Publisher: Engineering College, Al Muthanna University
Open Access

DOI:10.52113/3/eng/mjet/2025-13-03-/15-26, *Vol. (13), Issue (3), (2025)*

Muthanna Journal of Engineering and Technology
MJET

Submitted 26 August 2025, Accepted 25 September 2025, Published online 27 September 2025

Converting heavy fuel oil into light fractions by using thermal cracking

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Abstract

Global developments have led to a steady increase in demand for light petroleum derivatives, which are more valuable than heavy derivatives. Therefore, it was necessary to seek unconventional methods for supplying them, such as the use of thermal cracking technology. In this research, the residual crude oil produced from atmospheric distillation unit was selected and a study was conducted to convert it into light petroleum derivatives using the thermal cracking process. The efficiency and accuracy of the technology used in this research were studied by selecting parameters such as API gravity and conversion ratio from used heavy fuel oil (API = 15.5) to light petroleum products. To achieve optimum operating conditions and the best desired process results, experiments were designed using response surface methodology (RSM). The process study was selected to be within the temperature range (350-450°C) and time (30-60 min), where the best possible specifications for the resulting light petroleum derivatives would be achieved, with an API density of (32.1), a conversion rate of (79.2%), a time of (45) min, and a temperature of (421)°C

Keywords: *AR, atmospheric residue ,API, Thermal Cracking, Residual crude oil, , Conversion rate*

1. Introduction

Crude oil or petroleum is a liquid composed of various types of hydrocarbons in addition to various mineral compounds such as sulfur, oxygen and hydrogen compounds [1].

The remaining heavy oil is one of the products of the distillation process of crude oil, and its quantity and density increase the heavier the refined crude oil [2].

The components of crude oil are separated using an atmospheric distillation tower, where crude oil enters at a temperature of approximately 350 degrees Celsius to separate its light components such as naphtha, kerosene, diesel and gas oil, and the non-evaporating part remains to exit from the bottom of the distillation tower and is called atmospheric distillation residue [3].

The specifications of crude oil used in atmospheric distillation processes directly affect the percentage of heavy residues produced, which constitutes a significant percentage, reaching half of the incoming crude oil [4].

The vacuum distillation unit is used to treat the residues of the atmospheric distillation unit, which works similarly to the previous unit and has almost the same temperatures, except that the first unit operates under the influence of vacuum pressure, through which the light derivatives in the residues of the atmospheric distillation unit, such as LVGO and HVGO, are separated, and the non-evaporating part passes from the bottom of the tower to form what is called the vacuum residue [2].

One of the most important problems facing the oil industry is how to treat heavy oil residues. Therefore, it was necessary to provide new mechanisms to avoid these problems and convert these heavy residues into lighter products with greater benefit, a higher price, and wider use [5].

One of the methods used to treat heavy oil waste is the use of thermal cracking technology. The asphalt content of oil residues is a very important factor in the thermal cracking process due to the increased coke formation rate when using heavier oil residues, i.e., with a higher molecular weight and high asphalt content [4].

The feed material specifications for thermal cracking units and the selection of optimal operating conditions, such as temperature, pressure, and time required to complete the reaction, directly affect the efficiency of the process and its outcomes [5].

The thermal cracking process is the use of high temperature to convert long hydrocarbon chains into shorter chains by breaking the (C-C) and (C-H) bonds in the molecules of organic compounds [7].

The temperature range at which the hydrocarbon chains (C-C) and (C-H) break occurs is above 350°C [8].

Generally , thermal processes are those processes of primary improvement of heavy crude oils and residues in which the decomposition, rearrangement or combination of hydrocarbons molecules is carried out through the application of heat and without the aid of a catalyst[9].

The process is spontaneous and the non-selective hydrocarbon disintegration occurs simultaneously with condensation and polymerization reactions, as a result molecules with a wide range of molecular weight and boiling points are produced. This means that the products of the reaction are not only lighter than the feed, but also a heavier residual with less hydrogen content than the feed is produced, which depending on its size and concentration, could produce some precipitate. Consequently, the conversion levels in thermal cracking are usually limited by the stability of the residue generated. In all these processes the targets are usually as follows: higher yields of light cracked oil, increasing total liquid yield, reduction of utility cost and reduction of construction cost .Thermal cracking are economically preferable because no hydrogen or catalyst is required, and operational pressure is low compared to hydrogen addition technologies using catalyst and high pressure hydrogen [10].

Not all types of thermal cracking processes require the application of high pressure, but all types require the application of high temperatures. Thermal cracking of heavy residues results in petroleum materials that are less dense, have a lower boiling point and a lower molecular weight due to the breaking of the bonds between the reduced crude residues molecules [11].

Thermal cracking processes break the hydrocarbon chains that bind molecules together, creating what are known as free radicals. Thermal cracking naturally results in the formation of shorter hydrocarbon chains, but in limited cases, these short chains bond together to form longer chains.

The thermal decomposition mechanism occurs through chain reactions of the resulting free radicals [12]. These chain reactions can be summarized into two main types:

Primary reactions: These are reactions that result in shorter hydrocarbon chains by breaking the bonds between hydrocarbon molecules.

$$CH_3CH_2\,CH_2CH_3 \rightarrow CH_4 \ + CH_3CH \ = \ CH_2$$

$$CH_3CH_2\,CH_2CH_3 \ \rightarrow \ CH_3\,CH_3 + \ CH_2 \ = \ CH_2$$

Secondary reactions: These are reactions that lead to the formation of longer hydrocarbon chains through reactions that occur between free radicals resulting from the process of breaking bonds between hydrocarbon molecules [1].

$$CH_2 \ = \ CH_2 + CH_2 \ = \ CH_2 \rightarrow \ CH_3CH_2CH = CH_3$$

OR

$$R.CH = CH_2 \ + R^1.CH = CH_2 \ \rightarrow cracked\,residum + coke + product$$

The use of high-pressure hydrogen when adopting thermal cracking technology has a significant effect in reducing the formation of coal and thus increasing the efficiency of the process, but its use requires high costs [13].

In thermal cracking processes, the temperature used is inversely proportional to the reaction time, but its continued increase may cause the formation of a product of low quality and undesirable specifications [14].

The scientists (T. Kaminski, M. Husein, 2018) used the residuals of atmospheric distillation units as a raw material for the thermal cracking unit and studied the effects of temperature and reaction time on the process. He showed that when choosing the temperatures used within the range (400,420) degrees Celsius and choosing the reaction time within the range (1, 2) hours, the best results were at a temperature of (420) degrees Celsius and a reaction time of (2) hours, in which a product with an API equal to (34) and a conversion rate equal to (85%) was obtained [15].

Thermal cracking of crude oil residues was carried out under different conditions by . (Alsobaai, 2013), where he used a high-pressure batch reactor using hydrogen and chose temperatures within the range (400-480) °C, pressure within the range (180-120) kPa, and reaction time within the range (100-40) minutes. He concluded that the best results were obtained at conditions (480 °C , 100 min. , 178 kPa) for temperature, reaction time, and pressure, respectively [16].

By(Y. Syamsuddin, et al., 2005), the thermal cracking of atmospheric distillation residues of crude oil was studied using a high-pressure batch reactor. The best results were achieved at 340°C and a reaction time of 3 hours, with a conversion rate of 51.43 wt.% [17].

(Krishna et al., 1988) used the residues of heavy crude oil in Aghajhari as a raw material in the thermal cracking unit. The process was studied under a temperature within the range (427 - 500 °C), a flow rate within the range (2.04 - 2.91 lit/hr.) and a pressure equal to (17 kg/cm²) [18].

In our current study, we investigated the effect of thermal cracking on the crude oil residue produced from the bottom of an atmospheric distillation tower. We aim to convert heavy residue (AR) into lighter products with multiple uses and higher value. The results of the process were studied and analyzed using the response surface methodology (RSM), starting with the experimental design and ending with the analysis of the results. This includes identifying the levels of influence of the operating conditions and determining the optimal conditions that lead to the best desired results.

MATERIALS AND METHODS

2.1 MATERIALS

The raw material used in the current study of the thermal cracking process represents the crude oil distillation residue produced from the bottom of the atmospheric distillation tower of the AL-Diwaniyah Refinery, the specifications of which are shown in Table 1

Table 1: The AR (atmospheric residue) properties

Characteristics	Test Method	Value
API	ASTM D- 1298 , IP 160	15.5
Flash point	ASTM D-93,IP 34	92 °C
Density @ 15 oC	-	0.970 gm/cm3
Viscosity @ 50 °C	ASTM D-445,IP 71	285 cst
Sulfur wt.%	ASTM D-4294	3.9
Pour point	ASTM D-97,IP 15	-3 °C
W & S (volume %)	ASTM D-4007	0.06

2.2 Method

A 0.5-liter semi-batch stainless steel autoclave reactor was selected for the thermal cracking of the heavy residue resulting from the atmospheric distillation of crude oil, shown in Figure 1.

Initially, the reactor was filled with 100 grams of AR, followed by shutting down the reactor and inserting it into the currently off electric furnace. Nitrogen gas was then introduced into the reactor, and the production valve was opened to release the oxygen contained in the reactor's internal chamber.

When the electric furnace surrounding the reactor is turned on, the temperature is gradually increased. The experiment time begins when the oil temperature inside the reactor reaches the desired value by Thermometer Inserted inside the reactor to measure the temperature inside it.The temperature of the autoclave was selected and recorded using by system control, manual controller. The magnetic stirrer play with (500 rpm), .

The electric furnace is turned off after the specified time for the experiment has elapsed, after which the reactor is removed and placed in a water bath to cool it to room temperature.

During the experiment, the AR thermal cracking product passes through a production tube and is then cooled using a water heat exchanger. The resulting liquid is collected and its weight is calculated. the gas and coke products were assumed negligible. The weight of the material remaining in the reactor after cooling is also calculated. The information obtained is used in the conversion percentage calculations, as shown in the equation below:

$$\text{CONVERSION \%}= \frac{Mass_{Feed}-Mass_{Residue}}{Mass_{Feed}} \qquad (1)\ [2]$$

As for the API gravity, it can be calculated for the resulting liquid using the equation below:

$$\text{API gravity} = (141.5 / SG) - 131.5 \qquad (2)\ [2]$$

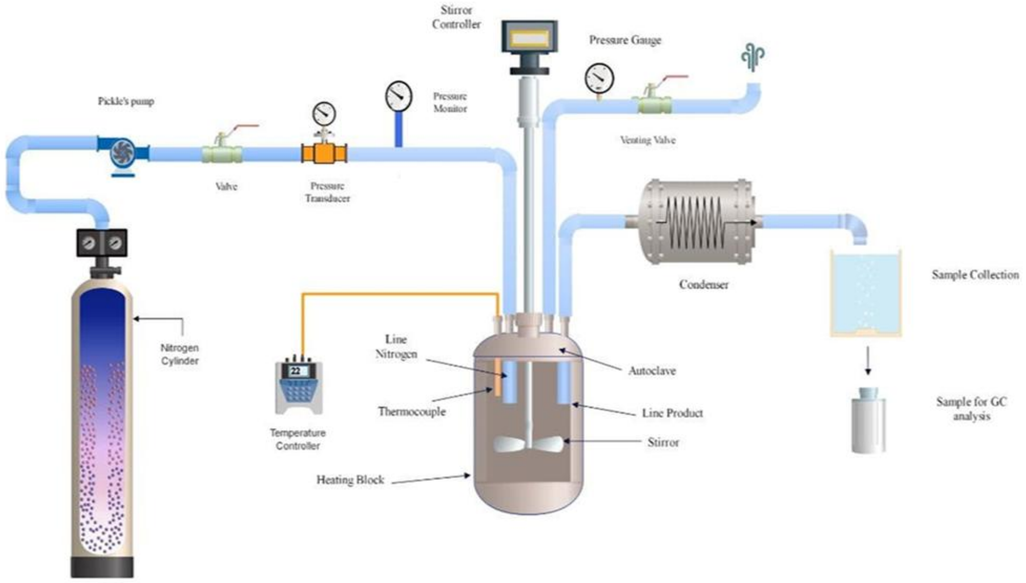

Fig. 1: Apparatus.

2.3 Design of experiments

The statistical program (Minitab-18) was used based on the statistical methodology (RSM) and (CCD((Central Composite Design)) to design the experiments and determine the influencing conditions and responses,to optimize processes by effectively approximating two-order response parameters, determining the relationship between input and responding variables, and accurately modeling the bending response . CCD design allows the identification of the factors most influencing the response, facilitating decision-making for product or process improvement. through the results of which we can determine the complex non-linear mathematical relationships related to the overlapping effect of the factors on the responses. As found in Equation 3, the second-degree polynomial shows the mathematical relationship between the operating conditions and the responses, and the calculation of the maximum value of the regression coefficient (R^2) and the analysis of variance of the results (Anova), which is considered a measure of the quality of the product of the thermal cracking process [19, 20].

$$Y=B_0+\sum B_i\ x_i+\sum B_{ii}\ x_i^2+\sum B_{ij}\ x_i\ x_j\ +\ \varepsilon \qquad (3)$$

Y is represent to the Responses the specifications of the results required from the process.

X_1,X_2 ,to X_q represents to the parameters that are adopted during the thermal cracking process.

(q) refers to the wholly number of parameters used in the thermal cracking process.

B_o it is a constant value that represents the intercept regression.

B_i it indicates the actual value of the extent of the linear regression effect.

B_{ii} it indicates the effective value of the quadratic regression.

B_{ij} refers to the effective value of the interaction regression.

ε denotes the value of the random error.

Based on the above studies, the operating conditions ranges of temperature, reaction time, used in the thermal cracking process were determined to investigate various behaviors and study their impact on the desired responses. (X1 , X2) refers to the value of the temperature in celsius and time in minute reaction, respectively, that used in the thermal cracking process, where the temperatures were chosen to be within the range (350-450) [6] [15] .degrees Celsius and the reaction time range within the range (30-60) minutes[21,22] .

Increasing pressure has a negative effect on the catalytic cracking process[23,24] .

so the pressure was taken to be constant at atmospheric pressure because pressure is an influential factor in the pyrolysis process, as its increase leads to an increase in the proportion of the resulting gas, so most of the processes of Pyrolysis at atmospheric pressure [9].

In addition, thermal cracking under high pressures is not beneficial due to increased deposition of asphalt and increased coke formation[6].

All experiments were performed under atmospheric pressure, and a mixer was used at 500 rpm. Using a CCD, the total number of experiments to be performed was determined and denoted by the symbol N.

$$N = q^2 + 2q + n \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (4)$$

n: Number of repetitions

The results of the responses, such as conversion ratio and API, were studied by conducting 11 runs, 4 factors, 5 central points and 4 pivot points, all of which were determined using the approved statistical program. Table 2 shows the ranges of operating limit levels used in the thermal cracking process. Table 3 shows the experimental design using Minitab 18 statistical program using RSM and CCD.

rotability $[\alpha=(2q)^{0.25}]$equals±1.414 .

Y refers to the desired responses as conversion ratio and API, calculated in equations (1 and 2).

Table 2: Process variables

Process parameters	central composite design range		
Coded levels	(-1) Low	(0) Middle	(+1) High
X1	350	400	450
X2	30	45	60

Table 3: Experiments design

Run	Blocks	Coded value		X1	X2
		X1	X2	Temp.	time
1	1	-1	-1	365	34
2	1	-1	+1	365	56
3	1	1	-1	435	34
4	1	1	+1	435	56
5	1	0	-α	400	30
6	1	0	+α	400	60
7	1	-α	0	350	45
8	1	+α	0	450	45
9	1	0	0	400	45
10	1	0	0	400	45
11	1	0	0	400	45

2. Results and Discussion

For the experiments carried out, Table 4 shows the calculated values for the expected results of the responses such as conversion ratio and API. Actual results were obtained that are consistent with the expected results calculated by the statistical program, which range between (3.4 - 77) and (30.4 – 39.1) as conversion percentage and API values, respectively.

Table 4: Results of the variables

Operational variables			Actual values		Predicted values	
Run	X1:	X2:	Conv.%	API gravity	Conv.%	API gravity
1	365	34	9.160	37.740	14.9724	37.5830
2	365	56	72.000	30.600	73.7225	31.2157
3	435	34	17.340	37.230	25.0650	36.8715
4	435	56	73.400	30.600	77.0351	31.0143
5	400	30	4.080	38.760	3.4122	39.1748
6	400	60	77.440	31.110	75.6737	30.4430
7	350	45	58.395	34.170	54.7169	33.8839
8	450	45	70.340	33.252	63.8568	33.2614
9	400	45	68.300	33.558	67.9850	33.8348
10	400	45	67.810	33.762	67.9850	33.8348
11	400	45	67.320	34.170	67.9850	33.8348

3.1. Mathematical correlation of the searched responses

The mathematical relationships in equations 5 and 6 were formulated based on the actual test values calculated and shown in Table 5, thus knowing the relationship that links the factors and variables of the operating conditions to the resulting responses such as the percentage conversion rate and the API value

Table 5: The regression coefficient

	Model Summary			
Conv.	S	R-sq	R-sq(adj)	R-sq(pred)
	6.70858	97.17%	94.34%	79.80%
API gravity	S	R-sq	R-sq(adj)	R-sq(pred)
	0.565169	97.99%	95.97%	86.39%

Table 6: ANOVA results for API and conversion gravity (underline numbers mean insignificant effect).

Source	DF	Adj SS	Adj MS	F-Value	P-Value
Model	5	77.7400	15.5480	48.68	0.000
Linear	2	75.8843	37.9422	118.79	0.000
Temperature	1	75.4822	75.4822	236.31	0.000
Time	1	0.4021	0.4021	1.26	0.313
Square	2	1.7907	0.8953	2.80	0.153
Temperature*Temperature	1	1.3408	1.3408	4.20	0.096
Time*Time	1	0.1006	0.1006	0.31	0.599
2-Way Interaction	1	0.0650	0.0650	0.20	0.671
Temperature*Time	1	0.0650	0.0650	0.20	0.671
Error	5	1.5971	0.3194		
Lack-of-Fit	3	1.4029	0.4676	4.82	0.177
Pure Error	2	0.1942	0.0971		
Total	10	79.3371			

Source	DF	Adj SS	Adj MS	F-Value	P-Value

Model	5	7723.86	1544.77	34.32	0.001
Linear	2	6278.72	3139.36	69.76	0.000
Temperature	1	6192.03	6192.03	137.59	0.000
Time	1	86.69	86.69	1.93	0.224
Square	2	1433.65	716.82	15.93	0.007
Temperature*Temperature	1	1433.62	1433.62	31.85	0.002
Time*Time	1	110.76	110.76	2.46	0.177
2-Way Interaction	1	11.49	11.49	0.26	0.635
Temperature*Time	1	11.49	11.49	0.26	0.635
Error	5	225.03	45.01		
Lack-of-Fit	3	224.55	74.85	311.74	0.003
Pure Error	2	0.48	0.24		
Total	10	7948.88			

Regression Equation in Uncoded Units

$$\text{Conversion} = -2458 + 11.18\,X1 + 5.54\,X2 - 0.01274\,X1*X1 - 0.0387\,X2*X2 - 0.00440\,X1*X2 \qquad (5)$$

$$\text{API} = 135.6 - 0.414\,X1 - 0.048\,X2 + 0.000390\,X1*X1 - 0.00117\,X2*X2 + 0.000331\,X1*X2 \qquad (6)$$

3.2. Analysis of variance (ANOVA)

Due to the difficulty and low accuracy of analyzing data from experimental results using traditional direct methods, it became necessary to find new methods such as analysis of variance (ANOVA), which is characterized by its high accuracy in calculating the quality of the results. In Table 6, the results of the analysis of variance for the conversion ratio and the API value are shown. The symbol p represents the degree of acceptability of the required responses as the conversion ratio percentage and the API value, where acceptability increases as its value decreases below (0.05). The value of the regression model is of great importance in evaluating the results of the conversion ratio and becomes unimportant for the API value if its value reaches (0.6). The F value obtained from the experimental results of this study were 34.32 and 48.68 for the conversion percentage and API value, respectively, from which we conclude the reasonableness of the results obtained. Acceptability is also enhanced by obtaining large regression coefficients and adequate fit of the calculated two-response results with adjusted R2 values. Equations 5 and 6 are arrived at after eliminating effects with P values greater than 0.05 (shown in the lower line of Table 6) and adopting the results of an ANOVA analysis. From these results, it can be concluded that the thermal cracking process is effective and can be used to successfully convert AR to light derivatives[25].

3.3 The effect of temperature and time on conversion

Figure 2 shows the extent to which the conversion rate is affected by temperature and reaction time during the thermal cracking process, where work was done at temperatures within the range (350-450) degrees Celsius and the time required to complete the process was within the range (30-60) minutes. It was observed that the conversion percentage values increased to the range (50-75) when the temperature rose to more than 390 degrees Celsius and the process took a time ranging within the domain (30-60) minutes. The excluded area is the one with the highest conversion percentage value, which is greater than 75%. While the conversion percentage decreased to the range (25-50)% when working at temperatures between 370 and 390 degrees Celsius when it takes a time equal to 60 minutes, noting the continued decrease in the conversion value in accordance with the decrease in the reaction temperature. Therefore, we conclude that increasing the process temperature will lead to an increase in the conversion rate, and the effect of the reaction time is at its highest levels when it is within the range (36-58) minutes, as shown in figure 3, which is consistent with the results presented in the research [16].

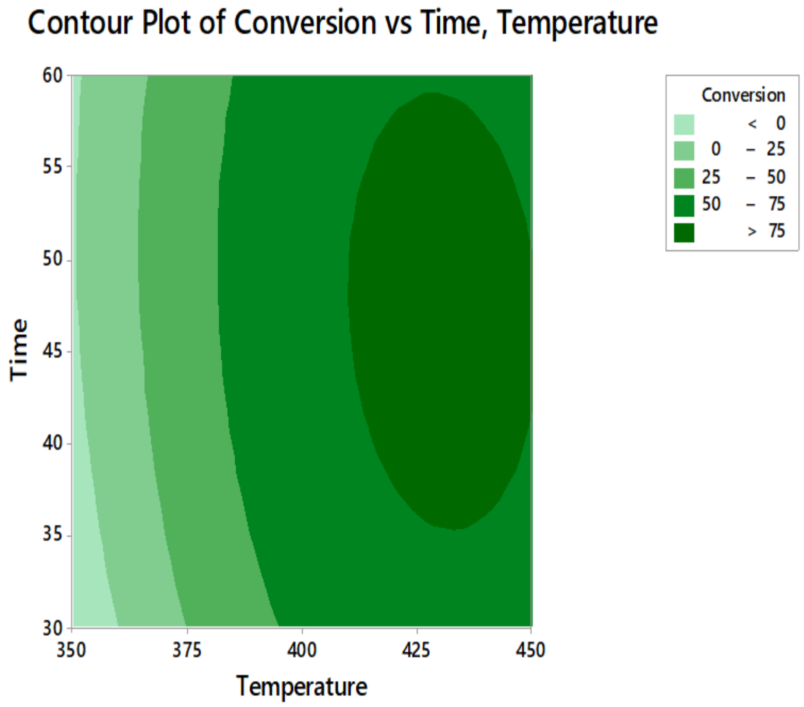

Fig. 2: counter plot temp. &time vs. conversion

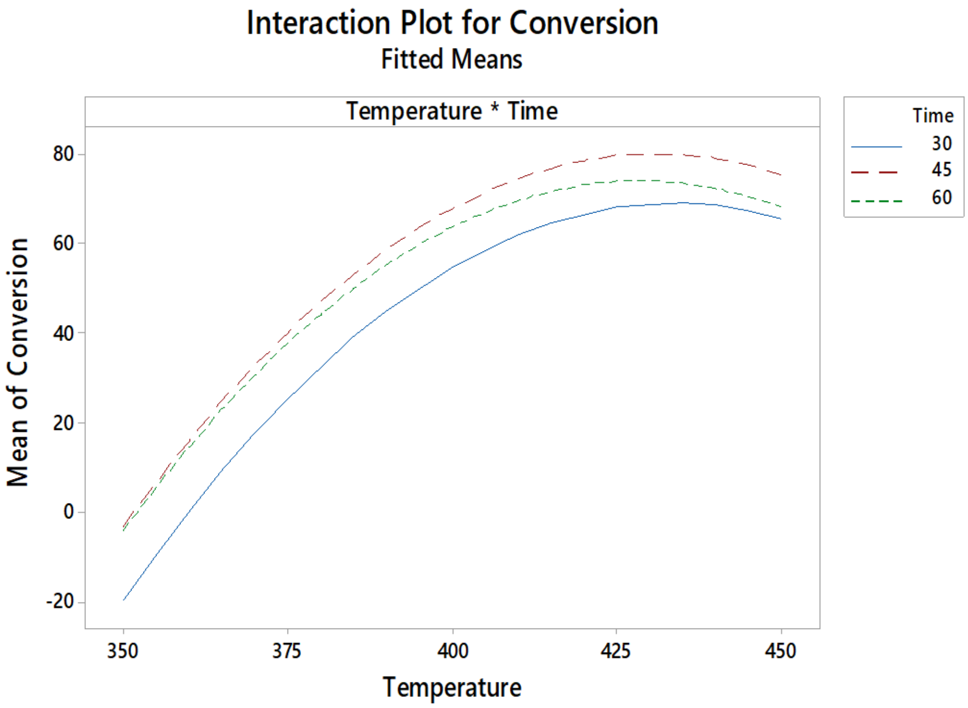

Fig. 3: interaction plot for conversion

3.4 The effect of temperature and time on API gravity

Figure 4 shows how the ApI value is affected by temperature changes within the range (350-450) degrees Celsius and when the reaction time takes within the range (30-60) minutes. It was observed that when working in the temperature range (350-360) degrees Celsius and also within the reaction time range (58-30) minutes, the highest value of API gravity was obtained. As the temperature and reaction time increased, the API gravity values decreased until the lowest API gravity value was recorded, approximately less than 32, within the temperature range of (420-450) degrees Celsius and when a reaction time of 60 minutes was consumed. It is clear from Figure 5 that increasing temperature and reaction time leads to a decrease in API gravity values, with difference that the effect of temperature is large, while the effect of reaction time is small. These conclusions are consistent with the results of the study presented previously [26].

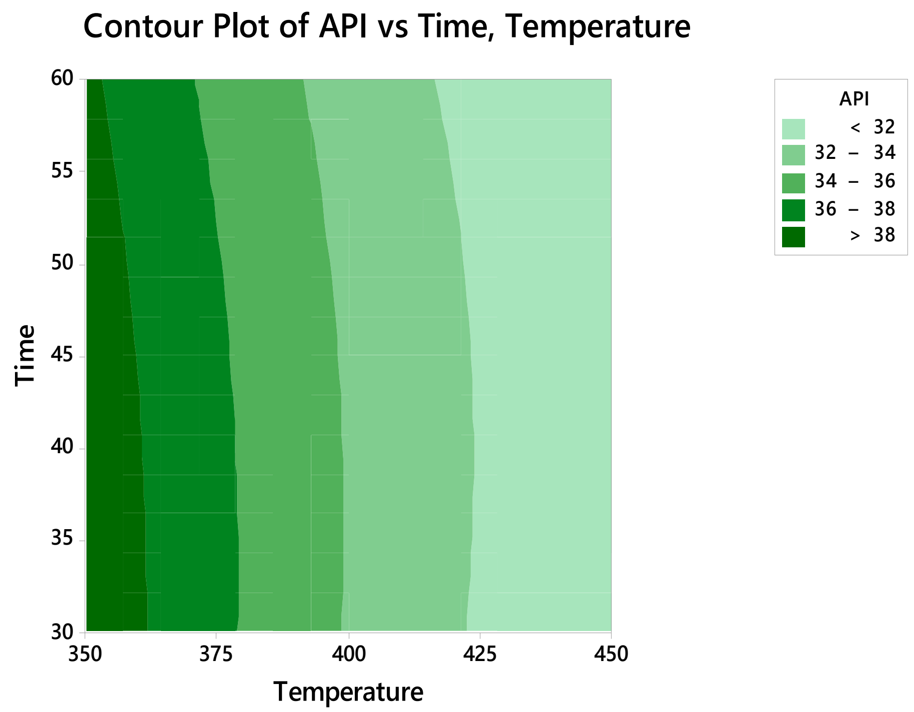

Fig. 4: The contour plot API vs. Time, Temperature

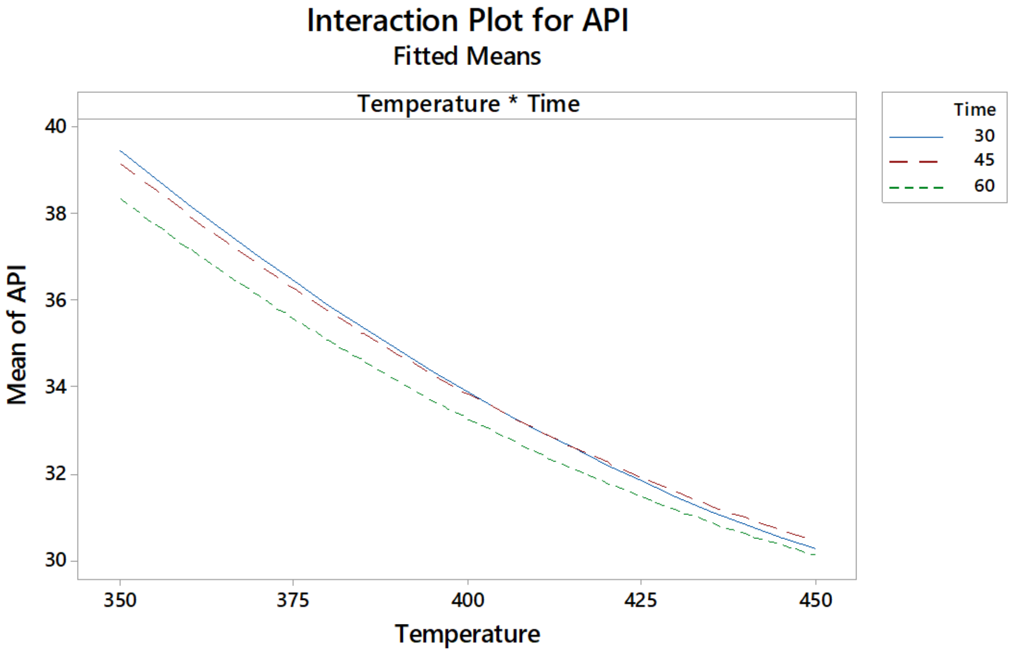

Fig. 5: interaction plot for API

3.5 The confirmation and optimization test

Table 7 shows the maximum values of the required responses that can be obtained when operating at the optimum limits of the operational variables, which were calculated and studied by statistical programming using Minitab-18. The best results were obtained at a temperature of 421°C and a reaction time of 45 minutes. Where the product was obtained with an API gravity value of 32.1 and the conversion percentage was 79.2%. This is evidence that the use of optimum values for operating conditions in the thermal cracking process has succeeded in achieving the best results for the required responses, which are represented by API gravity values and conversion percentage.

Table 7: The optimal values for maximum conversion and API

Response	Goal	Lower	Target	Upper	Weight	Importance
API	Maximum	30.6	38.760	38.760	1	1
Conversion	Maximum	4.08	77.440	77.440	1	1

Solution	Temperature	Time	API Fit	Conversion Fit
1	421	45	32.1	79.2

A practical experiment was conducted by applying the optimal conditions obtained from the statistical program, and the results were very close to the expected results, as we obtained the productivity of the liquid (65) and the API value (31.1). The necessary tests were carried out on the resulting liquid as shown in the table 8

Table 8 : Specifications of the product of the thermal cracking process at optimum operating conditions

Distillate%		Residue%	Lost%
70		12	18
Volume%	Temperature oC	-	-
10	167		
20	224		
30	240		
40	290		
50	311		
60	332		
65	335		

Table 9: The result of distillation test

Characteristics	Value
API	31.1
Density [gm./cm3] at (15 oC)	0.86
Viscosity at (50 °C)	3.4 cst
Sulfur wt.%	2.9
I.B.P.	82
E.B.P.	380

Atmospheric distillation of the resulting liquid was carried out in order to find out the percentages of the different fractions contained in the product, as shown in the table 9.

Through the process of atmospheric distillation of the liquid resulting from the thermal cracking process, light extracts such as gasoline, kerosene and gas oil were obtained in good proportions, and this indicates that the use of the thermal cracking process of AR is a successful process with good economic returns.

Fig. 6: The products of thermal and catalytic cracking process of RCR at the optimum operational conditions. (A) is RCR, (B) is thermal cracking product

4. Conclusions

The thermal cracking of crude oil refining residues can be studied for upgrading using thermal cracking technology through statistical analysis of process conditions and result values. The number of experiments was determined to be eleven experiments through designing experiments using the statistical program minitab-18 based on the response surface methodology RSM and CCD. Two operating conditions were adopted: temperature and time used in the thermal cracking process to obtain the required responses, which are represented by the conversion ratio and API gravity. The analysis of variance relied on the value of the regression coefficient R2 at a probability value less than 0.05 in order to benefit from it in determining the accuracy of the results obtained from the thermal cracking process experiments. There was a clear agreement between the experimental results and the expected results for ideal conditions. Data analysis revealed that the best results were obtained when the thermal cracking process was carried out at a temperature of 421°C and a reaction time of 45 minutes. The reactions achieved under these conditions resulted in a conversion rate of 79.2% and an API gravity of 32.1.

5. Acknowledgments

The authors would like to thank the staff and technicians of the Al-Muthanna University, College of Engineering, Department of Chemical Engineering, for their great support and assistance with technology.

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