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# Selectivity model of Fischer-Tropsch synthesis on the Cobalt-Based Catalyst

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## ABSTRACT
CO conversion model and two models of selectivity of $CH_4$ and $C_5^+$ productswere obtained for Fischer-Tropsch synthesis. The models were fitted to experimental data obtained by $Co/SiO_2$catalystin isothermal packed-bed reactor. The operating conditions are as follows: temperature 473K, pressure 0.5 MPa, space velocity 0.08-0.5 $cm^3$/ (g-s) and water partial pressure 0.01-0.26 MPa.The influence of operating parameters, interaction of them and the effect of water on the CO conversion and the selectivity of products were investigated. Results show that both of the two operating parameters, i.e., water partial pressureand space velocity are influenced.Also it has been shown that as water partial pressure increases, the $C_5^+$ selectivity and the CO conversion are increased while the $CH_4$ selectivity is decreased.

**Keywords:** CO conversion, Fischer-Tropsch synthesis, selectivity model, water effect

## 1. INTRODUCTION
The Fischer-Tropsch synthesis (FTS) is a promising approach to efficientconversion of coal, natural gas and biomass into a mixture of alkenes, alkanes, alcohols and other oxygenates with a range of Carbon number [1-4].This process is very effective for the conversion of expressedcompounds intoenvironmental less damaging fuels and useful chemical with lower emissions of pollutants[5-8].Numerous studies investigated this process in different aspects [9-16].In thisprocess, VIIIgroup metals are used as catalyst [17].Among the metals of this group, Iron and Cobalt catalysts are the two mainlyused one. [18, 19].Of these two, the cobalt-based catalysts (Co/SiO2, Co/activated Carbon, etc) aremore interesting for production of hydrocarbons from synthesis gasbeacause of the cobalt's intrinsic ability in hydrogenating dissociated carbon species, prompting chain growth and resistance to deactivation by carbonaceous deposits [17, 20]. Besides, other reasons of their widely usage in industrial process are: high selectivity and activity, low activity of water-gas shift reactionandtheir relatively low price. Also ithas been shown that cobalt-based catalysts are often suitablefor low temperatureFTS [21, 22].During the FT reaction,water isproduced in varyingquantities [23]. Whetherthis water isindigenous or added,it causesactivity increase [24, 25]and decrease on $Co/SiO_2$ catalysts[26].The deactivation process of this catalyst is increased in the high conversions and high partial pressure of water. In the low conversion when water is not added, the deactivation of catalyst is done much more slowly[25, 27].Decreasingthe deactivation of catalyst is irreversible in high waterpartial pressure [27].

Investigation of products selectivity in FT reaction is an important matter.To study the FTS products distribution, different wayshave been suggested in the literature which can be classified into two main categories: detailed Langmuir-Hinshelwood-Hougen-Watson (LHHW) kinetic models and hydrocarbon selectivity models [28].In achieving the optimum operating conditions for production, economical and temporal factors are of major significance.Hence, selectivitymodels can play an effective role in the designing ofFT process.In this study,appropriate modelswere obtainedfor products selectivity and CO conversion.Also the effects of operating parameters and their corresponding interactions on products selectivitywere studied by Analysis Of Variance (ANOVA) method.

## 2. EXPERIMENTAL
Experimentaldata were obtained fromKrishnamoorthy et al. (2002) studythatinvestigatedthe effect of water on the rate and selectivity of Fischer-Tropsch on cobalt-based catalyst. This study is carried outinthe followingoperating conditions:different ranges of water partial pressure andspace velocity, temperature473 K,pressure 0.5 MPa and $H_2/CO=2$ (Table 1).

$Co/SiO_2$ (12.7 wt%, 1.75 g 100- to-180 µm particles) catalyst used, here, first, was diluted by the $SiO_2$ (2.8 g, 100-to 180-µm particles), then it was reduced by $H_2$ ($12\times10^3$ $Cm^3$/h. g-Cat) for 1 hour within reactor while being heated to 598 K at 0.17 K/S.The FTSrates and selectivity were measured by an isothermal reactor with plug-flow hydrodynamics. FT reactions were carried out at two different pressures (0.5 and 2.0 MPa) and temperature473 K by syngas (synthesis gas, 62% $H_2$, 31% CO, 7% $N_2$ internal standard). All the concentrations of products(such as CO, $CO_2$and hydrocarbons)were measured by online gas chromatography. Water was added to the synthesis gas reactant stream, also, all flow lines were kept to prevent condensation at a temperature of 410 K or higher[24].

# 3. RESULTS AND DISCUSSION

In this work, the effects of water partial pressure ($P_{H2O}$) and space velocity(SV) on products selectivity ($CH_4$ and $C_5^+$) and CO conversion in FTS was investigatedthrough a statistical model.In order to obtain these models,there are two input operating parameters, i.e.,$P_{H2O}$and SV and three responses, i.e., selectivity of $CH_4$, selectivity of $C_5^+$and conversion of CO (table 1).

For each of these three responses, a model was fitted (Table 2). Then, the influence of two input parameters and their interactionswere studied by ANOVA and insignificant ones, considering p-values, were removed from the models. Since the accuracy of the model regression can be evaluated using $R^2$ and adjusted $R^2$ coefficients (checking the goodness of fit), they are calculated for all three models (table 3). As known for an accuracy model, the $R^2$ value must be near 1 and adjusted $R^2$ value must always be less than or equal to $R^2$. The final statistical models for $CH_4$ and $C_5^+$ selectivity and CO conversion are shown in table 2. SV is space velocity in ($cm^3$/(g.s)) and $P_{H2O}$ is water partial pressure in (MPa). Furthermore, P-value is used to verify the significance of the regression coefficients of parameters and the interaction between them. P-value of 95 % confidence (P-value less than 0.05) means that the calculated value of the parameter is accurate with 95 % confidence.

As table 3 shows, both of the two operating parameters, $P_{H2O}$ and SV influenced the selectivity of products and CO conversion. Also, all the interaction parameters influenced the distribution of products and CO conversion. Since the P-value of $P_{H2O}{}^2\times$ SV interaction parameter (0.5351) was inappropriate, thus, it was removed from CO conversion model. Finally, the agreement between predicted values with experimental values is can be seen in table 4.For the purpose of illustration, three-dimensional graphs are used for the selectivity variations trend of products. In fig. 1-2, variations of products selectivity as a function of the water partial pressures and space velocities have been depicted. Also,in fig. 3, the trend of CO conversion changesis shown.

As illustrated in the figures(Figs. 1, 2and 3), it can be seen thatas thewater partial pressure increases, the $C_5^+$ selectivity and the CO conversionare increased while the $CH_4$ selectivity is decreased.

According to Fig.1, initially,the CO conversion is increased with a simultaneous decrease and increase, respectively,in space velocity andwater partial pressure, thenafter reachinga maximum value, it decreases.Following thereactions occurring in the process might explain this reduction.

The Main reactions duringFT synthesis are as follows[29]:

Alkanes: $nCO + (2n + 1)\, H_2 \rightarrow C_nH_{2n+2} + nH_2O$ (1)

Alkenes: $nCO + 2nH_2 \rightarrow C_nH_{2n} + n\, H_2O$(2)

Water-gas shift: $CO + H_2O \Leftrightarrow CO_2 + H_2$(3)

On the other hand, the CO conversion degree can be expressed as follows:

CO conversion = ($CO_{input} - CO_{output}$) / $CO_{input}$(4)

Because reactants amount reduces during the reaction, the rate of first reaction (number one) decreaseswhile the rate of water-gas shift reaction increases(reaction number 2),hencethe CO is produced by increasing the residence time. Therefore,the CO conversion is reduced according to (4).

Variation of selectivity of $C_5^+$ with water partial pressure and space velocity are depicted in Fig. 2. As it is shown,the $C_5^+$ selectivityincreases with an increase inboth residence time and water partial pressure, the same result as in CO diagram (fig. 1).The observed increase in $C_5^+$selectivity is resulted from secondary reactions of primary olefin products in higher residence times.On the other hand, because water inhibits the secondary hydrogenation of primary olefins, therefore, more olefins are available to join for growing chain.Due toits dipole property, watercausescatalytic property in the catalytic system,thus, the selectivity of $C_5^+$ rises.However,asthe water concentrationincreases gradually, its rate decreases. Likewise,at low water concentrations, water molecules can acceleratethe reaction rate. While at high water concentrations (and higher residence time), beingabsorbed to the catalyst active sites, they are combined with the silicate and causes reduction rate,as a result of which, the selectivity of $C_5^+$isreduced.

Increasing selectivity of $C_5^+$ is coupled with decreasing $CH_4$ selectivity. In fact, with increasing the residence time, the smaller hydrocarbons have more opportunityto form the higher hydrocarbons.Hence,the $CH_4$ selectivity is decreased with increasing residence time at low water partial pressure (Fig. 3).

# 4. CONCLUSION

In this study two FT product selectivity models and CO conversion model were achievedand the effects of operating parameters and the water partial pressure on selectivity of products were investigated. As the resultshave shown, both of the two operating parameters $P_{H2O}$and SVand the interaction parameters,$P_{H2O}\times P_{H2O}$, SV × SV, $P_{H2O}\times SV^2$ and $P_{H2O}\times$ SV have influence on selectivity of products and CO conversion.In the catalytic system it has been observed thatincreasing the water concentration causesan increase inthe $C_5^+$ selectivity and CO conversion and a decreasein the $CH_4$ selectivity.

Due to decreasing of the water catalytic effect and raising deactivation of the catalyst, in high water concentrations, the CO conversion and the $C_5^+$selectivity are reduced gradually. Finally, comparison of results showed that the three models predict experimental data well with small errors in agreementwith the fitting.

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**Table 1. Effect of reaction condition on selectivity of hydrocarbons (12.7 wt% $Co/SiO_2$ at 473 K, 0.5 MPa, and $H_2/CO$=2)**

| Spacevelocity ($cm^3$/(g-s)) | Average $H_2O$ Pressure (MPa) | CO conversion (%) | CO conversion rate ($h^{-1}$) | $CH_4$ selectivity % | $C_5^+$ selectivity % |
|---|---|---|---|---|---|
| 0.08 | 0.07 | 56.8 | 16.9 | 6.4 | 88.2 |
| 0.17 | 0.02 | 26.7 | 16.8 | 7.1 | 86.4 |
| 0.25 | 0.02 | 18.2 | 16.6 | 7.4 | 85.9 |
| 0.35 | 0.01 | 13.1 | 16.8 | 7.3 | 86.3 |
| 0.50 | 0.01 | 9.8 | 17.8 | 7.4 | 84.9 |
| 0.50 | 0.06 | 10.2 | 18.5 | 5.9 | 87.2 |
| 0.50 | 0.11 | 11.4 | 20.8 | 5.6 | 87.6 |
| 0.50 | 0.16 | 13.5 | 24.6 | 5.2 | 89.2 |
| 0.50 | 0.21 | 14.1 | 25.7 | 3.4 | 92.3 |
| 0.50 | 0.26 | 13.9 | 25.3 | 3.2 | 92.9 |

A is space velocity in ($cm^3$/ (g.s)) and B is water partial pressure in (MPa).

**Table 2. Model of CO conversion and selectivity models of products**

| |
|---|
| CO conversion model = $39.639 - 95.181*A + 724.928*B - 5150.07*A*B + 68.131*A^2 - 219.869*B^2 + 7520.49*A^2*B$ |
| $CH_4$ selectivity model= $6.427 + 2.031*A - 32.066*B + 417.43*A*B - 0.233*A^2 + 15.393*B^2 - 776.51*A^2*B - 23.643*A*B^2$ |
| $C_5^+$ selectivity model= $84.387 + 23.515*A + 193.434*B - 1778.915*A*B - 45.352*A^2 - 648.051*B^2 + 2896.408*A^2*B + 1328.959*A*B^2$ |

**Table 3 . Analysis of variance**

| Terms | CO conversion model | $CH_4$ selectivity model | $C_5^+$ selectivity model |
|---|---|---|---|
| | P-value | P-value | P-value |
| $P_{H2O}$ | <0.0001 | <0.0001 | <0.0001 |
| SV | 0.0002 | <0.0001 | 0.022 |
| $P_{H2O}\times$ SV | 0.0088 | 0.0115 | <0.0001 |
| $P_{H2O}\times P_{H2O}$ | 0.0002 | <0.0001 | <0.0001 |
| SV × SV | 0.0004 | <0.0001 | <0.0001 |
| $P_{H2O}^2\times$ SV | 0.5351 | <0.0001 | 0.0002 |
| $P_{H2O}\times SV^2$ | 0.0054 | <0.0001 | <0.0001 |
| | R-sq = 99%<br>R-sq (adj) = 99% | R-sq = 96%<br>R-sq (adj) = 85% | R-sq = 97%<br>R-sq (adj) = 87% |

 

**Table 4. Compare calculated data from of each of obtained model with the experimental data**

| water partial pressure (MPa) | Space velocity ($cm^3/(g.s)$) | CO conversion value | | $CH_4$ selectivity value | | $C_5^+$selectivity value | |
|---|---|---|---|---|---|---|---|
| | | Exp. value | Calc. value | Exp value | Exp. value | Exp value | Calc value |
| 0.07 | 0.08 | 56.8 | 56.8 | 6.4 | 6.4 | 88.2 | 88.2 |
| 0.02 | 0.17 | 26.7 | 26.7 | 7.1 | 7.1 | 86.4 | 86.4 |
| 0.02 | 0.25 | 18.2 | 18.2 | 7.4 | 7.4 | 85.9 | 85.9 |
| 0.01 | 0.35 | 13.1 | 13.1 | 7.3 | 7.3 | 86.3 | 86.3 |
| 0.01 | 0.5 | 9.8 | 9.4 | 7.4 | 7.2 | 84.9 | 85.1 |
| 0.06 | 0.5 | 10.2 | 10.7 | 5.9 | 6.35 | 87.2 | 86.5 |
| 0.11 | 0.5 | 11.4 | 11.9 | 5.6 | 5.5 | 87.6 | 88.1 |
| 0.16 | 0.5 | 13.5 | 12.9 | 5.2 | 4.7 | 89.2 | 89.7 |
| 0.21 | 0.5 | 14.1 | 13.7 | 3.4 | 3.8 | 92.3 | 91.4 |
| 0.26 | 0.5 | 13.9 | 14.3 | 3.2 | 3.1 | 92.9 | 93.2 |

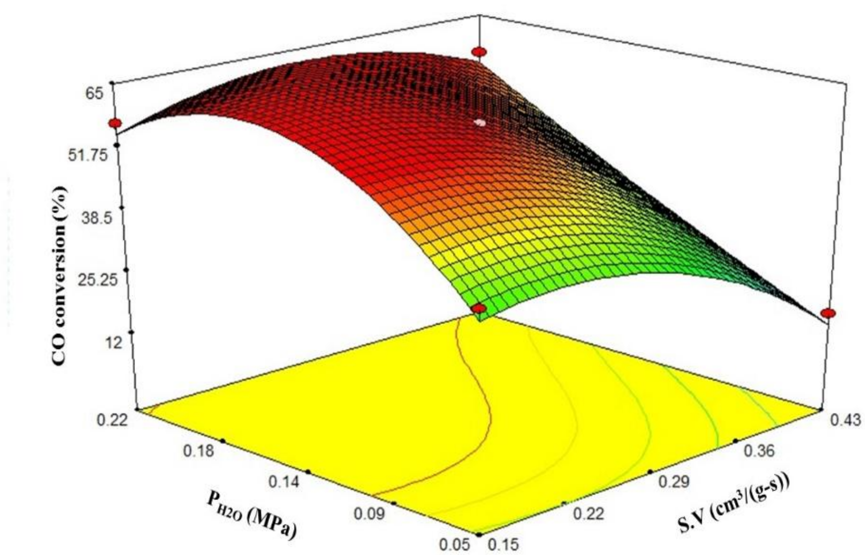


**FIG.1.** Variation of calculated conversion of CO as a function of water partial pressurevalues and space velocity values.

 

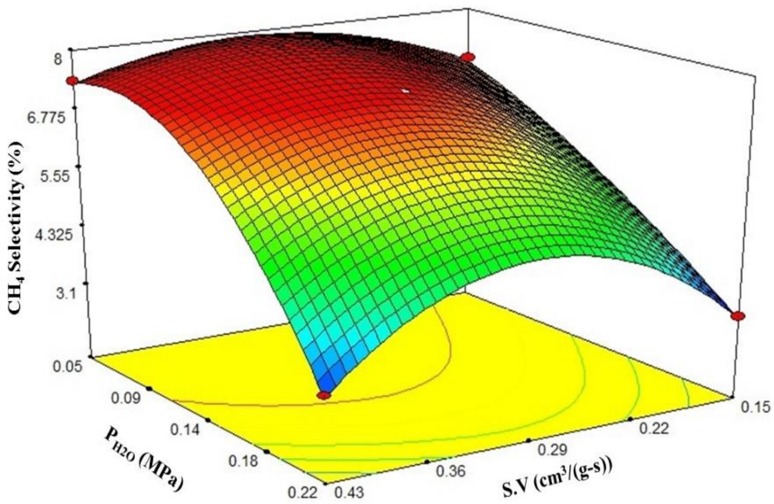


**FIG. 3.** Variation of calculated selectivity of $CH_4$ as a function of water partial pressurevalues and space velocity values.

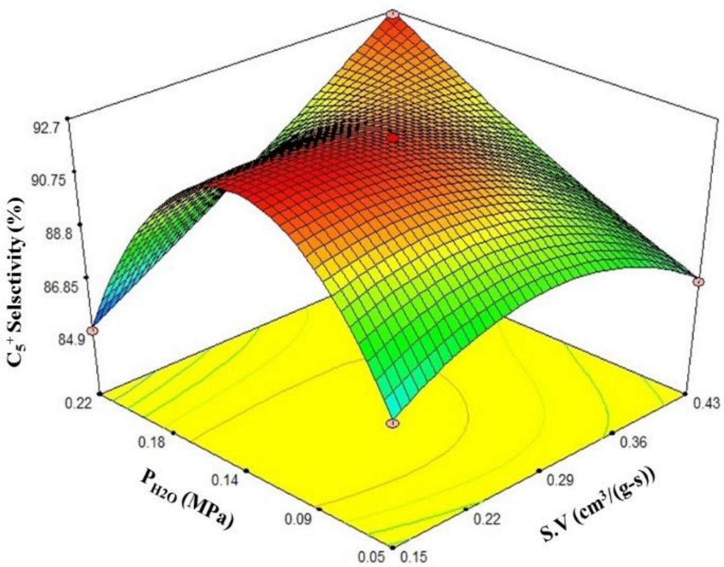


**FIG.2.** Variation of calculated selectivity of $C_5^+$ as a function of water partial pressurevalues and space velocity values.

 