

Propylene yield from naphtha pyrolysis cracking using surface response analysis

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ABSTRACT

The study was conducted in the actual world-scale olefin plant with a focus on measuring the impact of identified controlled variables at the steam cracker furnace towards the propylene yield. Surface response analysis was conducted in the Minitab software version 20 using the historical data after the clearance of both the outliers and residuals to ensure the analysis was conducted as normal data. Surface response analysis is a robust mathematical and statistical approach that is having a good potential to be systematically utilized in the actual large-scale olefin plant as an alternative to the expensive olefin simulation software for process monitoring. The analysis was conducted to forecast the maximum propylene yield in the studied plant with careful consideration to select only significant variables, represented by a variance inflation factor (VIF) <10 and p-value <0.05 in the analysis of variance (ANOVA) table. The final model successfully concluded that propylene yield in the studied plant was contributed by the factors of 0.00496, 0.00204, and -3.96 of hearth burner flow, dilution steam flow, and naphtha feed flow respectively. The response optimizer also suggested that the propylene yield from naphtha pyrolysis cracking in the studied plant could be maximized at 11.47% with the control setting at 10,004.36 kg/hr of hearth burner flow, 40,960 kg/hr of dilution steam flow, and 63.50 t/hr of naphtha feed flow. **Polyolefins J (2022) 9: 15-24**

Keywords: Response surface methodology; olefin process; statistical analysis; Minitab; ANOVA.

INTRODUCTION

Olefins or also known as alkenes are aliphatic hydrocarbons with a single C=C bond and tend to participate in various reactions such as hydration, alkylation, and polymerization [1]. Olefins are also the growing essential raw materials that are widely used in the petrochemical industry. They form the basis for many essential applications in plastic, pharmaceutical, cosmetics, adhesives, detergents, insulation, and solvents [2-6].

Olefins like ethylene and propylene are widely

produced in petrochemical plants to meet increasing industrial and consumer needs. Ethylene is by far the most sought-after olefin in the international market, with global output reaching 155 million tonnes per year [4]. However, demands for propylene are also showing significant interest and possibly may outpace ethylene soon [7] due to the continuous increase in worldwide demands. Many studies from various technologies had been conducted for the improvement of propylene yield [5, 8-11] as a result of this rising interest.

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Propylene yield monitoring is critical in the olefin plant as its value corresponds to the income generated from the steam cracker furnace. This study was conducted to evaluate the propylene yield at the newly commissioned olefin plant using naphtha liquid as the feedstock. The studied plant was designed to produce 645 KTA of polymer grade propylene from the pyrolysis cracking in the Short Residence Time (SRT) VII furnace. Performing the study in the large-scale olefin plant is challenging due to the frequent process fluctuation [7, 12-14] caused by dynamic operation in the upstream processes and other disturbances resulting from the large-scale operation.

The high operating temperature in the steam cracker furnace normally above 1,000°C is the result of continuous fuel combustion from the burners. It is required to generate enough heat [15] for the endothermic cracking reaction to happen inside the pyrolysis coils that pass through both the convection and radiation sections in the steam cracker furnace. The high-temperature reaction in the steam cracker furnace causes hydrocarbon bonds to break, resulting in the formation of smaller and unsaturated molecules [16, 17] such as ethylene and propylene. Figure 1 displays the configuration of the steam cracker furnace employed at the studied plant.

The naphtha feed from the upstream plant is introduced at the first convection bank before mixing with dilution steam (DS) at the middle bank. Following Le Chatelier's principles [18, 19], this mix is designed to enhance olefin yield selectivity from naphtha cracking, which is achieved through a reversible reaction between olefins and naphtha mixed with DS. When the chemical composition in

the reaction is changed, the equilibrium shifts to the side that opposes this change. The chemical reaction will attempt to partially counteract the disturbance in balance. As a result of this change, the rate of the reaction, and the products' yield will also change. In this case, reducing the partial pressure of naphtha feed from the DS introduction will therefore significantly increase the olefins yield.

This combined feed will next flow into the radiation section comprised of furnace coils that operated at the elevated tube metal temperature (TMT) reading of 1,050°C to 1,180°C. Different types of steam cracker furnaces will have different coil configurations following the specific olefin licensor's proprietary design. The cracked naphtha from these coils is next quenched in the transfer line exchanger (TLE) [20] at the exit of the radiation section before being transferred to the downstream equipment.

Paraffins, olefins, naphthenes, and aromatics (PONA) compositions in the naphtha feed at the studied plant during the analysis were 60.92 vol%, 1.02 vol%, 25.97 vol%, and 12.09 vol%, respectively. Table 1 represents the true boiling point (TBP) distillation curve for the naphtha feed in the studied plant throughout the study.

The steam cracker furnace is regarded as the most important piece of equipment in the olefin manufacturing process [21]. This due to its performance determines the quality and yield of the olefins produced [22] which later translates to the profit generation for the olefin plant. Besides, it is also essential to remember that safe and stable operation run for steam cracker furnaces is the key to ensure an excellent generation of olefin yield [23, 24] at the olefin plant.

Therefore, any improvement study to increase the olefins yield must carefully consider the safety and stable operation run for the steam cracker furnace. Pyrolysis cracking in the SRT VII furnace is among the most promising technologies to produce the best olefins yield that is currently available in the market [25, 26].

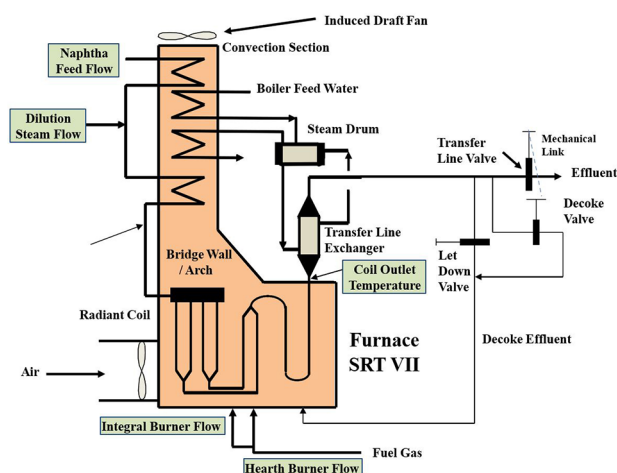


Figure 1. General arrangement for SRT VII with the chosen variables for the study.

Table 1. TBP distillation curve for the naphtha feed.

Parameter	Analysis result	
	Unit	Value
Initial boiling point (IBP)	°C	34.1
30 vol - %	°C	84.4
50 vol - %	°C	105.1
70 vol - %	°C	125.8
Final boiling point (FBP)	°C	166.1

The surface response analysis is a widely used statistical and mathematical approach for modeling the process in which the response of interest is affected by several variables [27] for response optimization. The recent studies conducted at various types of experimental scale furnaces [28-30] also successfully optimize the olefin process using the surface response analysis.

However, surface response analysis was not commonly used in the large-scale olefin plant. In the event of a process disruption that requires troubleshooting and rectification, the olefin plant normally relies on the process simulation software furnished by the olefin licensor. The software was undeniably robust and widely applied in the olefin plants. However, it comes with a high price, complex and sometimes with the limitation in some of the applications to safeguard the proprietary design by olefin licensors. This contributed to the challenging process monitoring by Process Technologist and Operations personnel in the olefin plant.

This study is significant as it provides a guideline for reliable model development utilizing available statistical software in the market which is cheaper and therefore saving the overall company's operating expenditures (OPEX). Statistical software is also practical and easier to be used by Operations personnel compared to utilizing the complex and restricted simulation software provided by some of the olefin licensors which are currently being practiced by most of the olefin plants worldwide.

Besides, the surface response analysis also may be analyzed from the actual available historical data in the large-scale olefin plant. Therefore, it directly represents the actual plant condition and may provide confidence towards the final model established for the process monitoring.

EXPERIMENTAL

Equipment/Tools

The SRT VII which adopting the technology designed by Lummus Technology Heat Transfer (LTHT) with 93 t/hr of naphtha feed processing capacity was utilized for the study. The main heater and all supporting auxiliaries at SRT VII were ensured in a stable and healthy condition before data selection, extraction, and analysis being conducted. This study was also carried out during SRT VII at the start of run

(SOR) condition where the development of coke in the furnace coils was expected low.

The data were collected with the PONA composition in the naphtha feed to the SRT VII did not exceed 5% of variations throughout the study duration. The historical data were extracted using Process Information Management System (PIMS) software, PI Process Book version 2015. The relevant process instrumentation at the identified locations was also calibrated for reliable data extraction. The surface response analysis was then conducted adopting Minitab software version 20 to develop the final equation model. The relevant 2D and 3D tools in Minitab were also used to evaluate the relationship and impact of each studied variable towards propylene yield.

Methodology

5 variables were chosen as the independent variables to the propylene yield which were hearth burner flow, integral burner flow, dilution steam flow, naphtha feed flow, and coil outlet temperature (COT). The variable selection in this study was based on the most frequently adjusted variable in the studied plant. They were chosen to give appreciation to the Operations personnel in understanding the operating behavior of the normally adjusted variables towards realizing the maximum propylene yield.

The analysis were conducted on 24th January 2020, 1900 hrs to 2nd February 2020, 1200 hrs (207 hrs total). The data were extracted from the PI Process Book on an hourly basis (average, time-weighted), continuously with a total of 1,242 data (represented by 1 dependent and 5 independent variables). Table 2 shows the tags and units used for the selected variables in the surface response analysis.

The data stability verification was first conducted utilizing three tools namely box plot, individual-moving range (I-MR), and run chart. The data normality verification was then continued utilizing the

Table 2. Controlled variable for the analysis.

Tag	Variables	Unit	Data type
Y1	Propylene yield	t/hr	Dependent
A	Hearth burner flow	kg/hr	Independent
B	Integral burner flow	kg/hr	Independent
C	Dilution steam flow	kg/hr	Independent
D	Naphtha feed flow	t/hr	Independent
E	Coil outlet temperature	°C	Independent

graphical summary and normality test plot. All 1,242 data were analyzed using these 5 tools in Minitab to identify the normality and stability of the collected data before the statistical evaluation was conducted.

Surface response analysis was adopted to these variables in the studied steam cracker furnace via the historical design of experiment (DOE) methodology as normal data, without Box-Cox data transformation if both stability and normality test passed. The insignificant variables were eliminated one-by-one in the surface response analysis, starting with the 2-way interactions, squares, and lastly followed by linear relations. These sequences had to be followed to keep the surface response analysis model in the hierarchical order.

The variable removal began for the variable with the highest variance inflation factor (VIF) until all variables achieve VIF <10. Once realized, the variable elimination was conducted to the remaining variables with the highest p-value and continued until all variables recorded p-values <0.05. The normally accepted threshold for p-value is <0.05 for the statistical analysis at the stable and controlled experiment. However, the value of 0.05 was also chosen in this study regardless it was conducted under the actual fluctuating process condition to comfortably satisfy the 95% confidence level for the final model.

In general, all variables with p-values <0.05 were removed in the one-by-one variable elimination. However, for the variable with the p-value >0.05 in the linear relation that was still appeared in the square or 2-way interaction, it would be incorporated into the final model. This was required to sustain the hierarchical model in the surface response analysis. This exemption will be permitted if the model had a strong R-squared of 75% or greater [31, 32], which will be determined on a case-by-case basis.

Interaction plot and contour plot were also utilized from the final surface response model to see the relationship of each variable in the model towards propylene yield, followed by the response optimizer graphical tool. Response optimizer in Minitab is useful to portray the combination of variable settings that jointly optimize a set of responses for the statistical model. In this graphical tool, the response was presented in the low and high operating ranges for each significant independent variable to achieve the maximum dependent variable.

RESULTS AND DISCUSSION

The stability test from Minitab revealed no outliers in the box plot analysis, seven residuals found in the I-MR Chart, and a p-value for ‘clustering’ was failed at <0.005 in the run chart (p-value for ‘mixtures’, ‘trends’, and ‘oscillation’, on the other hand, were successful with values of 1.000, 0.386, and 0.614, respectively). Due to at least one of the three stability checks was passed, the data was deemed stable.

Both the graphical summary and the normality test were initially failed for data normality verification, with a p-value of <0.005. In the subsequent analysis, a total of 27 poor (Y1– propylene yield) data (residuals, outliers, etc) were deleted from the source data, resulting in the final p-value of >0.05 in both the graphical summary and the normality test. After these outliers were removed, the updated data was observed as normal.

The surface response analysis was conducted using standard methodology without Box-Cox data transformation as both the stability and normality tests were passed. Table 3 presents the outcome of the final surface response analysis, which was conducted at the 15th analysis where a total of 8 squares, 4 2-way interactions, and 2 linear relations were removed in this analysis.

From the determined normal data, the 1st surface response analysis was performed to establish the overall relationship between each variable. The exclusion of variables with VIF >10 was continued in the 2nd through 7th surface response analysis. After the 7th analysis, the VIFs for the remaining variables were successfully decreased to <10. The variables with the p-values >0.05 were removed in the 8th through 14th

Table 3. Summary of ANOVA for the final surface response model.

Source	DF	Adj SS	Adj MS	F-value	p-value
Model	6	2.64296	0.440494	93.72	0.000
Linear	3	0.15785	0.052617	11.20	0.000
A	1	0.10749	0.107494	22.87	0.000
B	1	0.03416	0.034157	7.27	0.008
C	1	0.05239	0.052389	11.15	0.001
Square	1	0.71495	0.714950	152.12	0.000
A*A	1	0.71495	0.714950	152.12	0.000
2-Way	2	0.04623	0.023115	4.92	0.008
A*B	1	0.04623	0.046229	9.84	0.002
A*C	1	0.02912	0.029120	6.20	0.014
Error	175	0.82250	0.004700	-	-
Total	181	3.46546	-	-	-

analyses. The final model was established from the 15th surface response analysis where conditions for both VIF and p-value were met.

In summary, the elimination sequence for the 2nd through 14th surface response analyses was 2nd; (A – hearth burner flow)*(E – COT) with VIF: 178.50, 3rd; (C – DS flow)*(E – COT) with VIF: 52.42, 4th; (B – integral burner flow)*(E – COT) with VIF: 42.89, 5th; (D – naphtha feed flow)*(E – COT) with VIF: 40.38, 6th; (A – hearth burner flow)*(D – naphtha feed flow) with VIF: 39.22, 7th; (B – integral burner flow)*(D – naphtha feed flow) with VIF: 28.59, 8th; (A – hearth burner flow)*(B – integral burner flow) with p-value: 0.745, 9th; (B – integral burner flow)*(C – DS flow) with p-value: 0.151, 10th; (C – DS flow)*(C – DS flow) with p-value: 0.869, 11th; (D – naphtha feed flow)*(D – naphtha feed flow) with p-value: 0.0736, 12th; (B – integral burner flow)*(B – integral burner flow) with p-value: 0.534, 13th; (E – COT) with p-value: 0.726, and 14th; (B – integral burner flow) with p-value: 0.080. These eliminations were following the sequence that had been discussed in Section 2.2 starting from the 2-way, squares, and finally to linear relations to maintain the hierarchical order of the surface response model.

The R-squared for the equation model established from the 15th surface response analysis was recorded at 76.27%. This final model was accepted where it met the advised R-squared value of 75% or higher [31, 32]. Besides, given that the data was extracted from an actual world-scale olefin plant where process variation was normally faced, this result was also excellent. The final model developed from the surface response analysis is shown in Equation 1.

$$Y1=61.9+0.00496A+0.00204C-3.96D+0.000001A \times A-0.000001A \times C+0.000098C \times D \quad (1)$$

In general, 3 variables were identified as significant for (Y1– propylene yield) in the final model. The (D - naphtha feed flow) was showing the biggest impact with the factor of -3.96 compared to (A - hearth burner flow) and (C - DS flow) with the factors of 0.00496 and 0.00204 respectively. These determining factors were good as a guide for operating the studied SRT VII base on the most important variable to reach the highest (Y1– propylene yield).

Figure 2 portrays the probability plot for the

accounted residuals during the data validation to the final equation model. The p-value for the residuals from the probability plot was found at 0.139, which was >0.05. In explaining the validity of the final equation model, this high p-value demonstrated a good data distribution and acceptable model prediction for the final equation model. The residuals in this plot represent the comparison between the final model's projected data to the actual data in the studied plant analyzed from the surface response analysis. The acceptance of the final equation models from the surface response analysis was validated by a p-value >0.05 in this probability plot.

Olefin plants around the world are normally used to run the furnace at the higher COT to increase the propylene yield. It was also endorsed by other studies and reviews which were carried using the process simulation and pilot-scale plant [33-35] where the higher COT may result in the better olefin yields. However, (E – COT), on the other hand, was not found as one of the significant variables in this study.

From the one-by-one variable elimination, most of the relationship established with (E – COT) was removed due to the high VIF recorded from the analysis of variance (ANOVA). This was due to the high multi-collinearity response from (E – COT) with other parameters that were having similar behavior, for example; (A – hearth burner flow) and (B – integral burner flow). In this case, the higher (A – hearth burner flow) and (B – integral burner flow) will result in the higher temperature inside the furnace's radiation section which translated to the higher (E – COT) reading.

As surface response analysis found (E – COT) was less significant compared to (A – hearth burner flow)

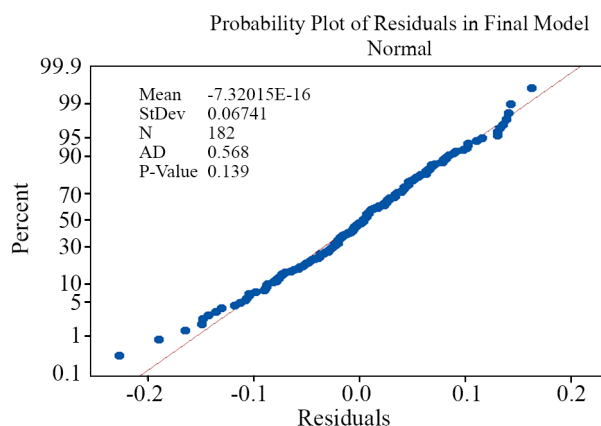


Figure 2. Probability plot of the residuals from the final equation model.

and (B – integral burner flow), its VIF value was recorded higher and therefore was removed during one-by-one variable elimination. It was important to ensure all significant variables and relationships achieve $VIF < 10$ to remove the high multicollinearity relationship between variables for the reliable final model in the surface response analysis. Besides, the recent study that has been conducted using statistical analysis in the large-scale olefin plant [36] for another important olefin (ethylene yield) also showed that the COT was also not accepted as the significant variable due to its high VIF value.

Figure 3 illustrates the interaction plot for the significant variables from the surface response analysis, together with their fitted means plot. The estimated average reaction at different levels of each significant variable was summarised in this fitted means plot, where leveraging over the levels of the other significant factors.

The square and 2-way interactions in the final surface response model were observed for (A – hearth burner flow), (C – DS flow), and (D – naphtha feed flow) in this interaction plot. Operating the higher (A – hearth burner flow) at 10993.9 kg/hr initially would result in the higher (Y1– propylene yield), however, due to its square and 2-way relation, the (Y1– propylene

yield) will reduce after reaching its optimum operating condition as shown in the interaction plot.

Besides, operating (D – naphtha feed flow) at the higher process range showing a clear relation in generating the higher (Y1– propylene yield). The interaction plot between both (D – naphtha feed flow)*(C – DS flow) and (C – DS flow)*(D – naphtha feed flow) were also showing the higher (Y1– propylene yield) at the higher (D – naphtha feed flow), 63.50 t/hr. However, a small 2-way interaction was also established at the lower (D – naphtha feed flow), 59.37 t/hr as shown in the interaction plot. In general, this plot is significant in showing the optimum condition of each significant variable for the process monitoring and optimization at the studied plant.

Figure 4 represents the surface plot of significant variables from the final model in achieving (Y1–propylene yield). The values for non-tested variables were kept constant in this plot at 10,499.11 kg/hr of hearth burner flow, 40,599.17 t/hr of dilution steam flow, and 61.43 t/hr of naphtha feed flow. On the surface plot, the light was also configured to represent the (Y1– propylene yield) at the optimum condition.

Figures 4 (a) and 4 (b) show that a lower (A – hearth burner flow) combined with a higher (C – DS Flow) or (D – naphtha feed flow) will result in a greater (Y1–

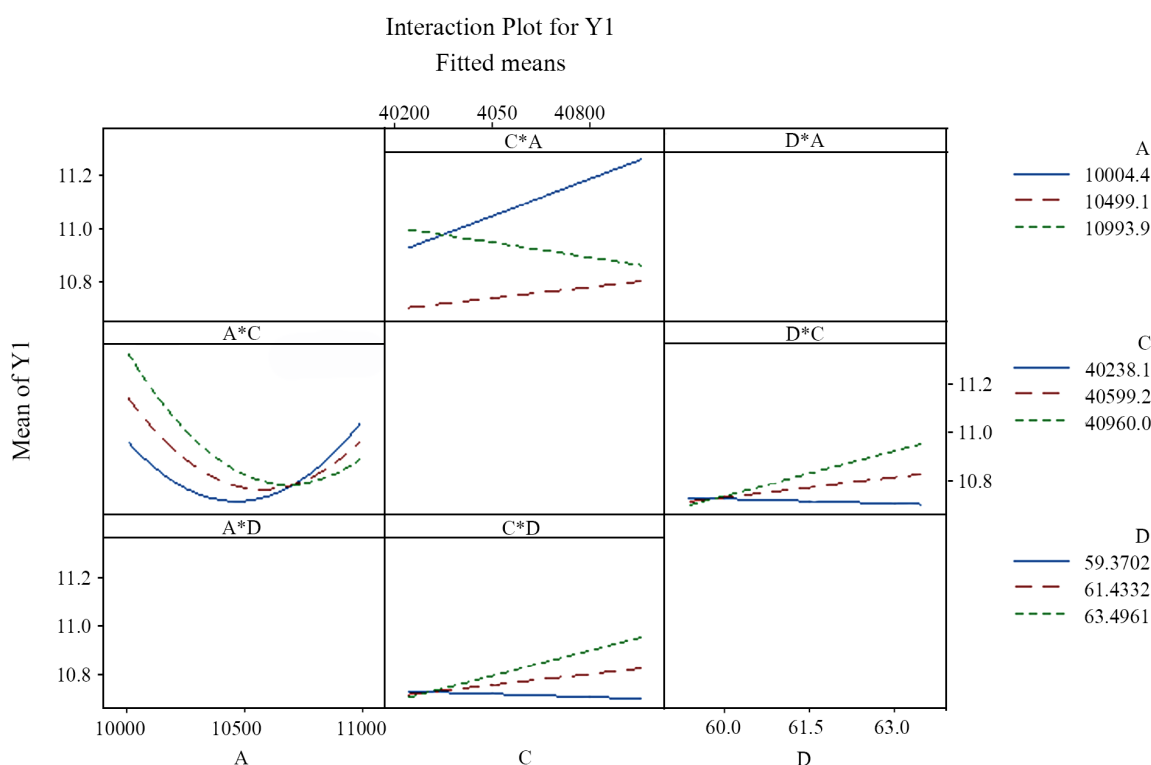


Figure 3. Interaction plot of dependent variable Y1 with the process condition of significant independent variables; A, C, and D.

propylene yield). However, because of the considerable quadratic and 2-way relationship of (A–hearth burner flow), the (Y1– propylene yield) will be significantly reduced after reaching the certain limit identified in the surface plot. From the plot, operating (A – hearth burner flow) at 10,500 kg/h with the lower

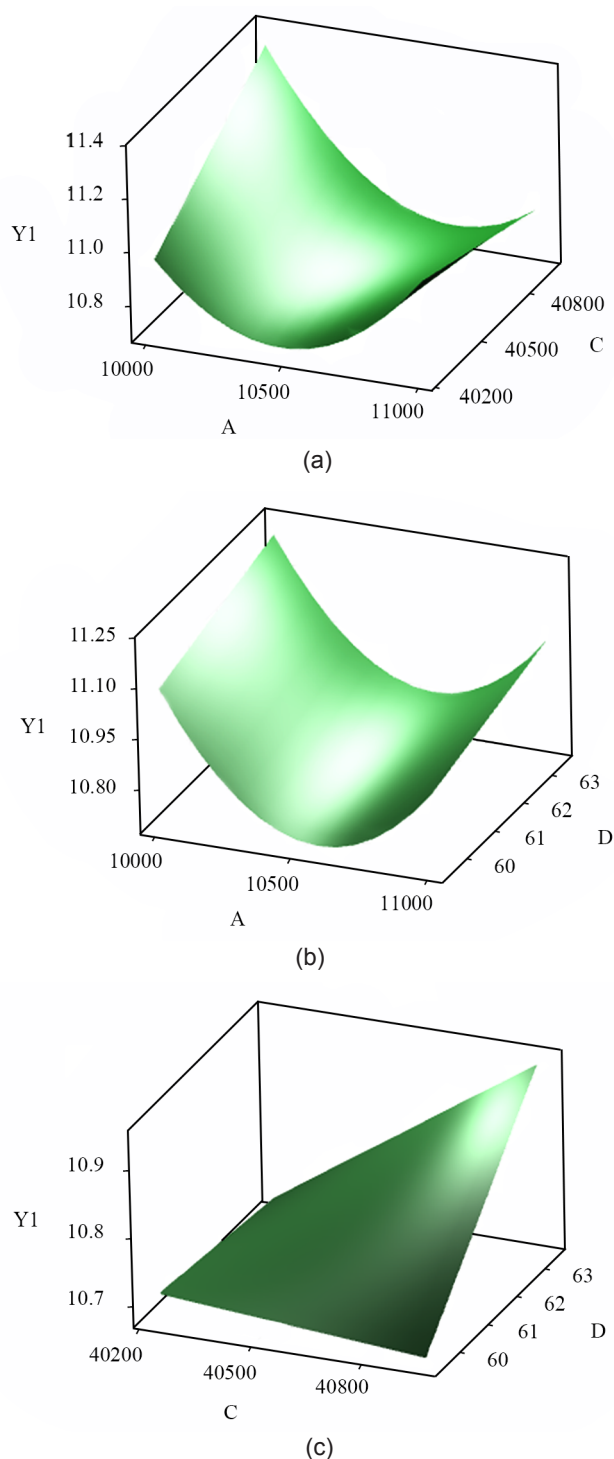


Figure 4. The relationship of significant variables in form of the 3D surface plot towards Y1 ; (a) A vs C, (b) A vs D, and (c) C vs D.

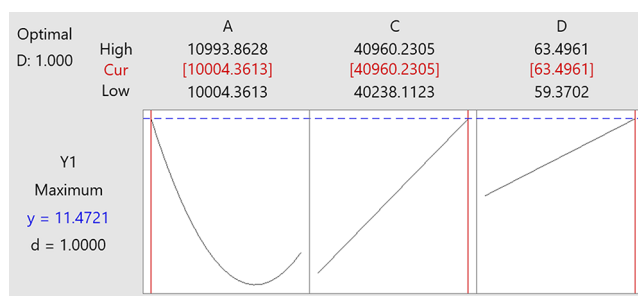


Figure 5. Prediction of maximum propylene yield with the process setting for the significant variables using surface response optimizer.

Table 4. Maximum propylene yield using the multiple response prediction.

Response	Fit	SE fit	Confidence	
			95% CI	95% PI
Y1	11.4721	0.0912	(11.2922, 11.6520)	(11.2470, 11.6972)

(C – DS flow) was more critical to be avoided to ensure the higher (Y1– propylene yield) compared to the combination with (D – naphtha feed flow).

Figure 4 (c) shows the high (Y1– propylene yield) also would be achieved through the combination of the higher (C – DS flow) and higher (D – naphtha feed flow). Operating lower (C – DS flow) with lower (D – naphtha feed flow) must be prevented as it would result in the worst (Y1– propylene yield) as shown from the plot. In summary, this surface plot is beneficial to show the clearer relationship in form of a 3D diagram for the overall process understanding at the studied plant.

Figure 5 depicts the configuration of significant factors in the surface response analysis to obtain maximum (Y1– propylene yield) in the studied plant, while Table 4 shows the multiple response prediction for the final model at the 95% confidence level.

The response optimizer plot revealed that the best control setting to accomplish maximized (Y1– propylene yield) at 11.47% was 10,004.36 kg/hr of hearth burner flow, 40,960 kg/hr of dilution steam flow, and 63.50 t/hr of naphtha feed flow. The low and high range setting in this plot was also to be observed and monitored closely by the studied plant to ensure the generation of (Y1– propylene yield) at the best confidence interval (CI) and prediction interval (PI) as shown in Table 4.

CONCLUSION

Surface response analysis using Minitab software version 20 successfully established the reliable final equation model with the R-squared stood at 76.27% and supported by the validated data for normality plot of residuals at the p-value of 0.139. The propylene yield at the studied plant could be maximized at 11.47% through a careful adjustment to the identified significant variables which were naphtha feed flow (factor of -3.94), hearth burner flow (factor of 0.00496), and dilution steam flow (factor of 0.00204).

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CONFLICTS OF INTEREST

None declared

ABBREVIATIONS

ANOVA	Analysis of Variance
CI	Confidence Intervals
COT	Coil Outlet Temperature
DOE	Design of Experiment
DS	Dilution Steam
I-MR	Individual-Moving Range
IDF	Induced Draft Fan
KTA	Kilo Tonne Per Annum
LTHT	Lummus Technology Heat Transfer
OPEX	Operating Expenditure
PI	Prediction Intervals
PIMS	Process Information Management System
PONA	Paraffins, Olefins, Napthenes, Aromatics
RSM	Response Surface Methodology
SRT	Short Residence Time
TBP	True Boiling Point
TLE	Transfer Line Exchanger
TMT	Tube Metal Temperature
VIF	Variance Inflation Factor

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