

SENSITIVITY STUDY OF THE PROPANE DEHYDROGENATION PROCESS IN AN INDUSTRIAL RADIAL MOVING BED REACTOR

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Abstract

Modelling and simulation of the propane dehydrogenation reaction is important for predicting an optimum operating condition to maximise the propylene yield. The present study performed the modelling and simulation study of propane dehydrogenation over a platinum based catalyst in radial moving bed reactor (RMBR). First order power law model was used to express the propane dehydrogenation reaction and side reactions. RMBR was discretized into axial and radial directions and the equations of the discretized bed were solved numerically. The kinetic parameters were optimised by comparing the simulation results with plant data. The predicted propane conversion, reactor outlet temperature and coke content deviated less than 5% from the plant data. The validated model was then used for the sensitivity study to evaluate the influence of different possible disturbances on the process. It was found that the reactor inlet temperature was the most influenced parameter to the reactor performance. The maximum propylene yield 30.34% was produced when the W/F was +10 K, H_2/HC was -0.2 and U_s was +100 kg/hr from the base case

Keywords: Propane dehydrogenation, Radial moving bed reactor, Modelling, Simulation, Sensitivity study.

1. Introduction

The growing consumption of propylene derivatives has profoundly increased the propylene demand in recent years. It has been increasing at an annual average rate of 5.7 percent since 1991 and it is expected to continue growing at an average yearly rate of 3.8 percent from year 2005 until 2015[1-3]. More than 60% of world's propylene production was used to produce polypropylene while the balance was consumed for the derivatives production such as cumene, propylene oxide,

Nomenclatures

a	Catalyst activity
C_m	Total concentration of the active sites, g active site/ g catalyst
F_A	Molar flow rate of component A , kmol/hr
h	height of catalyst bed, m
H_2/HC	Hydrogen to hydrocarbon molar ratio, mol/mol
i	Number of elements in the axial
j	Number of elements in the radial
k_1	Rate constant for forward reaction, kmol/(kg.hr)
k_{-1}	Rate constant for backward reaction, kmol/(kg.hr)
k_2	Rate constant for propane cracking, kmol/(kg.hr)
k_3	Rate constant for ethylene hydrogenolysis, kmol/(kg.hr)
k_c	Coking rate constant
K_{ads}	Adsorption equilibrium constants
K_{eq}	Equilibrium constant for propane dehydrogenation reaction, kPa
P_A	Partial pressure of component A , kPaG
R_1	are the bed inner radius, m
R_2	are the bed outer radius, m
T	Temperature, K
U_s	Catalyst circulation rate, kg/hr
Δr	elemental ring thickness
Δz	elemental height thickness

Abbreviations

FBD	Fluidized Bed Dehydrogenation
PDH	Propane Dehydrogenation Technology
RIT	Reactor Inlet Temperature
RMBR	Radial Moving Bed Reactor
STAR	Steam Activated Reforming
WHSV	Weight Hourly Space Velocity

isopropanol, acrylic acid, acrylonitrile, and other polygas chemical[3, 4].

The disparity of supply and demand for propylene has inspired the development of the on-purpose propylene production technologies such as olefin metathesis and propane dehydrogenation. Currently, the on-purpose production of propylene from propane is more economical than the other methods like naphtha cracking or other refinery processes due to the inexpensive price of propane[5]. Five licensed technologies with different type of catalyst, catalyst regeneration method, reactor design and operating condition are available for propane dehydrogenation. The technologies are *Catofin (Houdry Technology)* commercialized by ABB Lummus, *Oleflex* commercialized by UOP, *STAR (steam activated reforming)* by Krupp Uhde, *PDH (propane dehydrogenation technology)* by Linde-BASF-Statoil and *FBD (fluidized bed dehydrogenation)* by Snamprogetti-Yarsintez[6].

Propane is mainly derived from the non-renewable natural gas and petroleum resources. The continuous consumption of propane is depleting the natural gas and petroleum supplies. The propylene productivity should be maximised to sustain the propane dehydrogenation process. It can be done by increasing the

production yield under the optimum operating conditions such as pressure, temperature and H_2/HC ratio[7]. The propylene production industries who adopt the propane dehydrogenation process require an accurate reactor model before the process is optimised to increase the productivity.

Sahebdelfar and Bijani[8] have developed a simple model to predict the performance of a moving bed reactor for isobutane dehydrogenation. The reactor was assumed as a simple packed bed reactor. The predicted conversion of the second reactor well matched with the plant data. However, the conversion of first reactor was underestimated while the conversion of third reactor was overrated. Sahebdelfar et al.[9] used discretization method to model the radial moving bed reactors. The conversion, catalyst activity, and temperature profile were generated for the axial and radial directions of the reactors. It was found that the error between the simulated and actual reactor outlet temperature was approximately 30%. Besides, the deviation of the simulated total conversion from the plant data was approximately 25%. It was claimed that the error could be reduced by increasing the calculation step number.

Numerous sensitivity studies were carried out for propane dehydrogenation process but most of it was experimentally based. Sahebdelfar and Zangeneh[10] studied the influence of reactor temperature, H_2/HC molar ratio and WHSV (Weight Hourly Space Velocity) on the product selectivity in propane dehydrogenation process. It was found that lower reaction temperatures and higher hydrogen to hydrocarbon ratios resulted in higher propylene selectivity at the expense of lower propane conversion. Farjoo et al.[11] investigated the effect of temperature and residence time on the propane conversion and propylene selectivity. Reactor temperature was found to be the most significant parameter to the propane conversion. With the increment of 40 K in reactor temperature, increase propane conversion for about 10%. Zangeneh et al.[12] found the optimum reaction condition for propane dehydrogenation from the sensitivity study of reaction temperature, H_2/HC molar ratio and space-velocity to the propane conversion, propylene selectivity and propylene yield. It was found the optimum conditions for propane dehydrogenation to be $T = 893K$, $H_2/HC = 0.6$ and $WHSV = 2.2 h^{-1}$.

To date, the sensitivity study through simulation is limited. The radial moving bed reactor modelling and simulation considering both the radial and axial variations for propane dehydrogenation was not reported in the open literature. Chin et al.[7] modelled the radial moving bed reactor by assuming it was plug flow reactor. The deviations of the predicted composition of H_2 , C_2H_4 and C_2H_6 from the plant data were 21%, 14% and 11%. It was stated that these deviations may be attributed to the omission of the variations of concentration, temperature and reaction rate in the radial direction. The sensitivity study was carried out to examine the effect of reactor inlet temperature and H_2/HC molar ratio on the propane dehydrogenation and it was found that the operating condition to maximise the propylene production is $\Delta RIT_1 = -1$, $\Delta RIT_2 = +1$, $\Delta RIT_3 = +1$, $\Delta RIT_4 = +2$ and $\Delta H_2/HC = -0.02$ from the base case simulation.

In the present work, a two dimensional model for radial moving bed reactor (RMBR) was developed. Industrial plant data was used to validate the model. The model was then used for studying the effect of operating parameter on the propane conversion, propylene selectivity, propylene yield and also coke content.

Operating parameters used for sensitivity in this study were reactor inlet temperature (RIT), H_2/HC molar ratio and catalyst circulation rate (U_s).

2. Model Development for RMBR

2.1. Reaction kinetics

Propane dehydrogenation is known as an endothermic equilibrium limited reaction. The elevated temperatures and low pressures favor the forward reaction and hence increasing the yield of propylene. It is usually carried out at 873-923 K under the pressure of 15 - 250 kPaG in the presence of metal catalyst such as platinum based catalyst. The reaction and reaction rate for propane dehydrogenation are shown in Eqs. (1)-(2).



$$-r_1 = k_1 P_{C_3H_8} - k_{-1} P_{C_3H_6} P_{H_2} = k_1 \left(P_{C_3H_8} - \frac{P_{C_3H_6} P_{H_2}}{K_{eq}} \right) \quad (2)$$

where, k_1 is rate constant for forward reaction, k_{-1} is rate constant for backward reaction, K_{eq} is reaction equilibrium constant for propane dehydrogenation, $P_{C_3H_8}$, $P_{C_3H_6}$ and P_{H_2} are partial pressure for propylene, propane and hydrogen respectively. Equilibrium constant for propane dehydrogenation as a function of temperature (T) is shown in Eq. (3).

$$K_{eq} = 1.47 \times 10^7 \exp(-15403/T) \text{ kPa} \quad (3)$$

The elevated temperature also promotes side reactions such propane cracking and hydrogenolysis. The propane is cracked into methane and ethylene as a result of catalytic cracking on the catalyst surface [7, 11, 13-15]. The reaction and reaction rate for side reactions are shown in Eqs. (4)-(7).



$$-r_2 = k_2 P_{C_3H_8} \quad (5)$$



$$-r_3 = k_3 P_{C_2H_4} P_{H_2} \quad (7)$$

where, k_2 is rate constant for propane cracking, k_3 is rate constant for ethylene hydrogenolysis and $P_{C_2H_4}$ is partial pressure for ethylene.

The reaction and reaction rate for coke formation are expressed in Eqs. (8)-(9) [13,16].



$$\frac{da}{dt} = \frac{k_c P_{C_3H_6}}{C_m (1 + K_{ads} P_{H_2})} a \quad (9)$$

where, C_m is the total concentration of the active sites, k_c is the coking rate constant and K_{ads} is the adsorption equilibrium constants.

2.2. RMBR process description

The commercial radial moving bed reactor for propane dehydrogenation consists of a series of four reactors as shown in Fig. 1. The reactor comprises of two perforated coaxial cylinders to retain the catalysts. The feed gas enters from the bottom of first reactor and crosses radially through the catalyst bed that moves slowly downwards through the reactor by gravitational force. The catalyst from the first reactor is then collected in the catalyst collector and lifted to the second reactor. The outlet product from first reactor is fed to the second reactor. After passing through the four reactors in series, the catalyst at the outlet of last reactor is sent to the continuous catalyst regeneration unit. The catalyst is then regenerated and recycled back to the first reactor for the next reaction cycle. The outlet temperature decreases since the process is highly endothermic. Inter-stage heaters are required to increase the inlet temperature [8-9, 15, 17].

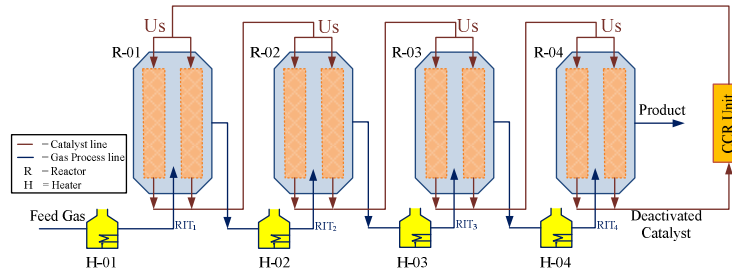


Fig. 1. Industrial RMBR for Propane Dehydrogenation Process.

2.3. Design equation

The simulation of RMBR in this study was performed by discretizing the catalyst bed into a number of ring shape element in axial and radial direction. The equation of the discretized bed is shown in Eq. (10).

$$\Delta W_{i,j} = \frac{W \cdot \Delta z \cdot \Delta r \cdot [2R_1 - (2j - 1) \cdot \Delta r]}{h \cdot (R_2^2 - R_1^2)} \quad (10)$$

where, i and j are the number of elements in the axial and radial directions respectively. R_1 and R_2 are the bed inner and outer radius, h is the height of catalyst bed, Δr and Δz are the elemental ring thickness and the height of ring respectively.

The mass balance and energy balance equations for RMBR are shown in Eqs. (11)-(12).

$$\frac{dF_A}{dW} = r'_{rx,n} \quad (11)$$

$$\frac{dT}{dW} = \frac{(\Delta H_{rx,n,T})(-r'_{rx,n})}{\sum F_A C_{pA}} \quad (12)$$

The rate equation for coke formation in Eq. (9) was then rewritten as a function of catalyst weight. The equation is shown in Eq. (13).

$$\frac{da}{dW} = \frac{k_c P_{C_3H_6}}{C_m(1 + K_{ads}P_{H_2})} \frac{a}{U_s} \quad (13)$$

where, U_s is the catalyst loading.

All the design equations for RMBR can be simplified in terms of radial and axial directions as in Eqs. (14)-(17):

$$W_{i,j} = f(\Delta r, \Delta z, R_1, R_2, L, j) \quad (14)$$

$$F_{A|i,j} = f(W_{i,j}; F_{A|i,j-1}; T_{i,j-1}; a_{i-1,j}) \quad (15)$$

$$T_{i,j} = f(W_{i,j}; F_{A|i,j}; T_{i,j-1}) \quad (16)$$

$$a_{i,j} = f(W_{i,j}; F_{A|i,j}; T_{i,j}) \quad (17)$$

2.4. Numerical solution

A set of differential equations was solved with 4th order Runge-Kutta method in MATLAB. The schematic for the calculation steps involved is shown in Fig. 2. Calculation was started from the inner ring at the 1st row of RMBR with $i = 1$ and $j = 1$. The catalyst weight at this point was first calculated. The outlet component molar flow of the current discretized bed ($W_{i,j}$) were then calculated based on the inlet component molar flow rate ($F_{A|i,j-1}$), inlet temperature ($T_{i,j-1}$) and initial catalyst activity ($a_{i-1,j}$). Subsequently, the outlet temperature and outlet catalyst activity were calculated. The outlet component molar flow and outlet temperature of the 1st ring became the inlet component molar flow and inlet temperature of the 2nd ring with $i = 1$ and $j = 2$. The calculation step for the first row was then repeated until the n^{th} ring of first row with $j = n$.

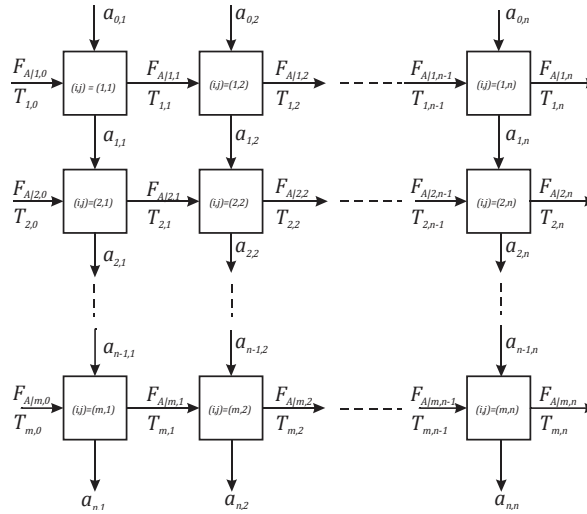


Fig. 2. Calculation Steps for the Numerical Solution of RMBR Model.

The calculations for the 2nd row with $i = 2$ were started once the calculations for the 1st row were completed. Similarly, the calculations were initiated from 1st ring with $j = 1$ until the last ring with $j = n$. Calculations were then repeated

until the bottom of reactor with $i = m$. The reactor outlet composition and temperature were obtained by the mass-average of the values at the outer wall of the bed while the reactor outlet catalyst activity was attained by the mass-average of the values at the lowest rings.

Optimization of the kinetic parameters was performed using *fminsearch* in MATLAB by comparing the simulation results with the plant data. The objective function for optimization is shown in Eq. (18).

$$\min = \sum (F_{A,data} - F_{A,predicted})^2 \quad (18)$$

The optimised kinetic parameters were then used for the sensitivity studies. The operating conditions of the plant that gave highest conversion and yield were taken as the base case of the sensitivity studies. The operating parameters were then varied to investigate its effect to the reactor performance. The range of the variation for each parameter is shown in Table 1.

Table 1. Operating Parameter Variation.

Operating Parameter	*Value	Variation
RIT ₁ ,K	RIT_1	± 10K
RIT ₂ ,K	RIT_2	± 10K
RIT ₃ ,K	RIT_3	± 10K
RIT ₄ ,K	RIT_4	± 10K
Hydrogen to hydrocarbon molar ratio	H ₂ /HC	± 0.2
Catalyst Circulation Rate (U _s), kg/hr	U _s	± 100 kg/hr

*due to its confidentiality, the operating parameter value is given in symbol.

3. Results and Discussion

3.1. Kinetic parameter optimization

Kinetic parameters required for Eqs. (2), (5), (7) and (9) were obtained by comparing the simulated mole fractions of the major components in the reactor outlet with the plant data. Table 2 shows all the values of the reaction kinetic parameters.

Table 2. Kinetic Constants of the Proposed Kinetic Models.

Parameter	Value	Unit
k_1	$k_{01} =$	13.920 kmol/(kg.hr.kPa)
	$E_1 =$	31.978 kJ/mol
k_2	$k_{02} =$	$1.573 \cdot 10^6$ kmol/(kg.hr.kPa)
	$E_2 =$	141.94 kJ/mol
k_3	$k_{03} =$	3.3965 kmol/(kg.hr.kPa ²)
	$E_3 =$	149.41 kJ/mol
k_d	$k_{0d} =$	1.097×10^{-1} (kg.kPa.hr) ⁻¹
	$E_d =$	146.21 kJ/mol
K_{ads}	$K_{0ads} =$	5.553×10^{-2} atm ⁻¹
	$\Delta H_{ads} =$	91.798 kJ/mol
C_m	$=$	0.0546 g active site/ g catalyst

As shown in the subsequent sections.

The predicted activation energies of the propane dehydrogenation and side reactions are comparable with the experimental data reported in the literature. These kinetic data were used for the simulation in the subsequent sections.

The corresponding parity plots for the mole fractions of propane, propylene and hydrogen (components in the main reaction) and the mole fractions of methane, ethane and ethylene (products of the side reactions) are shown in Figs. 3.

Ideally, the data points should lie on the $x = y$ line (shown in solid line). The propane dehydrogenation process in RMBR is well described by the estimated kinetic parameters since the data points of most of the components except C_2H_4 are within the marginal error lines of 20%. The C_2H_4 outlet composition is not accurately predicted due to its very low absolute value as compared to the other components.

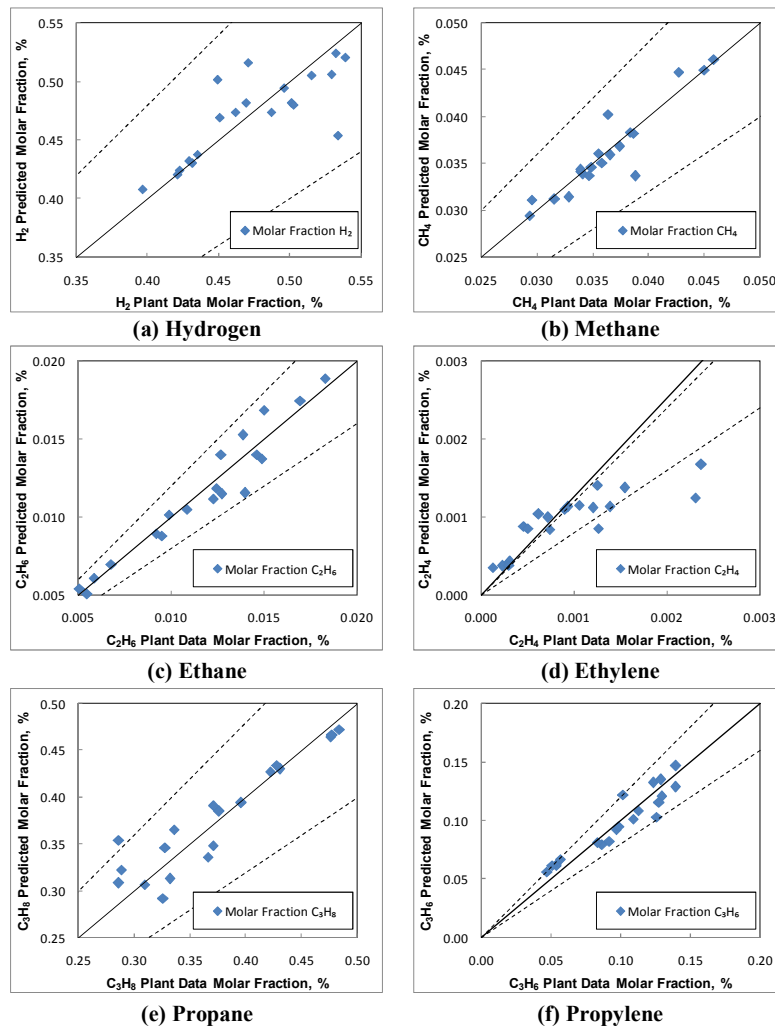


Fig. 3. Parity Plot of the Plant Data and Predicted Molar Fraction.

3.2. Sensitivity study

A base case simulation has been done before performing sensitivity study with parameters shown in Table 1. Operating parameter used for base case simulation was chosen among the parameter that gave highest yield obtained from propane dehydrogenation plant. The simulation result for propane conversion, propylene selectivity, yield and predicted coke formation were 32.54%, 86.35%, 28.09% and 4.01% respectively.

Table 3. Comparison of the Predicted and the Experimental Activation Energies.

Sources	Activation Energies (kJ/mol)		
	Main Rx	1 st Side Rx	2 nd Side Rx
	Eq. (1)	Eq. (4)	Eq. (5)
Larsson et al. [18]	34.8 ± 19.6	-	-
Gascón et al.[19]	35.5 ± 13.8	308 ± 13.8	-
Lobera et al. [20]	34.57 ± 9.13	137.31 ± 37.82	154.54 ± 15.09
Li et al. [13]	44.7 ± 16.9	104.8 ± 9.9	-
Present Study	31.978	141.94	149.41

3.2.1. Reactor inlet temperature (RIT)

The effect of RIT was studied by maintaining the value of H_2/HC molar ratio, feed flow rate, and U_s while varying the RIT by ± 10 K. The RIT of the propane dehydrogenation process is commonly represented by the Weighted Average Inlet Temperature (WAIT). WAIT can be calculated by summing up the product of the catalyst fraction and inlet temperature of each reactor as shown in Eq. (19)[21].

$$WAIT = \sum_{n_{reactor}=1}^N (WAIT_{n_{reactor}})(Wc_{n_{reactor}}) \quad (19)$$

where, $n_{reactor}$ is the reactor number. Figure 4 shows that the propane conversion and propylene yield increase with WAIT. However, the higher WAIT also promotes the side reactions such as cracking and coking. Therefore, lower propylene selectivity is attained and more coke is formed with the increase of WAIT. With the increment of 10 K in WAIT, the propane conversion, propylene yield and coke content increase by 1.28%, 0.87% and 0.14% respectively while the propylene selectivity decreases by 0.70%.and Wc is weight fraction of catalyst.

3.2.2. Hydrogen to hydrocarbon (H_2/HC) molar ratio

H_2/HC molar ratio was varied by ± 0.2 while the other parameters remain the same. Figure 5(a) illustrates the influence of H_2/HC ratio in the feed on the propane conversion and propylene selectivity. An increase in partial pressure of hydrogen has not only decreased the thermodynamic driving force of the reaction, but also kinetically reduced the rate of dehydrogenation reaction due to the competition of hydrogen with propane for the platinum active sites [22]. Furthermore, higher hydrogen concentrations in the feed also enhance the hydrogenolysis side reaction, as reflected by the decreasing of propylene selectivity and yield with the increase of hydrogen concentration. The reduction of H_2/HC molar ratio to 0.2 increases the propane conversion and propylene selectivity by 0.5% and 0.001% respectively.

The influence of H_2/HC molar ratio to the coke formation is shown in Fig. 5(b). Low H_2/HC molar ratio would accelerate the catalyst deactivation by coking. Therefore, an optimum H_2/HC molar ratio should be decided based on the restrictions imposed by the catalyst regeneration system. Lower H_2/HC molar ratio is always preferred as long as the catalyst regeneration system is able to cope the coke removal without significantly shortening the lifetime of the catalysts. A decline of H_2/HC molar ratio by 0.2 from the base case increases the propylene yield and coke content by 0.43% and 0.0079% respectively.

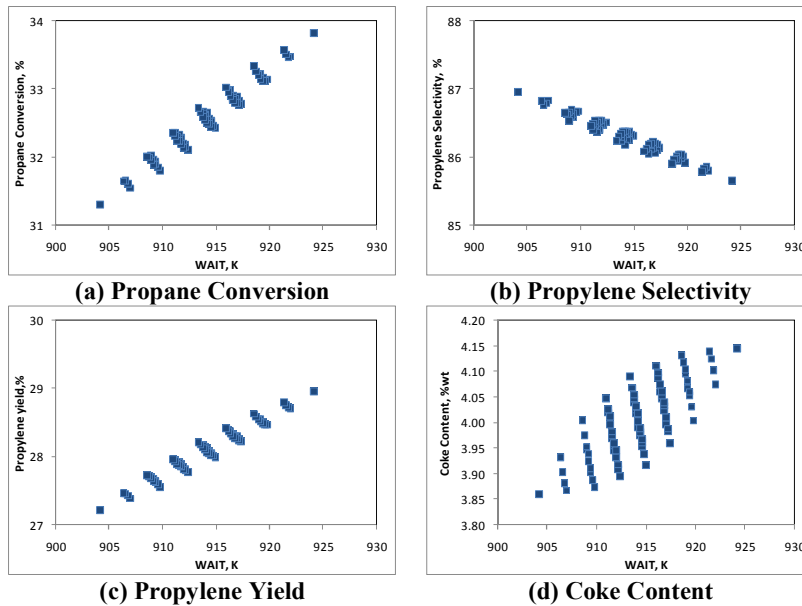


Fig. 4. Influence of WAIT to the Propane Conversion, Propylene Selectivity, Propylene Yield and Coke Content.

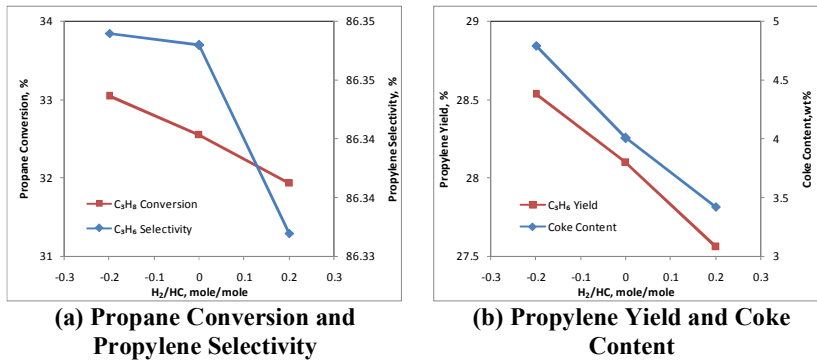


Fig. 5. Influence of H_2/HC Molar Ratio to the Propane Conversion, Propylene Selectivity, Propylene Yield and Coke Content

3.2.3. Catalyst circulation rate (U_s)

Figure 6(a) shows the influence of catalyst circulation rate to the propane conversion and propylene selectivity. Both propane conversion and propylene selectivity increase with the increase of U_s . The increase of U_s has increased the catalyst regeneration rate and reduced the reaction exposure time of the catalyst in RMBR. This has ascribed to the lesser coke content and higher catalyst activity as shown in Fig. 6(b). The increase of propylene selectivity with the increase of U_s might be attributed to the lower propane concentration in the reactor which promotes the hydrogenolysis reaction[10]. The increase in propane conversion and propylene selectivity has resulted an increase in propylene yield with the increase of U_s . With the increment of 100 kg/hr in U_s , the propane conversion, propylene selectivity and yield increase by 0.16%, 0.04% and 0.15% respectively. The corresponding coke content reduction is approximately 0.006%.

Based on the sensitivity study, the optimal operating condition for propane dehydrogenation was obtained. The maximum propylene yield 30.34% was produced when the WAIT was +10 K, H_2/HC was -0.2 and U_s was +100 kg/hr from the base case. This optimum operating condition is practical if the catalyst regeneration system is able to remove the coke content of 4.14%.

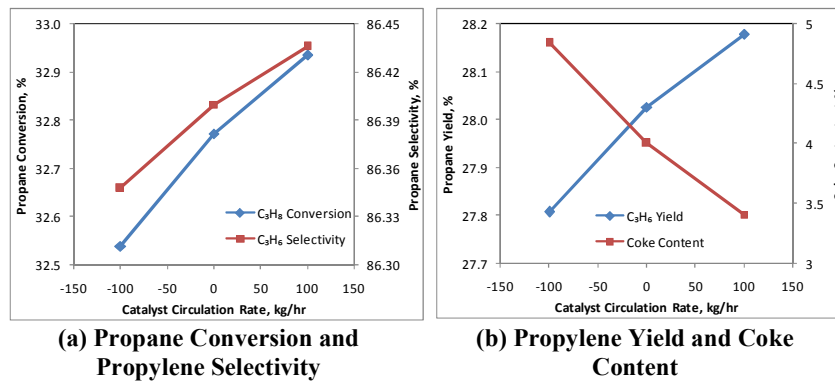


Fig. 6. Influence of Catalyst Circulation Rate to the Propane Conversion, Propylene Selectivity, Propylene Yield and Coke Content.

4. Conclusion

The RMBR model with the optimised kinetic parameters well predicted the reactor outlet composition, reactor temperature, catalyst activity and coke formation. WAIT and H_2/HC molar ratio significantly affect the propane dehydrogenation reaction. Comparing with the base case, a surplus yield can be obtained if the RMBR is operated at higher WAIT and lower H_2/HC . The increase in coke content under this condition can be removed by increasing the U_s considering the hydraulic limitation of the plant. The maximum propylene yield 30.34% was produced when the WAIT was +10 K, H_2/HC was -0.2 and U_s was +100 kg/hr from the base case.

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