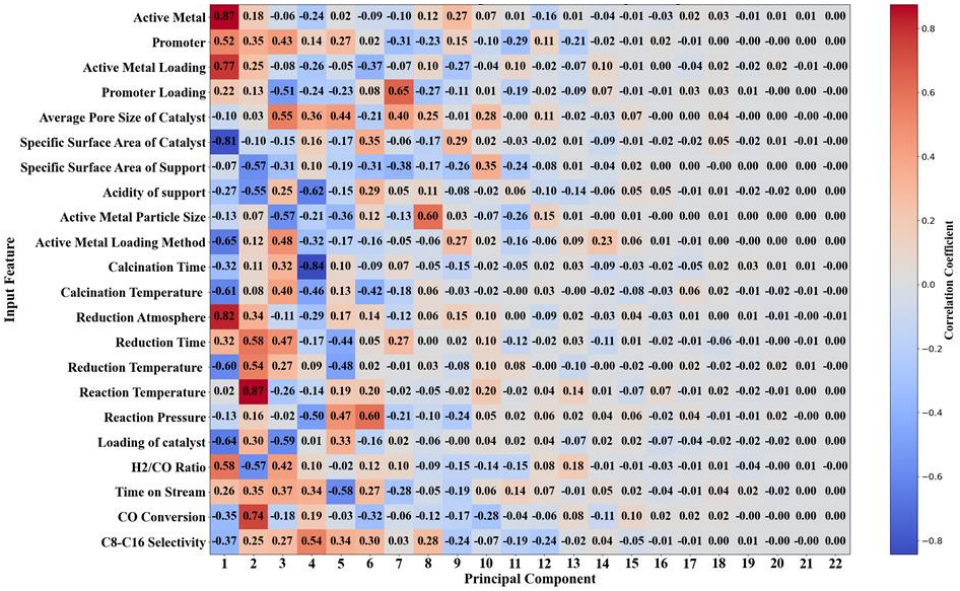

Figure 3: Principal component analysis of the dataset: correlation of the input features and responses with all principal components

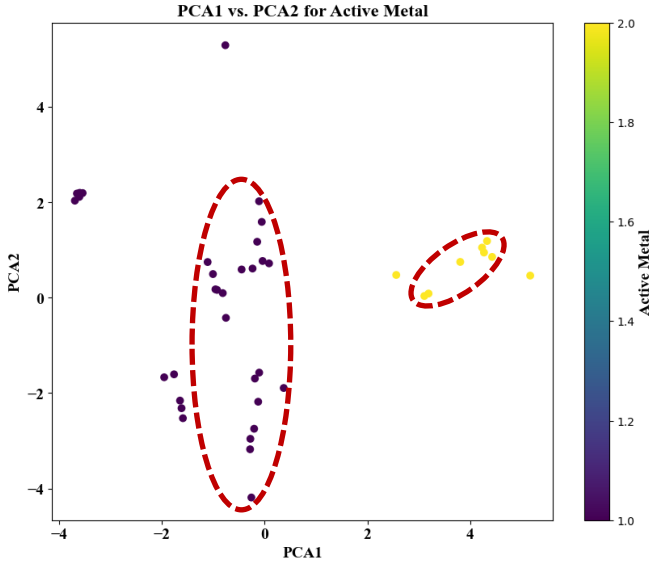

Figure 4: PCs categorized by type of active metal

3.2 Optimization of the machine learning model

In the next step, the performance of a random forest ML model in accurately predicting the CO conversion rate and selectivity for C_8-C_{16} hydrocarbons of a Fischer-Tropsch synthesis (FTS) catalyst under varying formulations and operating parameters (Figure 5) has been evaluated. The random forest model underwent optimization by adjusting the number of decision trees and their maximum depth. The optimal configurations were identified as 10 trees with a maximum depth of 6 for CO conversion, and 6 trees with a maximum depth of 6 for C_8-C_{16} selectivity. The refined model displayed remarkable prediction accuracy, yielding average R^2 values of 0.92 (training) and 0.72 (testing) for CO conversion, and 0.90 (training) and 0.70 (testing) for C_8-C_{16} selectivity. Despite a slight decline in prediction accuracy on the test dataset compared to the training data, the overall performance was deemed sufficient for effectively extrapolating the FTS activity and selectivity across different catalysts and operating conditions. Furthermore, the study evaluated the model's performance with and without utilizing PCA for dimensionality reduction. The findings indicate that the random forest model without PCA may perform better in directly capturing the FTS performance.

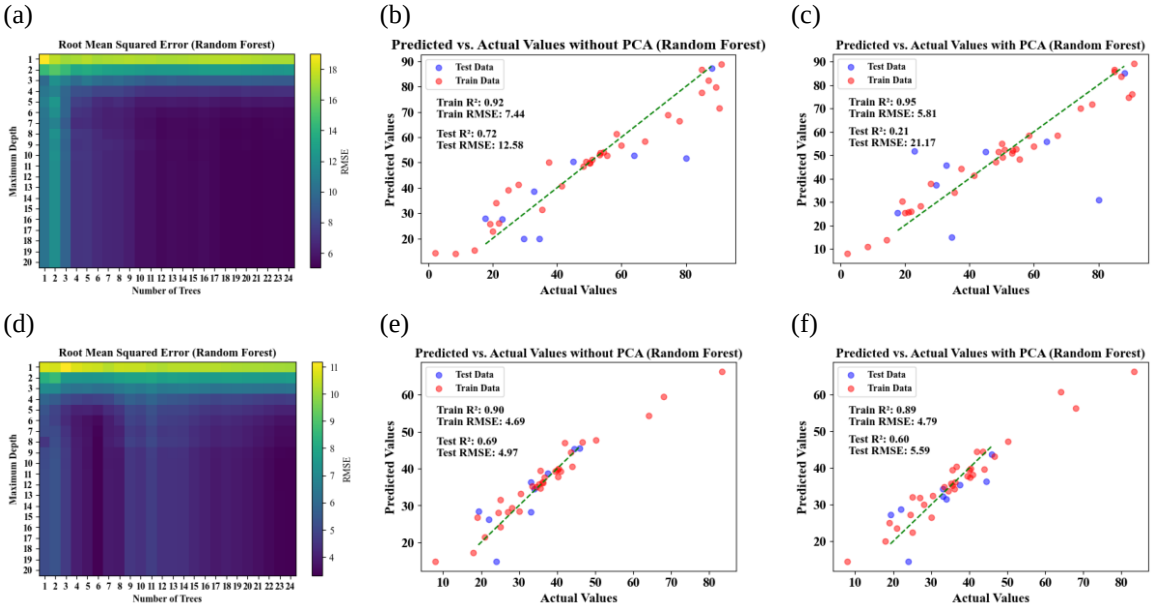

Figure 5: (a) RF hyperparameter tuning for CO conversion, (b) and (c) comparison of predicted CO conversion with original experimental data, (d) RF hyperparameter tuning for C_8-C_{16} selectivity, and (e) and (f) comparison of predicted C_8-C_{16} selectivity with original experimental data

3.3 Impacts of the input features on catalyst activity and selectivity

The pie chart illustrated in Figure 6 reveals that catalyst preparation conditions (type of active metal, promoter, promoter loading) were identified as the least significant features influencing the catalytic activity compared to catalyst characterization and FTS operational conditions.

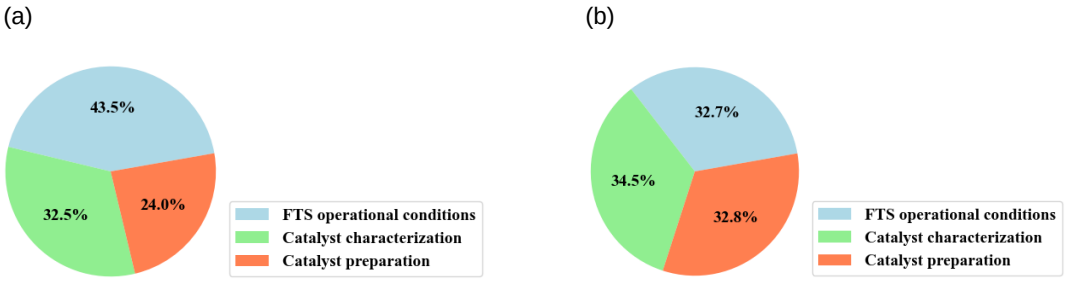

Figure 6: Effects of input features on (a) CO conversion, (b) C8-C16 selectivity

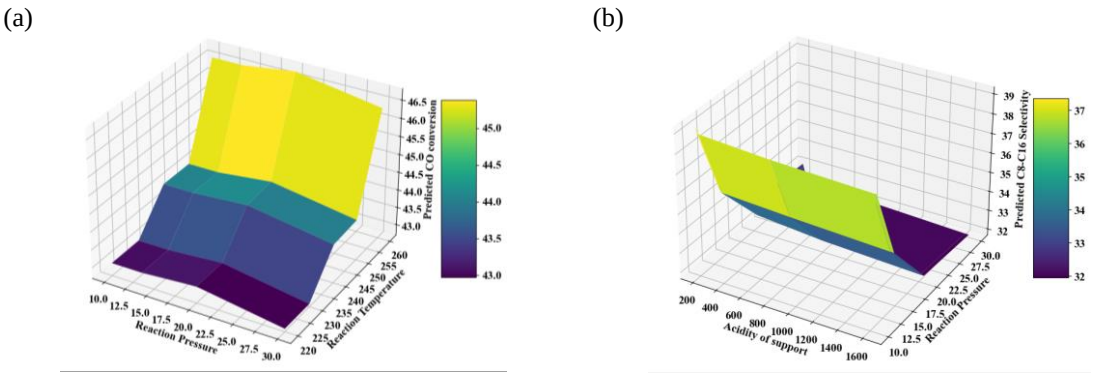

Figure 7: Two-way PDPs of (a) CO conversion and (b) C_8-C_{16} selectivity

While FTS operational conditions predominantly affect the CO conversion, catalyst characterization plays a more significant role in determining the C_8-C_{16} selectivity of the catalysts. The two-way partial dependence plots (PDPs) illustrated in Figure 7 further validate the high interactions and non-linear relationship between the temperature and pressure of the reaction, and catalyst activity. Notably, within the pressure range of 12.5-20 bar, higher CO conversion was observed at temperatures that exceed 250 °C, as depicted in Figure 2. These plots collectively underscore the dominance of catalyst characterization parameters; particularly support acidity and specific surface area, in determining the selectivity towards C_8-C_{16} hydrocarbons. These findings highlight the necessity of tailoring catalyst properties to achieve the desired product distribution in the FTS process. Specifically, the utilization of zeolite supports with engineered textural properties and surface chemistry is crucial for designing catalysts with moderate acidity and optimum surface area. By carefully controlling these support characteristics, it becomes feasible to finely adjust the catalyst's selectivity towards the desired C_8-C_{16} hydrocarbon range.

4. Conclusions

This study developed a machine learning (ML) framework utilizing a random forest (RF) model to predict the performance of Fe/Co catalysts in producing jet fuels (C_8-C_{16}) through the Fischer-Tropsch synthesis (FTS) process. The RF model demonstrated high prediction accuracy for CO conversion (R^2 of 0.92 train, 0.72 test) and C_8-C_{16} selectivity (R^2 of 0.90 train, 0.70 test). Partial dependence plots revealed coupling effects between catalyst textural properties, synthesis conditions, and FTS reaction conditions on the efficiency of the process. This framework offers insights for designing improved FTS catalysts for selective jet fuel production and holds potential for expansion by incorporating additional catalyst descriptors. Moving forward, the dataset will be refined to include further data points, and to incorporate additional descriptors such as specific metal-support interactions or surface chemistry characteristics, to improve the model predictive capabilities. Additionally, exploring advanced ML techniques, such as deep learning algorithms may offer opportunities to capture more complex relationships within the data and to improve the accuracy of predictions. Furthermore, integrating kinetic modeling approaches with ML models could provide a deeper understanding of the reaction mechanisms and support the production of more robust results. Lastly, conducting experimental studies to validate the predictions of the ML framework under real-world FTS conditions is crucial for its practical application in catalyst design and process optimization.

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