Propylene Yield Assessment utilizing Response Surface Methodology for Naphtha Pyrolysis Cracking

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Abstract: The study was carried out at the steam cracker furnace, Short Residence Time (SRT) VII type to investigate the impact of selected input variables on the Propylene Product Yield. The statistical analysis was conducted as normal data after successful outliers and residuals clearance using data stability and normality verification tests. The Response Surface Methodology (RSM) analysis was performed to establish the reliable Propylene Product Yield model using statistical software, Minitab version 21. RSM is a robust statistical approach that may utilize the available statistical software that is cheaper and practical to be used by Operations personnel at the olefin plant. It is an excellent alternative to the Olefins Licensor's simulation software that is currently used by the olefin plant worldwide. The analysis of variance (ANOVA) table was utilized to select only significant variables with a p-value of <0.05 for the model. The established final model indicated that Coil Outlet Temperature (COT) was the most important input variable for the Propylene Product Yield generation with a factor of 49.4, compared to -7.76 for Flow of Naphtha, -4.3 for Flow of Integral Burner, 0.292 for Flow of Dilution Steam, and 0.05947 for Flow of Hearth Burner. Besides, the Response Optimizer evaluated that the Propylene Product Yield at the studied SRT VII could be maximized at 13.24% by controlling the process operating parameters with; Flow of Hearth Burner at 9476 kg/hr, Flow of Integral Burner at 609 kg/hr, Flow of Dilution Steam at 40960 kg/hr, Flow of Naphtha at 63.50 t/hr, and COT at 810 °C.

Keywords: Minitab; Olefins; Response Surface Methodology; Naphtha Cracking; Manufacturing

1. Introduction

Pyrolysis cracking, also known as thermal cracking, is a high-temperature cracking reaction that leads to the formation of tiny unsaturated molecules[1, 2] like olefins in a petrochemical plant. Olefins, also known as alkenes, are often produced by the petrochemical industry. They are aliphatic hydrocarbons established from a single C–C bond, and serve as the basis for a variety of crucial consumer applications, including solvents, detergents, polymers, pharmaceuticals, and skincare products[3-5].

Ethylene and propylene are examples of olefin products generated by pyrolysis cracking at elevated temperatures[2, 6, 7] from the olefin plant. With a global production of 155 million tonnes per year[3], ethylene appears to be the most in-demand olefin in the global market. However, the continuing rise in global demand for propylene products may also possibly overtake ethylene[8] in the future market.

The research on propylene manufacturing technologies is also gaining significant interest from researchers such as propane dehydrogenation[9], olefin metathesis[10], Fluidized Catalytic Cracking (FCC)[4], and reaction optimization in the FCC process[11] due to the increase in its worldwide demand. Currently, the most prevalent method for generating a viable Propylene Product Yield is through the pyrolysis cracking in the olefin plant through a steam cracker furnace.

Steam cracker furnace is the most critical equipment[12] to be monitored and analyzed by the Operations personnel in the olefin manufacturing plant. The yield of the olefins[13] is highly influenced by its performance, leading to sustainable profit generation for the olefin plant. The Short Residence Time (SRT) VII is an example of a reliable steam cracker furnace in the market[14, 15] to produce a good olefins yield.

Process optimization implies fine-tuning a process to maximize a specific set of parameters while adhering to a specific set of restrictions. It is often used to maximize throughput, efficiency, and production cost. Previous research had successfully demonstrated different process simulations to improve the production of olefins[16-19]. These studies were effectively conducted in a lab-scale experiment that manipulated various controllable factors in the olefin process, which resulted in the significant optimization of olefin production.

Different operational variables are actively being experimented with to improve the olefin process. The majority of the research was completed in the lab, with some verification performed at the small pilot plant. The subject of analyses include the study on profit enhancement[20, 21], operational expenditure (OPEX) saving[17, 22-24], production rate improvement[25], key product yield maximization[19], power consumption optimization[26, 27], utility saving[25], exergy destruction minimization[28, 29], ethylene leakage reduction[27], hydrogen recovery improvement[27], CO_2 emissions minimization[26], Coil Outlet Temperature (COT) enhancement[18, 30, 31], and multiple optimization initiatives[19, 25, 26, 28, 29, 32, 33].

Statistical analysis may be used to analyze the data systematically, where its effectiveness was already proven in various studies[34-38] for data evaluation, monitoring, and process optimization. Response Surface Methodology (RSM) is an example of the robust statistical analysis used for simulating and evaluating processes where multiple factors influence the response of interest for response optimization[39]. Statistical software was proven robust in conducting various analyses at the normal type of furnace[40-42]. However, it was not widely used for complex furnace types such as steam cracker furnaces.

The RSM analysis in one of the special type furnaces was started with the vacuum furnace[43]. However, the study did not involve the olefin process. The later study[44] initiated the usage of Design of Experiment (DoE) in the steam cracker furnace. It utilized statistical analysis to improve olefin yield, conversion, and selectivity (from steam amount). The study using RSM was then continued[45] in the pilot-scale experiment, with a focus on the effect of conversion selectivity towards olefins yield.

However, the study to use the RSM analysis in steam cracker furnaces did not actively continue due to most researchers focusing on robust simulation software. However, with the recent improvement to the statistical software, it has a high prospect to be widely used in the olefin plant due to its practicality, simplicity, lower price, and robust performance[46]. In the actual plant condition, the Operations personnel always prefers to evaluate the process using less complex, user-friendly, and cheaper software without the usage limitation.

Currently, the reliance of the Olefins plant on the Olefins Licensor's software is due to its robustness, the mature field of the olefin process, and trust in the licensor's expertise. However, in the actual process at a large-scale steam cracker furnace, the statistical software has also a high potential to be further explored in terms of its practicality in usage and robustness in carrying out large data analysis directly by Operations personnel in an actual olefin plant. It will help Operations personnel to evaluate the frequent process fluctuation in the olefin plant[8, 47-49] and decide on process improvement.

The focus of this study is to assess the Propylene Product Yield from the steam cracker furnace at a newly commissioned olefin plant using naphtha liquid as a feedstock. It is essential as it offers guidelines for creating a credible model using commercially accessible statistical software, with a focus on the RSM. In comparison to the intricate and costly simulation software offered by the Olefin Licensors, it is less complicated, more useful, and simpler to be used by operations employees in the Olefins plant

2. Methods

2.1 Plant Equipment and Analysis Tools

The analysis was conducted in a large-scale Short Residence Time (SRT) VII type steam cracker furnace. The pyrolysis cracking at the studied SRT VII was designed to yield 645 KTA of propylene (polymer-grade) at the plant. The SRT VII furnace's layout is shown in Fig. 1, along with the location of the studied variables.

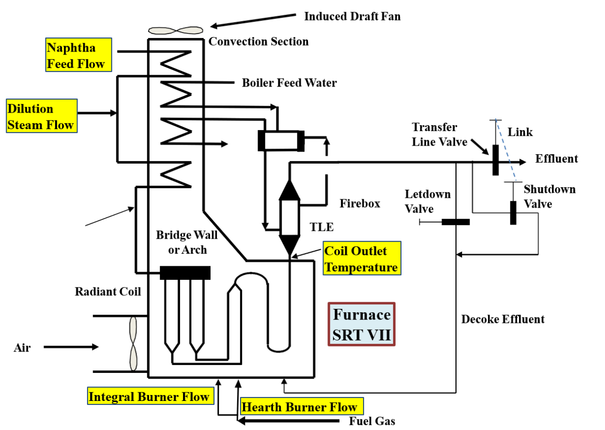

Fig. 1: The general arrangement of the SRT VII with the input variables for RSM analysis

The input variables in Fig. 1 were represented by *A* for Flow of Hearth Burner (HB), *B* for Flow of Integral Burner (IB), *C* for Flow of Dilution Steam (DS), *D* for Flow of Naphtha, and *E* for Coil Outlet Temperature (COT). The representation of input variables in *A* – *E* was required to ease the analysis in Minitab and minimize the possibility of the mistake during one-by-one variable elimination from analysis of variance (ANOVA) using

RSM analyses. The SRT VII was designed to process the continuous 93 t/hr of Naphtha feed during the normal process. The Paraffins, Olefins, Naphthenes, and Aromatics (PONA) contents in the Naphtha feed during the study duration were recorded at 61 vol%, 1 vol%, 26 vol%, and 12 vol%, respectively.

The relevant statistical and mathematical analyses were conducted utilizing Minitab Software version 21. All analyses from the data verification until the evaluation of maximum *Y1* (Propylene Product Yield) were carefully conducted using statistical software. It was to ensure the reliable equation model was successfully established from the final RSM analysis.

2.2 Methodology

The plant data were collected hourly (average, time-weighted) from 24th January 2020, 7.00 pm to 2nd February 2020, 12.00 pm (207 hrs), totaling 1,242 data points. These data were represented by one input and five output variables utilizing the PI Process Book - Process Information Management System (PIMS) Software version 2015.

The data stability was initially assessed to the collected data by employing three tools: Run Chart, Box Plot, and Individual-Moving Range (I-MR). Once completed, the Graphical Summary and Normality Test were then utilized to perform the data normality verification. The stability and normality of the collected data were evaluated using these five Minitab tools on all 1,242 data before performing the statistical analysis utilizing RSM analysis. These analyses were essential to guarantee the final RSM model was established from the collection of reliable data at the studied plant.

After the data passed the normality and stability tests, RSM analysis was carried out on the identified variables using a normal historical design of experiment (HDoE) approach, without any data transformation. Through RSM analysis, the insignificant variables were eliminated one at a time, starting with the Two-way, Squares, and ending with the Linear relations.

In the RSM analysis, variable removal started with the input variable with the highest p-value and continued until all variables had p-values <0.05. However, to maintain the hierarchical model in RSM, the variable with the p-value >0.05 in the linear relationship would be accepted if it was still seen in the square or two-way relation. This exemption would also be made if the model's R-squared was high at 75 percent or more[50-53].

Contour Plot and Interaction Plot were also adopted for the final RSM model to evaluate the correlation between each significant input and output variable. Finally, the Response Optimizer was used to forecast the maximum *Y1* (Propylene Product Yield) with its best operating condition at the studied SRT VII.

The Response Optimizer, in general, is a useful tool for visualizing a group of variable settings to optimize the set of responses for a statistical model. For each significant input variable, the targeted response was represented in the low and high operating ranges in order to obtain the highest *Y1* (Propylene Product Yield) output variable throughout this graphical tool.

3. Results and Discussion

The stability test showed that no outlier was found from the Box Plot analysis, seven residuals were identified in the I-MR Chart, and one plot in the Run Chart failed with the p-value <0.05 (Clustering: p-value <0.005). However, other plots in Run Chart were successful with p-values >0.05 (Mixtures: p-value 1.000, Trends: p-value 0.368, and Oscillation: p-value 0.614). The data was concluded as stable since at least one of the three stability tests was successful, which was Box Plot analysis.

Both the Graphical Summary and the Normality Test initially failed the normality verification as they both recorded p-values of <0.005. In the subsequent analysis, 27 bad *Y1* (Propylene Product Yield) data, including residuals and outliers, were eliminated from the source data. After eliminating the bad data, the final p-values of >0.05 were successfully obtained in both the Graphical Summary and the Normality Test. This filtered data was concluded as the normal data.

As both stability and normality evaluations were passed, the RSM analysis was performed using the normal methodology without Box-Cox data transformation. Table 1 displays the findings of the 7th analysis (final RSM), where a total of 2 Square and 4 Two-way relations were removed during one-by-one variable elimination.

Table 1. ANOVA for the final model.

Source	DF	Adj SS	Adj MS	F-value	p-value
Model	14	3.11311	0.222365	43.21	0.000
Linear	5	0.63088	0.126176	24.52	0.000
A	1	0.42254	0.422535	82.11	0.000
B	1	0.00699	0.006991	1.36	0.245
C	1	0.04676	0.046756	9.09	0.003
D	1	0.10138	0.101376	19.70	0.000
E	1	0.15357	0.153575	29.84	0.000
Square	3	0.35552	0.118505	23.03	0.000
*A*A*	1	0.23952	0.239518	46.54	0.000
*B*B*	1	0.14583	0.145830	28.34	0.000
*E*E*	1	0.03102	0.031021	6.03	0.015
2-Way	6	0.46143	0.076905	14.94	0.000
*A*B*	1	0.34190	0.341898	66.44	0.000
*A*C*	1	0.10122	0.101220	19.67	0.000
*B*D*	1	0.05200	0.052002	10.10	0.002
*B*E*	1	0.05932	0.059324	11.53	0.001
*C*D*	1	0.04207	0.042071	8.18	0.005
*C*E*	1	0.02615	0.026149	5.08	0.025
Error	174	0.89545	0.005146		
Total	188	4.00856			

The final model's R-squared was found to be 77.66%. It indicated that the model accounted for 77.66% of the variability in the data. Considering that the data was collected from the actual large-scale plant with dynamic process conditions in the upstream process[8, 47, 54, 55], this number was good. Furthermore, the established R-squared value was also sufficient for the research, which was recommended at 75% or above[50-53].

One variable with a p-value of higher than 0.05 was accepted into the final model, which was *B* (Flow of IB), with a recorded p-value of 0.245. Besides, its F-value also approached the value 1, which also indicated a less significant variable for the final model. However, this variable was accepted into the model as its relation existed in Square relation; *B* (Flow of IB) * *B* (Flow of IB), and also Two-way relation; *A* (Flow of HB) * *B* (Flow of IB), *B* (Flow of IB) * *D* (Flow of Naphtha), and *B* (Flow of IB) * *E* (COT).

Its inclusion was required to maintain the hierarchical model in RSM for the establishment of a reliable final model. Besides, it was also supported by the high R-squared value of 77.66 %. Equation 1 depicts the equation model derived from the final RSM analysis.

$$Y1 = -24755 + 0.05947\,A - 4.30\,B + 0.292\,C - 7.76\,D + 49.4\,E + 0.000001\,A^2 - 0.001471\,B^2 - 0.02440\,E^2 - 0.000063\,AB - 0.000001\,AC + 0.00603\,BD + 0.00782\,BE + 0.000105\,CD - 0.000358\,CE \quad (1)$$

The model showed that all five variables were considered in the final model, including *B* (Flow of IB). Understanding the operational behavior of the SRT VII at the studied plant in terms of *Y1* (Propylene Product Yield) generation would be made easier with the help of the coefficient factors in Equation 1.

The model also showed that *E* (COT) was the most impactful variable towards the *Y1* (Propylene Product Yield) with a significant factor of 49.4 compared to other variables, which were 0.05947, -4.30, 0.292, and -7.76 for *A* (Flow of HB), *B* (Flow of IB), *C* (Flow of DS), and *D* (Flow of Naphtha) respectively.

COT was the preferred variable used in olefin plants for monitoring the steam cracker furnace worldwide and was also suggested by various studies and reviews[3, 18, 30, 31, 56, 57]. Besides, increasing COT in achieving the higher olefins yield was proven successful from the experimental scale furnace and process simulations in other research[18, 19, 58].

Equation 1 from this study was significant in showing the factorial impact of the COT mathematically based on the actual olefin plant condition where process fluctuation was usually observed[8, 47, 54, 55].

Fig. 2 shows the Interaction Plot for the significant variables in the RSM model with their fitted means plot. This plot summarizes the estimated average response for the significant variable at different levels of one factor while averaging over the levels of the other variable. This Interaction Plot also confirmed Square and Two-way relations in the final RSM model for all variables. *A* (Flow of HB) * *B* (Flow of IB), *B* (Flow of IB) * *D* (Flow of Naphtha), *B* (Flow of IB) * *E* (COT), and *C* (Flow of DS) * *E* (COT) showed the most significant Two-way relations in achieving the highest *Y1* (Propylene Product Yield).

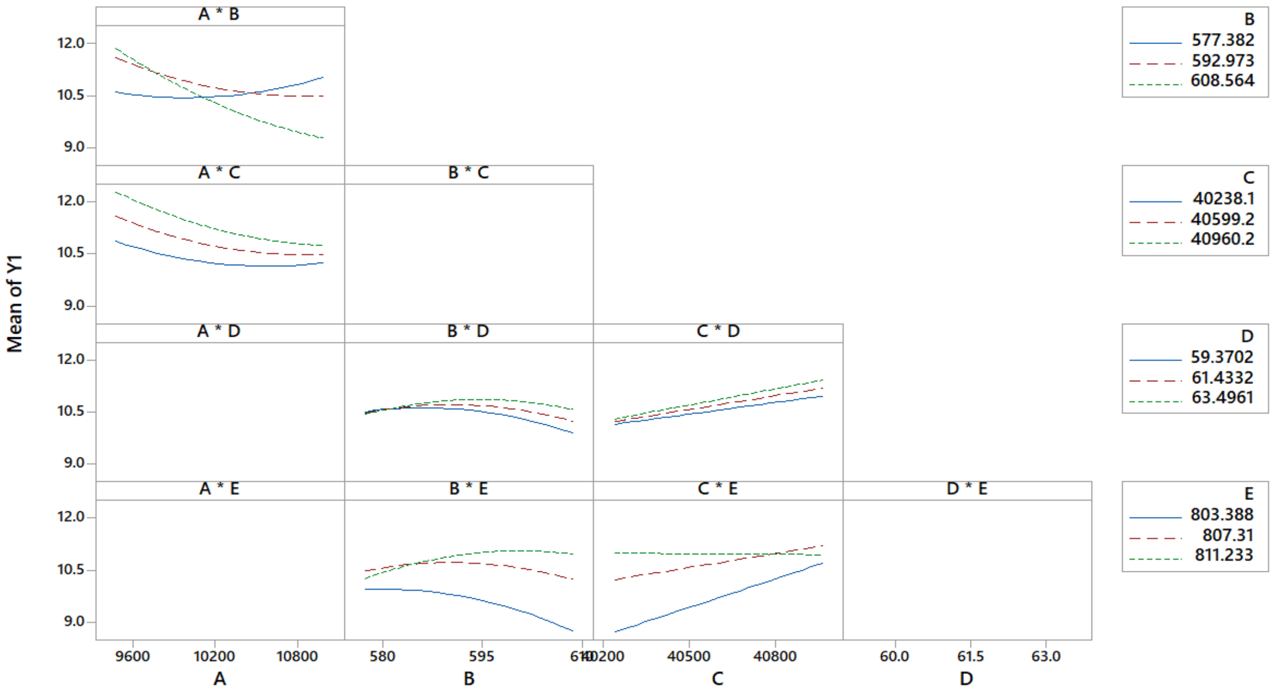

Fig. 2: Interaction Plot of identified significant variables; *A* (Flow of HB), *B* (Flow of IB), *C* (Flow of DS), *D* (Flow of Naphtha), and *E* (COT) towards *Y1* (Propylene Product Yield).

These relations were also seen in the final RSM model where their coefficient factors showed the highest contribution in the Two-way relation by the factor of 0.000063 *A* (Flow of HB) * *B* (Flow of IB), 0.00603 *B* (Flow of IB) * *D* (Flow of Naphtha), 0.00782 *B* (Flow of IB) * *E* (COT), and 0.000358 *C* (Flow of DS) * *E* (COT). The combinations of these variables were also necessary to be observed frequently in the studied plant as their changes would contribute to the bigger *Y1* (Propylene Product Yield) variations compared to the other variables.

Fig. 3 shows the Contour Plot of significant variables, derived from the final RSM model in achieving *Y1* (Propylene Product Yield). The values for non-tested variables were held at mean value, $\bar{x}$; Flow of Hearth Burner at10235 kg/hr, Flow of Integral Burner at 593 kg/hr, Flow of Dilution Steam at 40599 kg/hr, Flow of Naphtha at 61.43 t/hr, and COT at 807 °C. This figure was helpful to the Panel Operators as a guide in operating the studied plant with the targeted *Y1* (Propylene Product Yield) boundaries represented by the contour colors.

From the Contour Plot, *Y1* (Propylene Product Yield) could be best optimized by manipulating the lower *A* (Flow of HB) with a combination of higher *B* (Flow of IB), *C* (Flow of DS), *D* (Flow of Naphtha), or *E* (COT). However, it was also shown that operating the SRT VII at the higher *B* (Flow of IB) might also result in the lower Y1 (Propylene Product Yield) due to the Square and Two-way relations that existed in the model.

Besides, manipulating the higher *B* (Flow of IB) combined with higher *C* (Flow of DS), *D* (Flow of Naphtha), or *E* (COT) would also result in a higher *Y1* (Propylene Product Yield). However, the contour plot showed that there was a bigger challenge in operating *B* (Flow of IB) with the combination of *C* (Flow of DS) or *E* (COT) compared to *D* (Flow of Naphtha) in ensuring the *Y1* (Propylene Product Yield) maximization was at >12%.

Controlling *C* (Flow of DS) or *D* (Flow of Naphtha) towards *E* (COT) was also showing the same relation where the *Y1* (Propylene Product Yield) could be maximized with both combinations at the higher range. It could be achieved by following the operating envelope shown in the Contour Plot.

Table 2 displays the Multiple Response Prediction, derived from the final *Y1* (Propylene Product Yield) model with a 95% confidence level, while Fig. 4 illustrates the setting to achieve the maximum *Y1* (Propylene Product Yield) using Response Optimizer in Minitab.

Table 2. Multiple response prediction.

Response	Fit	SE Fit	Confidence	
			95% CI	**95% PI**
Y1 (Propylene Product Yield)	13.24	0.261	(12.725, 13.754)	(12.706, 13.773)

It indicated that the maximum *Y1* (Propylene Product Yield) in the studied plant was evaluated at 13.24% with the controlled operating parameters at 9476 kg/h of *A* (Flow of HB), 609 kg/h of *B* (Flow of IB), 40960 kg/h of *C* (Flow of DS), 63.50 t/hr of *D* (Flow of Naphtha), and 810 °C of *E* (COT). The high and low range settings in Fig. 4, represented 95% of the data confidence level to be referred by Operations personnel to maximize the *Y1* (Propylene Product Yield).

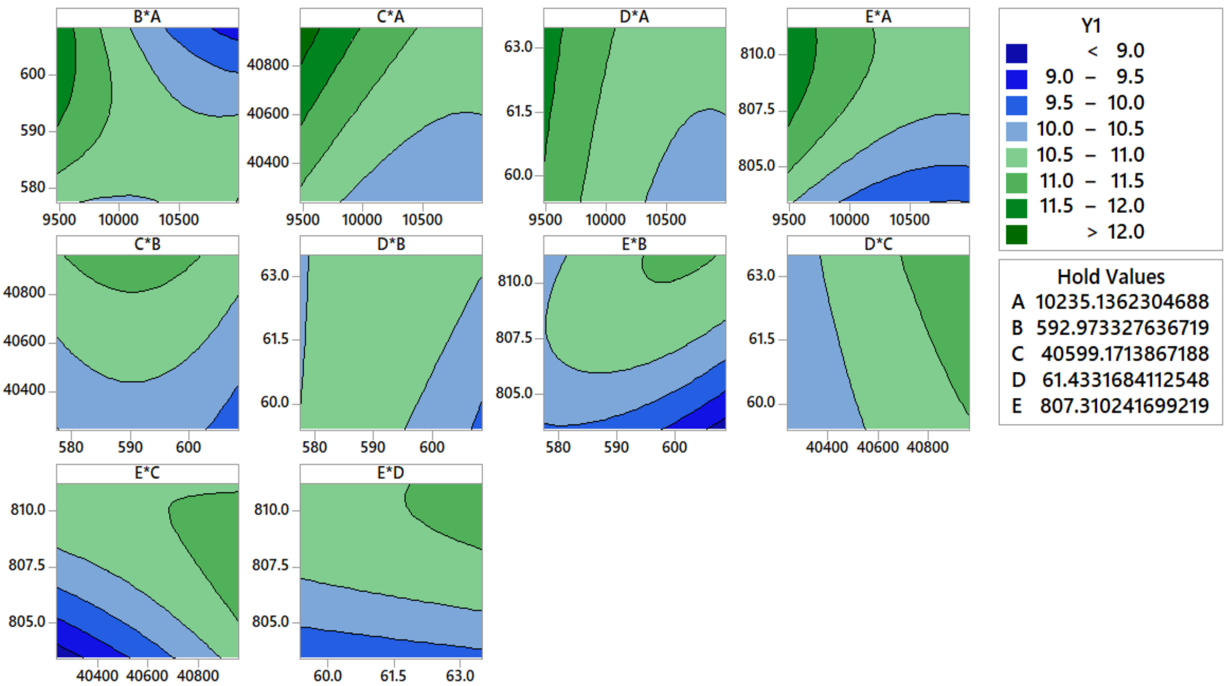

Fig. 3: Contour Plot of identified significant variables of *A* (Flow of HB), *B* (Flow of IB), *C* (Flow of DS), *D* (Flow of Naphtha), and *E* (COT) towards *Y1* (Propylene Product Yield).

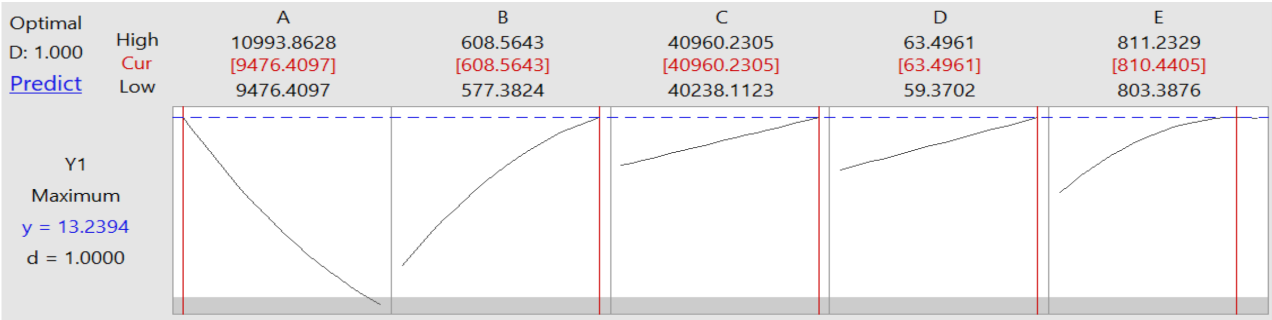

Fig. 4: Operating variables to establish the maximum *Y1* (Propylene Product Yield)

4. Conclusion

The model for Propylene Product Yield in the studied plant had been successfully developed using Minitab Software version 21. The COT had been identified as the most impactful variable in controlling the Propylene Product Yield by a factor of 49.4, compared to -7.76 for Flow of Naphtha, -4.3 for Flow of Integral Burner, 0.292 for Flow of Dilution Steam, and 0.05947 for Flow of Hearth Burner. The maximum Propylene Product Yield that could be achieved at the studied SRT VII was determined at 13.24% by following the process setting in the Response Optimizer.

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Nomenclature

A	Flow of Hearth Burner to SRT VII (kg/hr)
B	Flow of Integral Burner to SRT VII (kg/hr)
C	Flow of Dilution Steam mix with naphtha (kg/hr)
D	Flow of Naphtha to SRT VII (t/hr)
E	Coil outlet temperature from radiant (ºC)
Y1	Propylene Product Yield from SRT VII (%)

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