



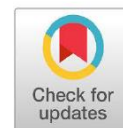
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Review article

Kinetic Overview of Catalytic Reforming Units (Fixed and Continuous Reforming)



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ABSTRACT

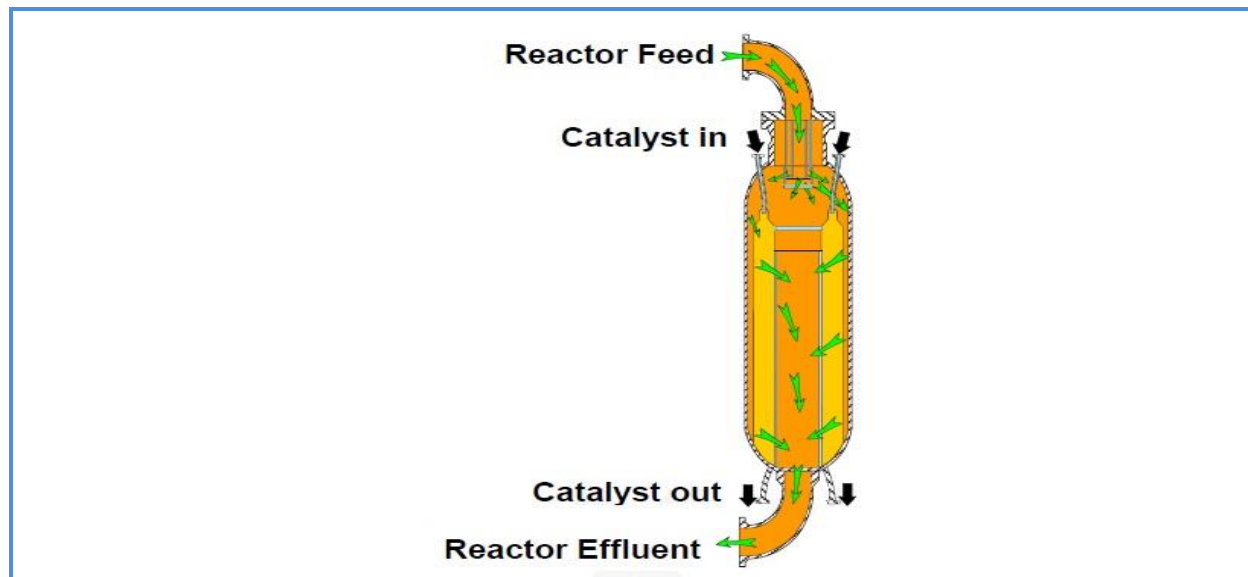
Naphtha catalytic reforming is one of the main processes of gasoline production with high octane number. Inactive catalysts and changing the products distribution is an important issue in this process. In this paper we discussed the kinetic overview of catalytic reforming units (fixed and continuous reforming). The catalyst activity model introduced in this research study has been used as a function of temperature and process time to detect the catalyst deactivation. The kinetic model and catalyst activity were estimated by using the genetic algorithm as well as overlapping the proposed model results with the experimental data. The results of the modeling showed that the amount of the aromatics during the reactor reduced the trend due to the decrease in the amount of paraffin and naphthen. After the process modeling, the effect of different factors such as time, reactor temperature changes, reactors operating pressure, and the ratio of hydrogen to hydrocarbon on the activity of catalysts and distribution of the products were investigated. The results revealed that, the aromatics rate decreased and coke formed rate on the catalyst surface increased by catalyst activity passing time. Also, increasing the input temperature and reducing the ratio of hydrogen to hydrocarbon enhanced the aromatics produced rate.

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Graphical Abstract



Introduction

Inactive catalysts and changing the products distribution is an important issue in this process. These units consist of three or four series fixed or movable catalytic bed reactors. Naphtha catalytic reforming is a process in which naphtha compounds with a low octane number are converted to high-octane gasoline compounds. In other words, mainly catalytic reactions of unbranched paraffin's in naphtha are converted to iso-paraffin and aromatics. Hydrogen and light hydrocarbons (C_1-C_4) are the process by-products.

Generally, the goals of catalytic reforming process are producing the high-octane gasoline, aromatics for petrochemical industries and the production of hydrogen for industrial uses such as hydrocracking and desulphurization [1-3]. Many studies have been conducted on the naphtha catalytic reforming process modelling [4-6]. In 1959, Smith [5] presented a set of model 3 (including naphthen, aromatic and paraffin) for the catalytic reforming process. The presented model was used in different studies [5-7]. Boomanon *et al.*, [8] improved the Smith model [9] in 1989 and determined the kinetic parameters by using experimental data. In 1991, Schroeder *et al.*, [10] studied the commercial catalysts inactivation of catalytic reforming process in a molecular reactor. In another study, Padmavasi *et al.*, [11] considered naphtha feed and catalytic reforming products as 26 sets that converts to each other by using six reactions. The catalyst deactivation has been considered due to the coke eclipse and an exponential descendant function [12]. The model presented by Padmavasi *et al.*, [12] has been used in other studies by Rahim pour [13] and Behin *et al.*, [14]. Rahim pour [15] has considered the catalyst activity as a

function of temperature and time. Also, Behin *et al.*, [16] studied the effect of substrate drop on modelling accuracy. Javares *et al.*, [17, 18] presented a set of models for the naphtha catalyst reforming. They considered the paraffin carbons range between 1 to 11 and aromatic and naphthen carbons range between 6 and 11 [19]. Other kinetic models were also presented by Lee *et al.*, [20], Weifing *et al.*, [21, 22]. Besides the kinetic studies in the field of naphtha catalytic reforming process, many efforts have been made to improve the catalysts of this process [25-18]. So, some attempts have been made to increase the activity and catalysts performance by adding one or two metals such as Re, Sn, and Ge to platinum. For the catalysts activity, the model presented in reference [23] has been used. This model was used by Rahim pour in 2006. Also, the Smith kinetic model [24] has been used for reactions. The results indicate that the catalyst activity as well as the products distribution of catalytic reforming process is a function of the input temperature and pressure and the ratio of hydrogen to hydrocarbons.

Table 1. Studies conducted for catalytic conversion units [22-24]

Further details	Year	Studies done	Scientist	Row
	1915	Introduction the molecular failure processes, increasing the quality and quantity of various oil products	Vladimir Shukhov (Russia)	1
	1916	Process modification and gasoline production using crude oil distillation	William Merriam Burton	2
Construction of its first industrial unit in 1936 in the United States by Houdri	1923	Introduction the catalytic cracking method	Ozhen Houdri (France)	3
The problem with using platinum is that it's expensive and rare	1940	The invention of first process of naphtha catalytic conversion by using of platinum-containing catalyst	Vladimir Hansel (Russia)	4
Led to significant economic advances in the hydrocarbons catalytic conversion process.	1947	Process modification using platinum fine metal particles on the surface of acidic base of alumina to perform chemical reactions in order to optimize and improve the gasoline octane number	Vladimir Hansel (Russia)	5
Construction of the first naphtha catalytic conversion platforming in 1949 at the old Dutch company Michigan refinery	1949	Production of high-octane gasoline from naphtha industrial scale by UOP under the name Platforming	Vladimir Hansel (Russia)	6
	1959	Presentation the first kinetic model of naphtha catalytic conversion reactions	Smith	7
	1960	The presentation of another model, containing crude 20 elements and hydrocarbons 8 to 10 carbon in the form of 53 reactions	Crown (United State)	8
	1972	Using the Langmuir kinetic model for the catalytic conversion process	Kemak	9

	1983	Development of a model consisting of 23 fictitious elements and hydrocarbon cutting 5 to 10 carbons	Marin	10
	1997	Optimization the Marin model by taking 35 fictitious elements and 36 reactions	Froment	11
	1998	The mathematical model for gasoline catalytic conversion of with the commercial name KR-104A and the use of hydrocarbon cutting of 60-150 ° C from petroleum crude materials	Ostrovsky	12
	2000	Presentation a model consisting of 12 elements for calculating the gasoline catalytic conversion in reactor radial and axial substrates	Sharikov	13
To simulate refinery processes	2000	Presentation the software Petro-SIM	Company KBC	14
using statistical methods to create mathematical models based on the principles of chemistry physics, taking into account the product and genus quality and type of catalyst and the feed composition percentages	2001	Using software and calculations of computer systems and taking into account the physical properties of naphtha feed cuts in the catalytic conversion process for testing and selecting a suitable platinum catalyst and optimizing its conditions and preparing it for industrial application and predicting operating conditions associated with the cycle catalyst recovery and increase unit efficiency based on process technology and composition of refinery feed hydrocarbon percentage	Kvartsov	15
Presenting two new versions of the benzene separation method from the reaction mixture, the first edition of which includes the gasoline reformat production containing 0.01% by weight of benzene, with a 12% reduction in the efficiency of catalytic conversion process, and the second edition of gasoline reformat containing 0.47% benzene and an increase of 2.9% of process efficiency	2006	Optimization the naphtha catalytic conversion process with the aim of producing gasoline reformat with benzene content of less than 1% weight by mathematical modelling	Noshkina	16
Applied techniques, robust databases, complete set of modules, flexible software system tools, and maintaining the specifications of the proposed kinetic model are all of this research strengths.	2006	Presentation a model consists of 18 new kinetic equation of naphtha catalytic conversion reaction process by the software SPAN PLUS as a functional module to simulate the process and development and expanding this model and its	Hu VI Feng	17

		optimization		
The decision variables were selected for four reaction temperature and pressure reactors and a hydrogen to naphtha ratio	2007	Using a multi-purpose strategy to provide a more complete network-based process model containing 20 chemical reaction kinetics equations for optimizing a continuous catalytic naphtha unit with the goal of achieving a high aromatic product for higher octane	Hu VI Feng	18
Their proposed model includes calculation of catalyst movement, catalyst activity change due to formation of coke along the length of reactor bed and catalyst activity relationship with their circulation ratio	2010	Mathematic modelling of gasoline producing catalytic conversion unit with continuous regeneration process	Gyngazova	19
Investigation the corrosion in the above units and providing a proper model by using the neural network	2019	Use of Data Mining in the Corrosion Classification of Pipelines in Catalytic Reforming Units (CRU)	Soroush Zarrinabadi, et al	20

Experimental

In the naphtha catalytic reforming process, the steam was mixed with hydrogen in a proportion of 3-5 mol of hydrogen with a hydrocarbon molecule at 450-530 °C. Dehydrogenation is the main reaction of this process, which is an endothermic reaction. In this reaction, the reactor temperature dropped rapidly, which reduced the reaction rate. The catalytic reforming process was carried out in a set of fixed bed reactors between which the furnace was placed. This mode of operation makes the process temperature almost constant and yields high [25]. The catalyst weight of total reactors was 1050 kg. The catalyst distribution percentage in the four reactors from the first reactor to the end was 10, 15, 25 and 50% for four reactors and 20, 30 and 50% for the three reactors, respectively. Before each reactor, the furnace was placed to increase the feed temperature to the reactor. The flow type inside the reactors was radial, and the feed flow rate to the reactor was 25/4 kmol/h. The catalytic bed reactor pressure was 4.5 barg, and the molecular mass for the input current was 170 Kg/Kgmole.

Table 2. Catalytic reforming unit data

Rate of reaction	Equation of Reaction	Name
$-r_1 = a(k_1 P_N - K_1 P_A P_{H_2}^3)$	$N \leftrightarrow A + 3H_2$	Di-hydro-generation
$-r_2 = a(k_2 P_p - K_2 P_N P_{H_2})$	$P \leftrightarrow N + H_2$	Di hydro-cyclists
$-r_3 = a \frac{k_3 P_N}{P_{tot}}$	$\frac{n}{N + 3 H_2} \rightarrow \text{Light Paraffin's}$	Hydrocracking of olefins
$-r_4 = a \frac{k_4 P_p}{P_{tot}}$	$\frac{n-3}{P + 3 H_2} \rightarrow \text{Light Paraffin's}$	Hydrocracking of paraffin's

Process modelling

Kinetic model: According to the data type that is a set of paraffin, naphthen, and aromatic, only the Smith kinetic model can be used. This model consists of thermal and endothermic and exothermic reactions as following.

Dehydrogenation: Dehydrogenation was one of the reactions that occur in the naphtha catalytic reforming process. This reaction is carried out in the metal part of the catalyst and is the fastest catalytic reforming reaction. This reaction is endothermic that may increase the product octane number. At high temperature and low pressure, the dehydrogenation reaction is well done.

De-hydro-cyclises: The conversion of paraffin's into olefins is one of the reactions that are carried out in the naphtha catalytic reforming process. This reaction is endothermic and relatively slow, as well as the dehydrogenation reaction was done at high temperature and low pressure.

Naphthen and paraffin's cracking: Naphthen and paraffin's cracking reactions were carried out in the catalytic reforming process. The products of these reactions were light hydrocarbons and considered to be undesirable. The cracking reactions were exothermic and should be at high temperatures and high pressures. The value of n was calculated using the following equation.

$$n = \frac{Mw - 2x_p + 6x_A}{14} \quad (1)$$

Also, the reaction and equilibrium (K_i and k_i) are ariance functions of temperature and are defined as:

$$k_i = k_{i,0} e^{-\frac{E_i}{RT}}, i=1, 2, 3, 4 \quad (2)$$

$$K_i = e^{\left(A_i - \frac{B_i}{T}\right)}, i=1, 2 \quad (3)$$

In relations (2) and (3), the values of A_i and k_i , 0 were constant, and used for the mean values of reference [9]. The values of E_i and B_i are parameters that calculated using the optimization for the proposed models.

Reactor model

The physical model considered for catalytic reforming reactor was radial and its schematic is shown in Figure 1.

$$\frac{dF_A}{dm} = r_1 \quad (4)$$

$$\frac{dF_N}{dm} = -r_1 + r_2 - r_3 \quad (5)$$

$$\frac{dF_P}{dm} = -r_2 - r_4 \quad (6)$$

$$\frac{dF_{H_2}}{dm} = 3r_1 + r_2 - \left(\frac{n}{3}\right)r_3 - \left(\frac{n-3}{3}\right)r_4 \quad (7)$$

$$\frac{dT}{dm} = -\frac{1}{F_{tot} C_{p,ave}} \sum r_i \Delta H_{r,i} \quad (8)$$

Table 3. Smith model reactions [4] and their rates [5]

(K) Reactors ΔT				H ₂ /HC	T _i (K)	(%Mole) Mole fraction			Raw
First	Second	Third	Forth			Aromatic	Naphthen	Paraffin	
-5	-11.5	-42	-75	5	520	12.8	29.47	58.4	1
-4.5	-11.0	-40.0	-75.0	4.9	520	12.3	28.75	59.42	2
-4.7	-10.0	-38.5	-77.0	4.85	521	12.8	27.70	59.41	3
-4.8	-10.0	-35.0	-69.5	4.5	520	12.75	27.40	60.44	4
-	-	-	-	-	-	56.74	3.50	40.55	5
-4.0	-10.5	-27.0	-61.5	4.4	521	11.48	32.47	56.74	6
-4.5	-11.0	-26.0	-59.0	4.4	528	11.55	32.27	58.19	7

To solve the ODE equations system, fourth rank Runge–Kutta numerical solution method was employed. C_p and ΔH_r are the mean specific heat capacity and the reaction heat, respectively. In this research study, temperature specificity of the specific heat capacity for hydrogen, paraffin, oil, and aromatics were extracted from reference, and the mean values of C_6 to C_9 for each carbon-containing group was used. Also, the reaction heat was obtained from the average reaction heat reported by Padmavasi for the carbon C_6 to C_9 .

Kinetic activity model

The function for the catalysts activity of this process is as follows [26].

$$-\frac{da}{dt} = k_d \exp \left[-\frac{E_d}{R} \left(\frac{1}{T} - \frac{1}{T_R} \right) \right] a^d \quad (9)$$

Where k_d , d , E_d , T_R are parameters that are obtained using empirical data.

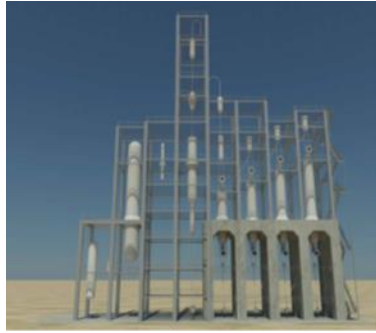


Figure 1. Catalytic reforming reactor model

Determining the parameters of the proposed model

The parameters of the proposed model should be determined in such a way that the model results with the empirical data are coherent. The optimization method for genetic algorithm was used to calculate the parameters. In this algorithm, the objective is to minimize the objective function that is expressed as follows.

$$OF = \sum_{i=A,P,N} \left[\frac{F_{i,out,calc} - F_{i,out,plant}}{F_{i,out,plant}} \right]^2 + \sum_{i=1}^4 \left[\frac{T_{i,out,calc} - T_{i,out,plant}}{T_{i,out,plant}} \right]^2 \quad (10)$$

By applying the objective function on the data in Table 2, the proposed model parameters (kinetic parameters and catalyst activity) were calculated. These parameters are presented in Table 4.

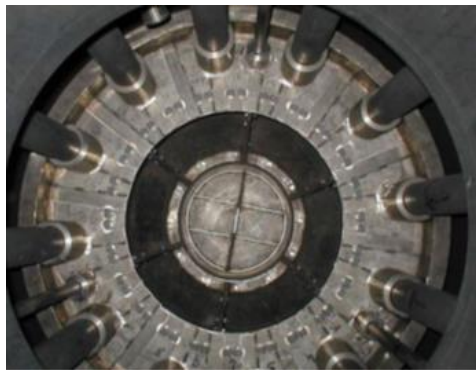
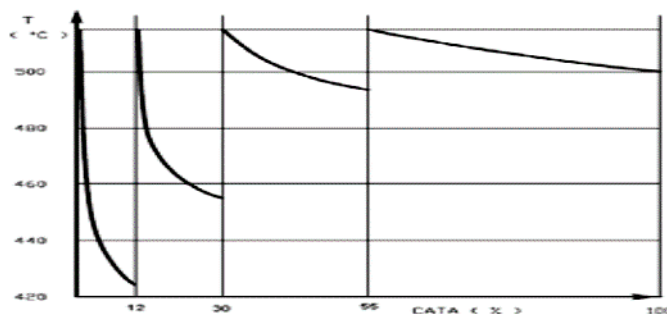
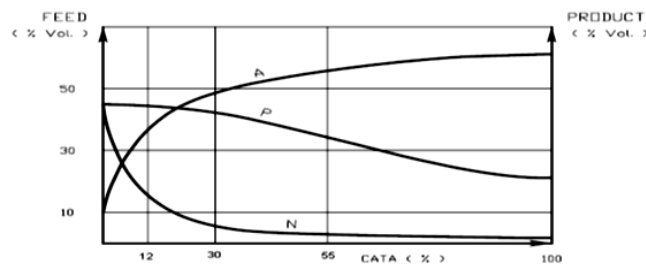


Figure 2. Internal of reactor in catalytic reforming units

Table 4. Presented model parameter

Amount	Parameter
21000	B_1
33000	B_2
34000	E_1/R
35000	E_2/R
26000	E_3/R
4739	E_4/R
0.000607	K_d
6.20	D
173140	E_d
750.55	T_R

The components and temperature concentration profile in terms of mass catalyst at different times can be calculated using the parameters obtained for the proposed models. (Figures 3 and 4) reveals the temperature and component concentration profiles in comparison with the experimental data. In three primary reactors, the increase in aromatic concentrations is due to the reduction of oil. In other words, the dehydrogenation reaction in these reactors is more than other reactions. In the final reactor, where the petroleum amount is low, formation of the aromatic production is due to the presence of paraffin's. In the last reactors, paraffin is first converted to naphthen, and then naphthen to aromatics. The effect of this speech can also be obtained in the temperature profile in the primary reactors, due to the endothermic dehydrogenation.

**Figure 3.** Temperature profile based on catalyst percentage**Figure 4.** Feed unit profile (volume percentage) based on catalyst amount

In the last reactor, the temperature drop has reached its minimum value, indicating that more exothermic reactions are performed along with the dehydrogenation reaction.

Investigating the effect of different factors on catalyst activity and products distribution

In this work, the effect of various factors including, time, reactor input temperature, hydrogen to input hydrocarbon ratio, and reactor pressure on catalyst activity and products distribution of catalytic reforming process were investigated.

According to the model presented for catalyst activity, time plays a significant role in the catalysts deactivation. To evaluate this factor, the time variations on the change in product distribution and catalyst activity are investigated for specific input conditions ($A=14$, $N=27$, $P=4.5$ barg, $H_2/HC=3.5$, $T=520$). Over time, the catalyst activity rate and aromatic production decreased slightly.

The hydrogen to hydrocarbons ratio may change the products distribution, which was evaluated in this study.

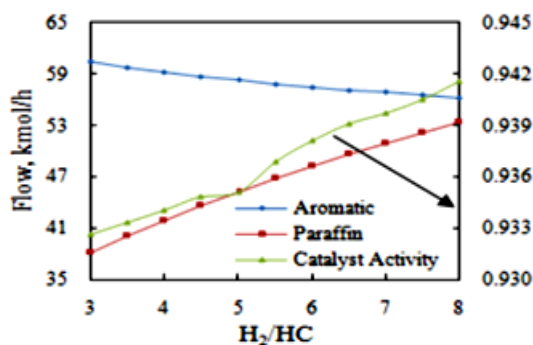


Figure 5. Effects of hydrogen to hydrocarbon changes on catalyst activity and products distribution

As demonstrated in Figure 5, the increase in temperature reduced the catalyst activity and also increased the aromatics amount in the product, also the paraffin amount was significantly decreased with the increase in temperature. Also, the effect of variations in the input temperature on each reactor was investigated separately (by constant input temperature for other reactors). The effect of the variation of the input temperature on the activity of the catalysts and products distribution is lower than the other inputs. The influence of changes in the fourth reactor was greater than the third reactor and the effect of the change in the third reactor temperature was greater than the second reactor. In the processes that carried out in the gas phase, pressure was one of the factors that affected the products distribution. The changes in the pressure of all reactors for feed created the optimal conditions. It is known that in the pressure between 4-5 barg, the highest amount of aromatics, paraffin and also the highest amount for catalysts activity is obtained.

Optimization the input reactors temperature and pressure

The objective function was defined in a way that the aromatics amount was produced and the catalysts activity was as high as possible.

$$OF = -a \times A \quad (11)$$

Using the kinetic model and the objective function of Equation 11, the values of input temperatures and reactor pressure were calculated at different times (Table 5). Table 5. also reveals the comparison between the catalyst activity and aromatics and paraffin production using the optimal values at any time, as well as the average values at all times. As seen in Table 5, there is a very little error indicating that operating conditions for reactors can be obtained based on the average optimal values.

Table 5. Optimum input temperatures and optimal pressures at different times these mean values

Pressure (kPa)				Temperature (K)				Time (day)
Reactor 1	Reactor 4	Reactor 3	Reactor 2	Reactor 4	Reactor 3	Reactor 2	Reactor 1	
4.3	4	4.82	4.25	520.85	520.75	520.88	520.78	0
4.62	4.83	4.32	4.25	520.96	520.00	520.63	520.68	20
4.14	4.08	4.82	4.75	521.85	521.81	521.63	521.18	30
4.64	4.45	4.07	4.75	521.97	521.95	521.88	521.43	40
4.85	4.39	4.32	4.75	789.99	522.88	522.63	522.68	50
4.64	4.83	4.53	4.25	522.85	522.75	522.23	522.68	60
4.85	4.83	4.07	4.25	789.97	523.88	523.63	523.06	70
4.85	4.89	4.03	4.38	789.97	523.95	523.71	523.66	80
4.14	4.83	4.82	4.25	524.85	524.75	524.22	524.18	90
4.12	4.88	4.75	4.32	522.92	522.86	522.16	522.15	Average

Table 6. The accuracy of using the mean values of input temperatures and reactor pressure instead of using different quantities at any time

Error(%)			Optimizer Data			Optimizer Data			Time (day)
Aromatic	Paraffin	Activity	Aromatic	Paraffin	Activity	Aromatic	Paraffin	Activity	
700.0	0.523	0.000	2.000	41.286	55.258	41.588	59.253	1.000	0
0.000	0.888	0.005	0.987	39.893	59.987	57.170	42.242	0.985	20
1.874	0.012	0.12	0.985	40.442	59.789	57.082	40.981	0.972	60
1.547	0.014	0.016	0.963	40.943	59.654	40.516	61.006	0.852	70
0.11	0.258	0.005	0.965	41.403	59.456	0.456	41.674	60.948	80
0.014	0.888	0.012	0.985	41.828	59.333	0.954	41.493	60.873	90
0.14	0.852	0.008	0.924	42.224	60.444	0.929	42.492	60.824	100
0.22	0.976	0.011	0.963	44.985	59.888	0.967	42.875	60.769	110
0.028	0.963	0.015	0.956	43.888	58.777	0.901	42.585	60.699	120

Investigation the effect of changes in the naphtha catalytic conversion unit capacity on gasoline octane number and the coke formed amount on the catalyst

The purpose of evaluating the effects of unit capacity on two important parameters of the catalytic conversion process is to determine the importance of process condition settings and the operating

variables for each capacity. The main result of this study is that by increasing the capacity of the catalytic conversion unit, the gasoline product octane number and coke formed content amount on the unit catalyst level increased. Of course, in this unit capacity variation, the coke formed percentage is within the permitted range. In the case of steady state of 22 ton/h, the circulating gas flow with a purity of 90% hydrogen and flow of 800 kg/hr catalysts and 520 °C of reactor input temperature, the range of heavy naphtha feed capacity of the continuous catalytic conversion unit for the conditions process is appropriate.

Investigation the effect of reactor input temperature of catalytic conversion units on octane number and the coke formed amount on the catalyst

One of the major results of this study is that the maximum allowable reactor input temperature was 530 °C. With increasing the reactor temperature, the gasoline octane number enhanced; however, the amount of coke increased as the reactor temperature enhanced. The slope of this coke formation at 520 °C increased dramatically, which is a problem that may enhance the temperature. As a result, the catalytic conversion reactors optimum input temperature in the fixed capacity of heavy naphtha feed unit of catalytic conversion and a constant mass flow of 22 ton/h recycling hydrogen gas with a purity of 90% and a constant catalyst flow rate of 800 kg/h at the range of 521-521 °C was obtained.

Conclusion

In this research study, the Smith's kinetic model was used for modelling, and the catalysts activity was assessed as a function of temperature and time. The kinetic parameters and catalyst activity were calculated using the single refining data. Then, the effect of various factors on the activity of the catalysts and products distribution was investigated. Over time, the catalysts were deactivated that resulted in decreasing the aromatic amount production. Also, increasing the input temperature to the reactors and reducing the hydrogen to hydrocarbons ratio reduced the catalyst activity and increased the aromatics produced amount. By changing the reactors pressure, it was found that the highest levels of aromatherapy would be produced at medium pressure.

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