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Process conditions effects on Fischer–Tropsch product selectivity: Modeling and optimization through a time and cost-efficient scenario using a limited data size

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A B S T R A C T

Analysis of renewable fuels production systems via modeling enable industries to economically investigate the effective factors besides working in optimized conditions. Accordingly, an innovative strategy using hybrid Artificial Neural Network/Response Surface Methodology (RSM) is proposed for data simulation in order to comprehensively analyze the effect of operating conditions including temperature, pressure, H_2/CO ratio, space velocity, and time on stream on Fischer–Tropsch product distribution. The quadratic response surface models were then validated using the correlation of determination (R^2_{cod}) proving the certainty of the proposed strategy with near-to-one values of R^2_{cod} which is capable of successfully implementing in industrial applications to explore every complex process. Ultimately, single and multi-objective functions were optimized showing that maximum amount of C_2 and minimum amount of other products can be achieved under the following conditions: $T = 500.13$ K, $P = 1.5$ MPa, space velocity = 1 NL/g_{cat}/h, and H_2/CO Ratio = 1.93.

1. Introduction

Energy has been always an everlasting demand for human. As the amount of energy resources such as gas and crude oil reservoirs deteriorates, inexhaustible attempts have been conducted to substitute viable energy supply. Among conventional approaches for energy production, Fischer–Tropsch Synthesis (FTS) with almost a century background, has appealed numerous researchers to delve into different aspects of this process as a promising approach for environmentally friendly fuel production [1–8] . FTS includes a collection of catalytic chemical reactions that converts CO and H_2 mixture (syngas) into a wide range of hydrocarbons. Variety of metallic catalysts can be used for FTS; however, of the different catalysts known for FT reaction, only iron- and cobalt-based have found industrial applications. The synthetic fuel produces almost no pollutants such as sulfur and aromatics resulting in dramatic decline in air pollution. The syngas, which is of great importance in FTS, is usually produced via gasification or steam reforming of carbonaceous material such as coal, CH_4, or biomass. Besides providing syngas via conventional routes, H_2 production is recently established through bioethanol steam reforming [9–11].

The process conditions such as temperature, pressure, gas space velocity, H_2/CO ratio, time on stream as well as catalyst parameters like density and surface area, promoters, and supports can be manipulated to affect the product distribution. Several studies have been done to investigate the effects of such parameters on product distribution of FT process, based on experimental efforts or mathematical modeling. Therefore, the literature review on FT product distribution is divided into two sub-sections: experimental and mathematical.

1.1. Experimental studies on product selectivity

There are numerous reports published regarding the effects of catalyst parameters on product distribution of FTS including supports [12–15] and promoters [16–22] as well as catalyst particle size [23–25]. Additionally, the impacts of process conditions have been considered in several studies. The effects of temperature, pressure, H_2/CO ratio, and space velocity on the performance of a Fe–Mn catalyst were reported by Liu et al. [26]. Similarly, de la Osa et al. [22] studied the effects of reaction conditions on the FT activity and product distribution of an alkali earth metal promoted cobalt based catalyst. Moreover, Todic et al. [27] investigated the effects of temperature, pressure, H_2/CO ratio, space velocity, and time on stream on FTS product distribution of a *Re*-promoted cobalt-alumina catalyst.

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1.2. Mathematical modeling of product selectivity

Modeling of chemical processes provides beneficial outcomes to mimic the effects of operative parameters. Numerous efforts have been conducted to model the product distribution of FT process, so far. Anderson–Schulz–Flory (ASF) [28–34], as one of the mathematical routes to model the product selectivity of FTS has been considered by various studies. Due to significant deviations from the ASF distribution reported in the literature [35], the researchers have to develop modified distributions to decrease the errors . In a most pioneering work Donnelly et al. [28] developed a method by using modified Schulz-Flory to characterize the carbon number distribution of products of the FTS. Nakhaei Pour et al. utilized the concept of two superimposed ASF distributions to investigate effects of reaction conditions and nano-particles size of Fe/Cu/La catalysts [29] as well as the influence of reaction conditions and zeolite presence [32] on product distribution in FTS. Laan and Beenackers [33] proposed a new product distribution model for linear hydrocarbons based on ASF distribution. Furthermore, Schulz and Claeys [36] simulated typical deviations from ideal distributions and validated their model by using experimentally observed data with cobalt and iron catalysts. Recently, Förtsch et al. [34] presented an analytical extension of the classical ideal Anderson–Schulz–Flory (ASF) distribution for the products of Fischer–Tropsch reactions. Their model is capable of describing real distributions with the known deviations from the ideal ASF distribution for C_1 and C_2 components and, by including the known double-ASF concept, the deviations of longer-chain hydrocarbons (C_{10+}).

However, discrepancies of ASF distribution make scientists develop other methods of investigating FTS product distribution [37]. Laan [38], presented a multicomponent mathematical model for a large-scale slurry bubble column reactor operating in the heterogeneous or churn-turbulent flow regime. Their model predicts the composition of the gaseous and liquid streams as a function of the operating parameters. Similarly, Fernandes [39] developed a mathematical model of a slurry reactor used for syngas polymerization to verify the effects of temperature, operating pressure, catalyst holdup, and syngas composition on syngas conversion and carbon product distribution.

1.3. Motivation of this study

Statistical approach offers a constructive scenario to analyze the effective parameters of a multi-dimensional system and also helps the control purposes. Response Surface Methodology (RSM) has widely been employed as a successful statistical modeling tool by researchers who deal with experiments in practical applications, with lots of successful reports. Nowadays, many studies [40–45] apply RSM for the process yield improvement, time development reduction, and overall costs regulation. It, however, may not be applicable for noisy processes, as will be discussed later. Moreover, Artificial Neural Networks (ANNs) play an important role in pattern recognition of nonlinear systems.

The scientific motivation driven the work in the present study primarily arisen from precisely implementing RSM in FTS using ANN based on a few experimental data and followed with the objective of presenting response surface models to comprehensively investigate the effects of process conditions on products selectivity of FTS. Although several experimental and mathematical contributions have been devoted to FT product distribution in the literature, developing response surface models for FT product selectivity has so far been poorly addressed [46–48]. The response surface models provide a fascinating route for modeling FT product distribution and for investigating various effective parameters, in comparison with ASF or other mathematical modeling. The obtained polynomial selectivity models for FT product selectivity enable the researchers to probe the effect of each parameter, their interactions, significance of each parameter, and also optimization. Therefore, considering lack of studies on developing statistical models via RSM, proposing such models would be of importance for industrial applications.

Neural network has been widely utilized in FTS as a kinetic parameters estimator [49], as a modeling and control tool [50], as a water-gas-shift investigator [51], and as an optimization tool [4,52]. Moreover, application of ANN–RSM has been largely addressed in numerous scientific field [53–56], unlike a few studies in FTS [57,58]. Most previous studies of ANN–RSM compare ANN and RSM results, however the main distinctive feature of the present study is that ANN and RSM are coupled to reduce the data required for response surface modeling as well as eliminating the probable adverse effects because of switching between operating conditions. Actually after utilizing performance evaluation methods for training and validating ANN, it is used for data simulation and then construction of response surface models to scrutinize the effects of five adjustable process parameters on the FT product selectivity. The introduced method for developing response surface models for FT product distribution remarkably reduces time and cost for FT data gathering and offers the correct implication of RSM in FTS.

2. Experimental

The experimental data was borrowed from Todic et al. [27] experiments which were conducted over 0.48%*Re*–25%Co/Al_2O_3 catalyst. The catalyst was prepared based on the method elaborated in reference [59]. Experiments were carried out in a 1-L stirred tank slurry reactor with almost 14 g catalyst. To minimize heat and mass gradients, they used catalyst pellets in the sieving range of 44–90 µm. The product distribution data was collected from three temperatures (478, 493, 503 K), two pressures (1.5 and 2.5 MPa), two H_2/CO feed ratios (1.4 and 2.1), a range of time of stream (148–367 h) and different space velocities (1–22.5 NL/g_{cat} /h).

3. Problem statement and methodology

The present study aims to develop a robust and novel technique for reliable investigation of process conditions effects on FTS product selectivity via statistical modeling based on limited data size. Followed by training some NNs based on a few experimental data, RSM was used to design an experimental plan. Afterwards, the designed experiments were performed using the trained NNs. Actually, the trained NNs were employed as a virtual operating unit to experiment the designed plan. Finally, analysis of variance was used to evaluate the significance of the RSM models.

Response Surface Methodology (RSM) is a collection of statistical and mathematical techniques which are useful for developing, improving, and optimizing processes. Moreover, it has important applications in the design, development, and formulation of new products, as well as the improvement of existing product designs. The most extensive applications of RSM are in the industrial world, particularly in situations where several input variables potentially influence some performance measure or quality characteristic of the product or process which is called response [60].

Despite beneficial features of RSM, a designed plan by using RSM may not be executable in a FT experimental set up. Actually the working Fischer–Tropsch synthesis is a complex dynamic system and any sudden change may disturb the steady state. The concept of RSM include variation of the input variables and monitor the output responses in order to indicate the effects of those input variables. While this strategy may be an effective route to investigate the impact of some parameters on some responses in static