Mathematical Statistician and Engineering Applications
ISSN: 2094-0343
2326-9865

Performance Assessment of Effectiveness Factor of Catalyst by Adjusting Reformers Operating Temperature in Basra Refinery Plant

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Article Info
Page Number: 388-399
Publication Issue:
Vol 71 No. 3s2 (2022)

Abstract:
Prediction of effectiveness factors for catalyst particles is one of the complicated steps in chemical reaction engineering. It combines chemical reaction kinetics with transport phenomena. In this study, the proposed model incorporates three semi-regenerative catalytic reformers with furnaces developed for the Basrah Refinery B.R.P in Basrah City, one of Iraq's most important production plants. The current study used mathematical modeling based on the Smith model to estimate the product molar and temperature distributions related to reactor bed radius. In each reformer, simultaneous catalytic reforming processes occur, where the reactions are of first-order for the reactants. The feedstock and product flows are considered ideal gases. Only variables in the radial direction are considered, with no respect for axial dispersions. An approximate technique for evaluating catalyst effectiveness factors is prediction. Based on collected and analyzed the properties of light naphtha production and carefully selected reaction temperature for improving the catalyst effectiveness factor, then R.O.N. instead of the conventional methods which depend on catalyst regeneration processes. The results show that the accuracy of the predicted model relative to Ke-min results is about (92%). Data collection has been carried out for more than one year under actual operating conditions and then analyzed to predict catalyst effectiveness as a function of time. The results showed the existing profile of catalyst effectiveness factor as a function of actual operating conditions and time has changed from (0.927-0.546) when the operation temperature changed between (470 to 485). consequently, the values of R.O.N. of light naphtha have improved from 80 to 90 without regeneration processes.

Article History
Article Received: 28 April 2022
Revised: 15 May 2022
Accepted: 20 June 2022
Publication: 21 July 2022

INTRODUCTION

Catalytic reformation is a method that entails converting heavy naphtha's low-octane hydrocarbons into more useable high-octane gasoline components while retaining the boiling point range. In the (C_5 –C_{12}) range, naphtha and reformate are complicated combinations of paraffin, naphthenes, and aromatics. Olefins are also found in naphtha as produced by catalytic or thermal cracking [1]. This method utilizes naphtha or cracking oil as a feedstock to create high aromatics and high octane value liquid products. Simultaneously, as a byproduct, it generates liquefied petroleum gas (L.P.G.) and hydrogen (H_2) [3]. The catalyst is regenerated and coke removed to keep the catalyst effectiveness at an acceptable range by combustion with air. In any case, regeneration or replacement of inactive catalysts is required from period to period [4]. Catalyst deactivation can occur by various factors, including working period, coke deposition on the catalyst surface, and

deactivation of catalysts by a sudden plant closure. Based on [5], the frequency of catalyst renewal determines categorization. Manufactured catalysts in contemporary catalytic reforming units consist of Gama Alumina support, certain metals like Platinum, Rhenium, Germanium, and Iridium (less than 1% wt), and additives like chlorine to promote the isomerization reaction [3]. Six responses occur throughout the catalytic reforming process, as mentioned in [6,24-28]: dehydrogenation, isomerization, dehydrocyclization, hydrocracking, hydrodealkylation, and coke formation. The first three reactions are essential and favorable because they raise the octane number, but the last three are unfavorable because they lower the number.

Several scholars have developed several kinetic models to depict catalytic reforming. Smith [4] offered a satisfactory kinetic analysis in 1959 focused on perfecting the complicated combination of naphtha. Individual components describe the naphthene, aromatic, and paraffin groups. In 2005, Ke-min et al. [7,19-23] presented a theoretical model that explains catalytic reformation using four packed bed reactors in sequence with radially flowing under adiabatic circumstances. Butheana et al. [8] created a naphtha catalytic reformation unit forecasting model employing Artificial Neural Networks (ANN) in 2014. In the present work, a mathematical model is prepared for predicting and showing the profile of the effectiveness factor of catalyst as a function of time and described the technical procedure which has been used in B.R.P. to improve the catalyst effectiveness factor then R.O.N. by controlling on operating temperature without using the regeneration processes and this procedure due to limitations such as equipment service life and catalyst replacement requirements.

DESCRIPTION OF THE BASRAH REFINERY PLANT

The Basrah refinery is one of the petroleum facilities in the south of Iraq. This facility is composed of several sub-units., including reformation units. One of the essential plants in this project is the reformation plant. It converts low-octane heavy naphtha (around 40-65) to high-octane light naphtha (approximately 80-96). The plant generates about 10000 brails of light naphtha per day. The reformation process involves three fixed bed catalytic reactors. The catalytic reforming units are working under relatively high operating conditions, with temperatures ranging from (460-513)°c, pressures ranging from (1.5-2.5 MPa), and a molar ratio of hydrogen to hydrocarbons of around 5.5. Installation of Platinum-alumina catalysts inside the reactors, and the feedstock density ranges from (728.8 to 743.7 kg/m3). They are using Pt/Al_2O_3 (RG682) as a catalyst in all of them. The catalyst capacity in the first, second, and third reactors is approximately 13.8, 20.7, and 34.5 m^3, respectively, while the other characteristics have shown in table (1).

Nevertheless, the reformation feed must be preheated with hydrogen during the first portion of the furnace to heat the input combination to the temperature required (around 508-513 °C) before entering the first catalytic reactor. Most favorable processes, such as dehydrogenation and isomerization, occur inside the reactor. Following heating up, in the furnace's second half, the product from the first reactor entered a second reactor, as well as the product from the second reactor entered the third reactor. In any case, every reactor is followed by a furnace since the total reforming reactions in the reactors are endothermic.

Table (1) shows the overall design dimensions of reformers utilized at B.R.P.

Reactor No.	Out Reactor Diameter (mm)	Catalyst Bed Diameter (mm)	Volume of Bed Catalyst (m^3)	Height of Bed Catalyst (mm)	Weight of Catalyst (Tons)
I	466	1640	13.8	3397	8
II	466	1946	20.7	3939	12
III	466	2446	34.5	4595	20

Reactor No.	Inlet Temp (°C)	Outlet Temp. (°C)	Operation Pressure
I	(508-513)	(465-477)	1.5-2.5 MPa
II	(508-530)	(473-484)	
III	(508-513)	(478-487)	

MODELING DETAILS AND ASSUMPTIONS

In predicting the model of the catalytic reforming system and taking into account the following assumptions:-

1- the system is Adiabatic, with steady-state and plug-flow modes.

2- Both the reactants and the products are ideal gases.

3- Catalytic reforming processes in each reactor happen simultaneously and are first-order regarding reactants.

4- neglected longitudinal molar and temperature distributions, only radial directional factors are analyzed.

5- kinetic analysis focused on perfecting the complicated combination of naphtha. using Individual components to describe the naphthene, aromatic, and paraffin groups based on the Smith model.[4]

5- The three feedstock chemicals (paraffin, naphthenes, and aromatics) have the same number of carbon atoms[4].

6- neglected Estimating the distribution of the molar flow of light gasses (C_1-C_5) formed by paraffin and naphthenic hydrocracking reactions since they are tiny.

7-Kinetics proposed by Smith was adopted to describe the reactions occurring during catalytic reforming [7].

The reaction network used in this model and the corresponding rate equations are presented as follows[7].

Conversion of napthenes to aromatics $\longleftrightarrow$ C_nH_{2n} C_nH_{2n-6} + $3H_2$ …(1)

Conversion of paraffins to napthenes $CnH_2n+2 \longleftrightarrow$ CnH_2n + H_2 …(2)

Hydrocracking of parffins $CnH_2n+2 \longleftrightarrow$ (n-3)/3 H_2 $n/15C_1+n/15$ C_2 $+n/15C_3+n/15C_4+n/15C_5$ …(3)

Hydrocracking of napthenes $CnH_2n+n/3$ H_2 $n/15$ $C_1+n/15C_2+n/15C_3+n/15C_4+n/15C_5$ …(4)

$r_1= \eta k_{f\ 1} (P_E –P_A P_H^3 /K_{eq1})$ …(5)

$k_{f1}= 9.87\exp(23.21–34750/T)$ …(6)

$K_{eq1}=P_A P_H^3 /P_E=1.04\times10–3 \exp(46.15–46045/T)$ …(7)

$r_2= \eta k_{f\ 2} (P_EP_H–P_P/K_{eq2})$ …(8)

$k_{f2} = 9.87\exp(35.98–59600/T)$ …(9)

$K_{eq2} = P_P/(P_E\cdot P_H)=9.87\exp(800/T –7.12)$ …(10)

$r_3 = \eta k_{f\ 3} P_P /P$ …(11)

$k_{f3} = \exp(42.97–62300/T)$ …(12)

$r_4 = \eta k_{f\ 4} P_E / P$ …(13)

$k_{f4} = \exp(42.97–62300/T)$ …(14)

To calculate the effectiveness factor from the following equations[13]:

$\eta= 1/\theta(1/\tanh(3\theta) -1/3\theta)$ …(15)

For reversible reactions of the nth order, the general expression of the Thiele module is:

$\theta = L[n+1/2]^{0.5}$ [k ρ_b $C_{AS}^{n-1}/De]^{0.5}$
…(16)

and for irreversible reactions of the order I:

$\theta = L[k$ ρ_b $/De]^{0.5}$
…(17)

The following equation represents the effective diffusion coefficient;

$$De = \frac{1}{1/Dij} + \frac{1}{1/DKA}$$
…(18)

Also, to calculate the diffusivity, using the Fuller equation to predict the diffusivity through the mixture [14, 15, 16],

$$D_{ij} = \frac{1*10^{-7}\, T^{1.74} \sqrt{\frac{1}{M_i}+\frac{1}{M_j}}}{P[\Sigma \upsilon_i^{\frac{1}{3}} + \Sigma \upsilon_j^{\frac{1}{3}}]}$$
…(19)

Calculation of the Knudsen diffusivity is independent of pressure P [17, 18]:

$$D_{KA} = 0.97\, r \sqrt{\frac{T}{M_A}}$$
…(20)

For every constituent (Paraffins, Naphthenes, and Aromatics) and Recycle Hydrogen, the kinetic rate formulae can be derived as follows [7] :

$dN_A/dR = 2\pi RL\rho_b r_1$
… (21)

$dN_N/dR = 2\pi RL\rho_b(-r_1 - r_2 - r_4)$
…(22)

$dN_P/dR = 2\pi RL\rho_b(r_2 - r_3)$
…(23)

$dN_H/dR = 2\pi RL\rho_b(3r_1 - r_2 - ((n-3)/3)r_3 - (n/3) r_4)$
…(24)

The varying molar rates of aromatics, naphthenes, paraffin, and hydrogen throughout the bed layer radius represent from Eq.(21) to Eq.(24). The following equation is applied to determine the temperature distribution along the reactor based on an energy balance across the differential reactor control volume, As mentioned in [7] :

$dT/dR = (2\pi RL\rho b)/(\Sigma FiCpi) \times \{3\ r_1\ (-\Delta H_1) + r_2\ (-\Delta H_2) + [(n-3)/3]\ r_3\ (-\Delta H_3) + (n/3)\ r_4\ (-\Delta H_4)\}$
...(25)

The value of R.O.N. can predict by the following equation, As mentioned in [12]:-

$RON_{mix}=Y_P \quad (RON)_P \quad +Y_A(RON)A+Y_N(RON)_N$ …(26)

$RON=a+ \quad bT+ \quad cT^2 \quad + \quad dT^3 \quad + \quad eT^4$ …(27)

Where R.O.N. denotes the research octane number and T denotes the boiling point in kelvin. Information obtained from the API-TDB, the coefficients a - e were determined shown in the table(2) below:

Table(2) RON estimation coefficients

Hydrocarbons	**A**	**b**	**C**	**D**	**e**
Paraffins	487.837	-230.5	475	-780	310
Naphthenes	-77.536	471.59	-418	100	0
Aromatics	145.668	-54.336	16.276	0	0

RESULTS AND DISCUSSION:

Basrah refinery suffers from several technical problems. Accordingly, the plant is operating at a lower than the design capacity. Also, the value of the octane number of light naphtha production fluctuates from time to time due to the service life of the equipment expiring; also, the catalyst's performance in the reformer system is required replacement. The regeneration process and improving the catalyst effectiveness factor are necessary to plant turn off while there is a severe shortage of gasoline supplies in the city. For this reason, to ensure and cover the gasoline lack in gasoline stations, the controlling of reformer operation temperature is represented as one of the technical and temporary solutions until the new light naphtha project is complete.

A mathematical model constructs relative to the smith model to evaluate the catalyst's performance in a reformer system. As shown in equations (21 to 25), the differential equations of reaction rates were solved using the Euler method to find molar and temperature distribution. The theoretical model results compare with Ke-min results, where the agreement was more than 90%. Table(3) shows the temperature and molar profiles from the simulated model and Ke-min. Table (4) shows the percentage of errors relative to Aromatics compounds.

Table (3) Simulation Results related to Ke-min work.

Reactor ID	**Position**	**Bed radius (mm)**	**T(°C) Simulated**	**Aromatics (kmol/h) Simulated**	**T(°C) Kemin**	**Aromatics (kmol/h) Kemin**
I	**Inlet**	510	521	16.180	521	16.180

	Outlet	170	454.068	51.103	468.26	51.084
II	**Inlet**	585	521	51.103	521	51.084
	Outlet	170	471.249	77.342	478.78	79.457
III	**Inlet**	760	521	77.342	521	79.457
	Outlet	170	492.519	94.706	492.72	101.342

Table (4) Percentages of errors in the simulated and Ke-min models

Temperature Error %	**Aromatics (Kmol/h) Error%**
6.461	3.744

The performance assessment of catalyst effectiveness factor in B.R.P. evaluates relative to actual operating conditions and output temperatures from the third catalytic reformer, as shown in Tables (5) and (6). The effectiveness factor values, which ranged from 0.92 to 0.546, were used to calculate the model's results, and the results showed that the simulation and plant operating conditions were in agreement.

Table (5) shows the B.R.P. operating conditions.

Operation Conditions	**Value**
Inlet Temp. (°C)	508-513
Operating Pressure (MPa)	1.5-2.5
Feed flow rate kmol/hr	436.494
Mole% Naphthenes	66.459
Mole% Aromatics	14.890
Mole% Paraffins	17.213
RON	40-65

Table (6) shows the comparison between B.R.P. and model results.

Item	**Plant Results**	**Model Results**
Inlet Temp. (°C)	480	480
Outlet Temp. (°C)	475	463.882

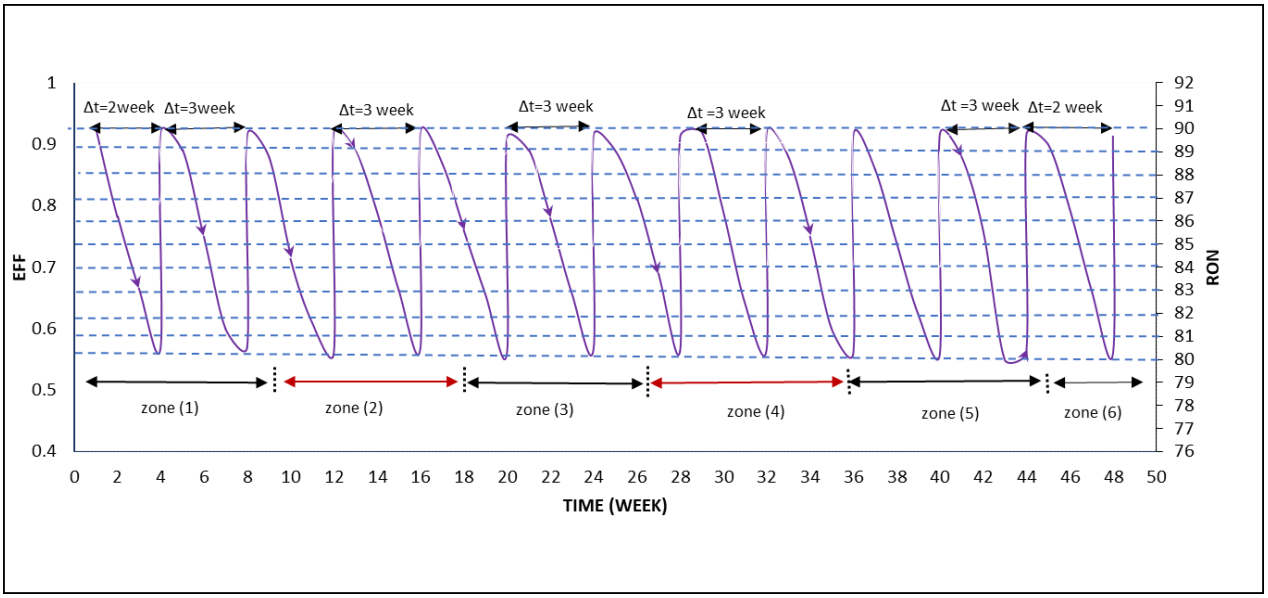
Mole% Naphthenes	3.306	3.693
Mole% Aromatics	31.292	34.783
Mole% Paraffins	63.374	61.523
R.O.N IBP= 30 (°C) , EBP=80 (°C)	87	89.780

Data collection of outlet operating temperature, light naphtha composition, operating pressure, and measured octane number for more than one year recorded and analyzed to predict catalyst effectiveness factors' values. Table (7) shows some of the selected plant operation conditions recorded through the research period and the expected catalyst effectiveness factors.

Table (7) shows the real operation conditions of B.R.P

Time (week)	**feed (kg/hr)**	**R.G (Nm^3/hr)**	**RON**	**Inlet temp. (°C)**	**Outlet temp. (°C)**	**P mol%**	**N mol%**	**A mol%**	**Operating pressure (MPa)**	**Effectiveness factor**
1	31300	66500	90	470	458.37	0.66	0.06	0.27	2	0.92
2	33700	63700	86	475	462.27	0.66	0.06	0.28	2	0.78
3	38500	67000	83	480	466.43	0.66	0.06	0.29	2	0.66
4	38500	69000	80	485	471.30	0.65	0.05	0.30	2	0.569
4	31300	73000	90	470	459.00	0.66	0.07	0.27	2	0.92
5	31300	66714	89	470	458.39	0.66	0.06	0.27	2	0.89
6	33700	67117	85	475	462.61	0.66	0.06	0.28	2	0.75
7	37500	65700	81	480	466.46	0.65	0.06	0.29	2	0.6
8	41000	67000	80	485	470.70	0.65	0.05	0.30	2	0.57
8	31600	71000	90	470	458.75	0.66	0.07	0.27	2	0.915

Table (7) illustrates the results of the catalyst's effectiveness factor to the exact operating conditions of the Basrah refinery, demonstrating that raising the flow rate reduces the catalyst's effectiveness factor. This drop increases the relative impedance to mass transfer inside the catalyst body.



Increasing the flow rate decreases the resistance to heat transfer through the liquid, which minimizes the temperature rise of the catalyst. The results show that the efficacy factor has varied between 0.927 and 0.546, while the octane number has varied between 80 and 90, as seen in figure (1).

Figure (1): The effect of operating conditions on effectiveness factor values of the catalyst and R.O.N. with time.

Figure(1) shows the fluctuation of the catalyst effectiveness factor as a function of time. For the first period and after 14 days of operation, the octane number and effectiveness factor values have reduced from 90-80 and 0.92-0.,547 respectively, while the operating temperature is about (470 - 480 ^{o}C). After this period, for Improving the values of octane number to be more than 90, the operating temperature has increased to (485 ^{o}C). The same behavior in the value of catalyst effectiveness factor is repeated from time to time, and then it is improved by controlling the operating temperature. The results showed that the process of raising the reaction temperature helps to renew the activity factor of the catalyst and to ensure the production of high-octane gasoline. This procedure is considered beneficial as it increases the activity of the catalyst for a more extended period until the catalyst is replaced or reactivated.

CONCLUSIONS

The accuracy of the model's results relative to actual operating condition results is about (90%). Also, the results showed the detailed profile of catalyst effectiveness factor as a function of actual operating conditions and time have changed from (0.927-0.546) when the exact operating condition of temperature is the changed from(470-485 **°C**) and pressure (2 MPa) for enhancing the value of R.O.N. between of (80-90).

NOMENCLATURES

A= Denote to Aromatic.
B.R.P= Basrah Refinery Plant.
Cp= constant of Heat capacity (kJ/ kmol H_2. °C).
F= flow rate of feedstock (kmol/hr).
F_i= component rate of Feed flow (kmol/hr).
H= Denote Hydrogen.
$Hr°_T$ (1,2,3&4) = Heat of reaction at any temperature for reaction 1,2,3&4 respectively (kj/ kmol).
k_{f1}= Is the constant reaction rate related to r_1, kmol/(h.kg cat.MPa).
k_{f2} = is the constant reaction rate related to r_2, kmol/(h.kg cat.MPa^2)
k_{f3}, k_{f4} = is the constant reaction rate related to r_3, and the constant reaction rate related to r_4, kmol/(h.kg cat) **K_{eq1}**= is the cycloalkane equilibrium constant converted to arene, $(MPa)^3$.
K_{eq2}= is the cycloalkane-to-alkane conversion equilibrium constant (MPa^{-1}).
L= catalyst bed height of reactor(mm).
M_f = feedstock average molecular weight (kg/kmol).
M_A= Arene molecular weight (kg/kmol).
M_E= Cycloalkane molecular weight (kg/kmol).
M_P= Alkane molecular weight (kg/kmol).
n= carbon atoms number.
N= Denote to Naphthene.
N_i= component Molar flow (mkol/ hr).
P= Denote Paraffin.
pi = the pressure of the process or the partial pressure of the constituents
P =Total pressure (MPa).
R_i= radius of Inlet reactor.
R_o= radius of Outlet reactor.
r_1= Denotes the rate of cycloalkane transformation to arene. in kmol/(h.kg cat).
r_2= Denotes the rate at which cycloalkane is transforming to alkane. in kmol/(h.kg cat).
r_3= Denoted the rate of a reaction hydrocracking of paraffin in kmol/(h.kg cat).
r_4= Denoted the rate of a hydrocracking reaction of naphthenes in kmol/(h.kg cat).
T= Temperature in of reactor (^oC).
T_o= Temperature out of reactor (^oC).
V= Volume of the reactor (mm^3).
y_i= component mole fraction.
ρ_b= Bulk density of catalyst (Kg.cat / mm^3).
η = Catalyst effectiveness factor.
RON_{mix}= denotes the research octane number of mixture
RON_P= denotes the research octane number of Paraffin.
RON_N= denotes the research octane number of Naphthene.
RON_A= denotes the research octane number of Aromatic.
θ = denotes the Thiele Modulus.
Dij = denotes the Diffusivity of i through j m^2/sec .

DK = denotes the Knudsen diffusivity m^2/sec .
De = denotes the Effective diffusivity m^2/sec .

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