Material Balance And Reaction Kinetics Modeling For Penex Isomerization Process In Daura Refinery

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Abstract. Penex Deisohexanizer isomerization of light straight run naphtha is a significant process for petroleum refining and proved to be effective technology to produce gasoline components with a high octane number. Modeling of the chemical kinetic reactions is an important tool because it is a better tool for optimization of the experimental data into parameters used for industrial reactors. The present study deals on the isomerization process in Daura refinery. Material balance calculations were done mathematically on the unit for the kinetics prediction purpose. A kinetic mathematical model was derived for the prediction rate constants K1 and K2 and activation energy E at operating temperatures range 120-180°C. According to the model, the results show that with increasing of temperature leads to increased K2 directly, where the K2 values proportional inversely. The activation energy results show that Ea1 (nC6) < Ea1 (C5) < Ea1 (CH), and for Ea2 (IC5) < Ea2 (2,2-DMB), < Ea2 (2,3-DMB) < Ea2 (MCP).

1 Introduction

The Penex Deisohexanizer (DIH) isomerization is a process operated using the catalytic reaction of n-pentanes, n-hexanes, and mixtures thereof to produce isomerate hydrocarbons. The reactions take place in a hydrocatalytic fixed bed reactor to promote conversion and minimize hydrocracking [1]. The process of light naphtha can produce isomerate hydrocarbons with higher octane number, in addition, that the typical Penex unit product can blend into gasoline pool, and the non-converted low octane components (nC5, CH and MP’s) from the deisohexanizer column (DIH) can be recycled to the reactor section for further upgrading [2].

The major elements of Penex (DIH) isomerization processes are reactors operated using chlorinated–alumina (Pt/Al2O3-Cl) as catalyst [3, 4]. This catalyst is proven to be active at lower temperatures (120-180°C) in which equilibrium favorable to produce iso-paraffins. [5]. The scheme of Penex (DIH) process is illustrated in Fig. 1. [6]

The most important process variables in Penex isomerization unit are the reactor temperatures. The higher temperatures than equilibrium lead to increase the amount of hydrocracking and increase the carbon formation on the catalyst.

A typical UOP Penex unit is provided with two reactors in series. All of the benzene rings in the LSRN feed are hydrogenated in the first reactor and some conversion of cyclohexane (CH) and methyl cyclopentane (MCP) to hexanes also occurs, as does some hydrocracking of C7 components to C3 and C4.

The hydrogenation of benzene rings, naphthene conversion to hexane, and C7 hydrocracking are exothermic reactions and, for a typical feedstock, contribute more to the temperature rise in the reactor than exothermic reaction of paraffin isomerization.

The concentrations and outlet temperature will be influenced by the catalyst and by the mass of C6 cyclic and C7 components in the feed.

2 Penex Unit Reaction Mechanism

2.1 nC5 Reaction Mechanism [7]

The Isomerization reaction of normal paraffins to isoparaffins is slightly exothermic and thermodynamically favorable at a temperature range 120-180°C. The kinetics of the reaction proceed in series through an olefin as an intermediate product formed by dehydrogenation of n-paraffins through an adsorption mechanism on the surface of metal site catalyst.

\[ n \text{pentane} \rightarrow n \text{pentene} \]

Because of low equilibrium conversion of paraffin isomerization, n-olefins can be converted to carbonium ion with the aid of injection of strong chloride acid as initiator.

\[ n \text{pentene} + [H^+] [A^-] \rightarrow \text{carbonium ion} + [A^-] \]

This initiator ion allows equilibrium forward to removes n-olefin from the first hydrogenation reaction, and then to rearrange the molecules.

\[ \text{carbonium ion} \rightarrow \text{iso pentane carbonium ion} \]

The high catalytic acidity causes a hydrogenation reaction to proceed at a higher reaction rate. Then, the carbonium ion converted iso-paraffin to iso-olefin by dehydrogenation step.

\[ \text{iso pentane carbonium ion} \rightarrow \text{iso pentene} \]

In the last reaction, the iso-olefin is hydrogenated again to form iso-paraffin in the presence of surface of the catalyst.

\[ \text{iso pentene} \rightarrow \text{iso pentane} \]
Generally, the final steps of (n-C<sub>6</sub>) isomerization proceed similarly, thus (n-C<sub>6</sub>) several iso-products like 2-MP, 3-MP, 2,2-DMB, and 2,3-DMB is can be produced.

2.2 nC6 Reaction Mechanism [8]

\[
\begin{align*}
    n \text{pentane}_{(RON= 24.8)} & \xrightleftharpoons{pt} 2MP_{(RON= 73.4)} \\
    n \text{pentane}_{(RON= 24.8)} & \xrightleftharpoons{pt} 3MP_{(RON= 74.5)} \\
    n \text{pentane}_{(RON= 24.8)} & \xrightleftharpoons{pt} 2,2 \text{ DMB}_{(RON= 91.8)} \\
    n \text{pentane}_{(RON= 24.8)} & \xrightleftharpoons{pt} 2,3 \text{ DMB}_{(RON= 104.3)}
\end{align*}
\]

The relationship between equilibrium conversion vs. isomerization reaction temperatures of (C<sub>5</sub> and C<sub>6</sub>) paraffins are shown in Fig. 2 and 3, respectively [5].

2.3 Other Reactions [9]

There are several reactions occurring inside and outside of the reactors.

2.3.1 Inside Reactors:

a) Naphthene ring of methyl cyclopentane (MCP) and cyclohexane (CH) present in the Penex feed will hydrogenate to n-paraffins, and then to iso-paraffins. The increasing in reactor temperature leads to increase Naphthene ring opening reactions.

b) As the temperature increased, the naphthenes shift forwards to (MCP) production.

c) Very quickly hydrogenation reaction of the benzene ring in the presence of a catalyst (Pt/Al<sub>2</sub>O<sub>3</sub>-Cl) proceed at very low temperature.

\[
\begin{align*}
    \text{benzene}_{(RON> 100)} +3H_2 & \xrightleftharpoons{pt} \text{cyclohexane}_{(RON= 84)} \\
    \text{C}_7\text{Cl}_4 + 5H_2 & \xrightleftharpoons{(Pt/Al_2O_3-Cl)} 4\text{HCl} + \text{C}_5\text{H}_6
\end{align*}
\]

d) Hydrocracking of Penex feed according to types of feed quality. For example, C<sub>7</sub> molecules tend to hydrocrack easily than smaller ones, paraffins of C<sub>5</sub> and C<sub>6</sub> hydrocrack to some extent. The severity of hydrocracking tends to reduce yield and increase product temperature.

2.3.2 Outside Reactors:

a) Hydrogen chloride is injected annually at period of maintenance time, to limit corrosion problems in units, the reaction run until removed any rust present in the unit.

\[
6\text{HCl} + \text{Fe}_2\text{O}_3 \rightarrow 2\text{FeCl}_3 + 3\text{H}_2\text{O}
\]

b) Perchloroethylene (C<sub>2</sub>Cl<sub>4</sub>) is injected before reactors at approximately the temperature of (110°C) or higher. The present hydrogen will react with this promoter in the presence of a catalyst (Pt/Al<sub>2</sub>O<sub>3</sub>-Cl) to produce hydrogen chloride.

\[
\begin{align*}
    \text{C}_2\text{Cl}_4 + 5\text{H}_2 & \xrightleftharpoons{(Pt/Al_2O_3-Cl)} 4\text{HCl} + \text{C}_4\text{H}_8
\end{align*}
\]

c) Neutralization of hydrogen chloride formed in above reaction using caustic soda (NaOH) to form salt and water in the scrubber.

\[
\text{HCl} + \text{NaOH} \rightarrow \text{NaCl} + \text{H}_2\text{O}
\]
3 Material Balance

As shown by Fig. 1, there are three streams entering to the rector of Penex unit, the feed (LSRN), makeup gas (H₂) and recycle (from Deisohexanizer) and outlet stream exit as a product from the reactor. The product stream (F3) exits from the reactor, enters to stabilizer column. In this process, overhead light gases (CH₄, C₂H₆, C₃H₈, and i-C₄H₁₀) and some of nC₅ and iC₅ which is removed and exit from the top of column, in addition to H₂ and HCl, which then enter to scrubber.

The outlet product from the top of stabilizer column enters to the scrubber column where NaOH solution is injected into the scrubber. From the bottom of the stabilizer, the product stream contains (nC₅, iC₅, nC₆, 2-MP, 3-MP, 2,2-DMB, 2,3-DMB, MCP and CH) which goes to Deisohexanizer (D IH) column. The outlet product from the bottom of stabilizer enters to the DIH column. There are two streams exit from DIH column, top stream (D) and bottom stream (W).

The mass balance calculations on the unit assuming feed rate of Penex unit = 10,000 BPD is presented in Table (1) and (2) respectively.

Table 1: Material Balance Calculations on the Reactor and Stabilizer Unit Assuming Feed Rate = 10,000 BPD

| Comp. | Mwt | Density (Kg/m³) | F1 in | F2 in | R in | F3 out | F3 in | F4 out | F5 out |
|-------|-----|----------------|-------|-------|------|--------|-------|--------|--------|
| nC₅  | 72  | 626            | 11276.16 | 0     | 199.731 | 5278.9 | 5278.9  | 18.47  | 5260.43 |
| iC₅  | 72  | 616            | 5688.17  | 0     | 135.817 | 12020.96 | 12020.96 | 281.52 | 11739.44 |
| nC₆  | 86  | 665            | 14808.4  | 0     | 17440.5 | 17736.79 | 17736.79 | 0     | 17736.79 |
| 2-MP  | 86  | 653            | 4989.84  | 0     | 24902.4 | 25382.58 | 25382.58 | 0     | 25382.58 |
| 3-MP  | 86  | 664            | 3915.2   | 0     | 21467.1 | 21451.51 | 21451.51 | 0     | 21451.51 |
| 2,2-DMB | 86 | 674        | 139.37    | 0   | 719.031 | 12145.51 | 12145.51 | 0     | 12145.51 |
| 2,3-DMB | 86 | 695        | 1572.3    | 0   | 6559.16 | 11356.35 | 11356.35 | 0     | 11356.35 |
| MCP   | 84  | 749            | 39.199    | 0     | 23.968 | 8738.38  | 8738.38  | 0     | 8738.38 |

Table 2: Mass Balance Calculations on the Scrubber and Deisohexanizer Unit Assuming Feed Rate = 10,000 BPD

| Comp. | Mwt | Density (Kg/m³) | F4 in | S1 out | S2 out | F5 in | R out | W out | D out |
|-------|-----|----------------|-------|--------|--------|-------|-------|-------|-------|
| nC₅  | 72  | 626            | 11276.16 | 0     | 199.731 | 5278.9 | 5278.9  | 18.47  | 5260.43 |
| iC₅  | 72  | 616            | 5688.17  | 0     | 135.817 | 12020.96 | 12020.96 | 281.52 | 11739.44 |
| nC₆  | 86  | 665            | 14808.4  | 0     | 17440.5 | 17736.79 | 17736.79 | 0     | 17736.79 |
| 2-MP  | 86  | 653            | 4989.84  | 0     | 24902.4 | 25382.58 | 25382.58 | 0     | 25382.58 |
| 3-MP  | 86  | 664            | 3915.2   | 0     | 21467.1 | 21451.51 | 21451.51 | 0     | 21451.51 |
| 2,2-DMB | 86 | 674        | 139.37    | 0   | 719.031 | 12145.51 | 12145.51 | 0     | 12145.51 |
| 2,3-DMB | 86 | 695        | 1572.3    | 0   | 6559.16 | 11356.35 | 11356.35 | 0     | 11356.35 |
| MCP   | 84  | 749            | 39.199    | 0     | 23.968 | 8738.38  | 8738.38  | 0     | 8738.38 |

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MATEC Web of Conferences 111, 02012 (2017)
The concentration of reactants in the isomerization reactions (nC₅, nC₆ and CH) in the inlet stream to the reactor is calculated from the mass balance, these concentrations are used to calculate K₁ (rate constant for the formation of intermediate olefin). The results are shown in Table (3).

Whereas the mole fraction of the produced isomers outlet from reactor (iC₅, 2,2-DMB, 2,3-DMB and MCP) are calculated from the mass balance, these mole fractions are used to calculate K₂ (rate constant for the formation of isomers).

Table 3 Concentration of Reacted Components

| Component | \( F_{Ao} \) (Kmole) | \( V \) (m³) | \( C_{Ao} = \frac{F_{Ao}}{V} \) (Kmole/m³) |
|-----------|-----------------------|-------------|----------------------------------|
| nC₅       | 156.613               | 18.013      | 8.694                            |
| nC₆       | 172.191               | 22.608      | 7.161                            |
| CH        | 104.03                | 11.23       | 9.26                             |

4 Development Model

In the typical isomerization process, several reactions are taken place: paraffin isomerization; naphthene hydrogenation; naphthene isomerization, benzene saturation, hydrocracking; and naphthene alkylation. Table (4) list the reactions and equilibrium conversion suggest for isomerization of normal paraffinic components in the feed of Penex process. [10]

The model developed in this study are taken following assumptions: 1st order reactions at steady state, isothermal conditions, gas phase reaction, constant physical properties, and neglected gradient in pressure drop, temperature and concentration respectively.

Mass balance has been developed over the cross section of the segment of the catalyst bed, as illustrated in Fig. 3.

The design equation for volume of tubular reactor:

\[
V = F_{Ao} \int_0^L \frac{dx}{-K_1 C_{Ao}}
\] (12)

And reactant concentration of \( C_A \) in gas phase:

\[
C_A = C_{Ao} \left( \frac{1 - x}{1 - ex} \right)^{\frac{T}{T_o}}
\] (13)

Where \( \epsilon \) is refer to voidage in reactor bed.

Putting equation (13) into (12) and re-arrange, to get the reaction rate constant K₁ as shown in equation (14).

From Arrhenius equation (15), the activation energy \( (E_{a1}) \) and frequency factor \( (K_0) \) can be calculated by plot \( (\ln K_1) \) vs. \( (1/T) \), as illustrated in Fig. 4.

5 Results And Discussion

The model shown by equation (14) can be used to calculate isomerization reaction rate constant K₁, using data given from Penex unit in the Daura Refinery: Volume of catalyst in each reactor =35.66 m³, Hold up = 75 %, \( \epsilon = 0.393 \), and \( T_o = 50^\circ C \).

Table (5) represents the results of K₁ at temperatures range of 120-180 °C. It was shown that calculated reaction constants of \( (K_1) \) values increased with increasing temperature and reach maximum value at a temperature (180°C).

Applying equation (15) and plot \( (\ln K_1) \) vs \( (1/T) \) for components listed in Table (6) are shown in Fig. 5.

The activation energy \( (E_{a1}) \) and Frequency factor \( (K_{01}) \) are calculated from these plots and tabulated in Table (7). The results show that \( E_{a1}(nC6^*) < E_{a1}(nC6^{**}) < E_{a1}(C5) < E_{a1}(CH) \).

Table 4 A Typical Reactions for LSRN Isomerization

| Reactions | Conv. % | Equ. No. |
|-----------|---------|----------|
| nC₅ ⇔ iC₅ | 54      | (1)      |
| nC₆ ⇔ 2,2-DMB | 35 | (2) |
| nC₆ ⇔ 2,3-DMB | 10 | (3) |
| BZ+3H₂ ⇔ CH | 100 | (4) |
| CH ⇔ MCP | 59      | (5)      |
| 2-MP → 3-MP |       | (6)      |
| 3-MP → 2,2-DMB + 2,3-DMB |       | (7)      |
Fig. 4: The Relationship Between (ln $K_1$) vs. (1/T)

Table 5: Results of $K_1$.

| T(°C) | T(K) | $K_1$ (h⁻¹) |
|-------|------|-------------|
|       |      | nC₅ | nC₆* | nC₆** | CH |
| 120   | 393  | 0.398| 0.241| 0.056  | 0.289|
| 135   | 408  | 0.413| 0.250| 0.058  | 0.300|
| 150   | 423  | 0.429| 0.260| 0.060  | 0.311|
| 165   | 438  | 0.444| 0.269| 0.062  | 0.322|
| 180   | 453  | 0.459| 0.278| 0.065  | 0.333|

Where nC₆* for nC₆ ↔ 2,2-DMB at x=0.35, and nC₆** for nC₆ ↔ 2,3-DMB at x=0.1

Table 6: ln$K_1$ and 1/T Values.

| 1/T   | ln$K_1$ | nC₅  | nC₆* | nC₆** | CH  |
|-------|---------|------|------|--------|-----|
| 0.0025| -0.9202 | -1.4164| -2.8759 | -1.2429|
| 0.0025| -0.8828 | -1.3789| -2.8385 | -1.2055|
| 0.0024| -0.8467 | -1.3428| -2.8024 | -1.1694|
| 0.0023| -0.8118 | -1.3080| -2.7675 | -1.1345|

Equation (11) can be used to calculate the rate constant $K_2$ for reaction on olefin to iso-paraffin. The required data were taken from Daura Refinery; as LHSV = 1.5, residence time $t = 1/ LHSV = 0.667$ h. The values of $K_2$ are shown in Table (8), which was shown that $K_2$ values decreasing with increasing temperature, which means that product isomerate concentrations were increased as a result of temperature sensitivity of isomerization reactions.

Applying equation (15) and plot (ln$K_2$) vs (1/T) for components listed in Table (9) are shown in Fig. 6. The values of the activation energy ($E_{a2}$) and Frequency factor ($K_{o2}$) are calculated from these plots and tabulated in Table (10). The results show $E_{a2}(iC5) < E_{a2}(2,2-DMB) < E_{a2}(2,3-DMB) < E_{a2}(MCP)$.

Fig. 5: Plot (ln $K_1$) vs. (1/T)

Fig. 6: Plot (ln $K_2$) vs. (1/T)

Table 7: Activation Energy and $K_{o1}$ Results.

| Component | $E_{a1}$ (KJ/mole) | $K_{o1}$ |
|-----------|---------------------|----------|
| nC₅       | 2738.822            | 0.460    |
| nC₆*      | 1525.743            | 0.263    |
| nC₆**     | 1835.182            | 0.061    |
| CH        | 2880.394            | 0.334    |

Table 8: Results of $K_2$.

| T(°C) | T(K) | $K_2$ (h⁻¹) |
|-------|------|-------------|
|       |      | iC₅ | 2,2-DMB | 2,3-DMB | MCP |
| 120   | 393  | 0.1547| 0.2566| 1.2062| 0.2566|
| 135   | 408  | 0.1480| 0.2477| 1.1512| 0.2477|
| 150   | 423  | 0.1430| 0.2389| 1.0998| 0.2389|
| 165   | 438  | 0.1384| 0.2308| 1.0529| 0.2308|
| 180   | 453  | 0.1346| 0.2232| 1.0099| 0.2232|

Table 9: ln$K_2$ and 1/T Values

| 1/T   | ln$K_2$ | iC₅  | 2,2-DMB | 2,3-DMB | MCP |
|-------|---------|------|---------|---------|-----|
| 0.0025| -0.8106 | -0.5907| 0.0814  | -1.8327|
| 0.0025| -0.8298 | -0.6062| 0.0612  | -1.9066|
| 0.0024| -0.8446 | -0.6218| 0.0413  | -1.9469|
| 0.0023| -0.8588 | -0.6368| 0.0224  | -2.0044|
| 0.0022| -0.8711 | -0.6513| 0.0043  | -2.0132|

Table 10: Activation Energy and $K_{o2}$ Results.

| Component | $E_{a2}$ (KJ/mole) | $K_{o2}$ |
|-----------|---------------------|----------|
| iC₅       | 1.503               | 0.281    |
| 2,2-DMB   | 1.519               | 0.349    |
| 2,3-DMB   | 1.931               | 0.602    |
| MCP       | 4.632               | 0.038    |

6 Conclusions
1. The increasing temperature leads to increased $K_1$ according to reactor model (proportional directly).

2. The $K_2$ values decreased when the temperature increased for the isomerate (proportional inversely) which indicates the increasing in the isomerate concentration.

3. The activation energy results show that $E_{a_{i(C_5)} < E_{a_{i(C_6)}} < E_{a_{i(CH)}}$, and for $E_{a_{2}(iC_5) < E_{a_{2}(2,2-DMB)} < E_{a_{2}(2,3-DMB)} < E_{a_{2}(MCP)}$, which means the reaction of lower activation energy have higher reaction rate.

### Abbreviations

| Abbreviation | Definition |
|--------------|------------|
| dz           | Integration Step for the Reactor Length |
| 2,2-DMB      | 2,2-Dimethylbutane |
| 2,3-DMB      | 2,3-Dimethylbutane |
| 2-MP         | 2-Methylpentane |
| 3-MP         | 3-Methylpentane |
| BZ           | Benzene |
| $nC_6^*$     | Concentration for $nC_6 \leftrightarrow 2,2$-DMB at $x=0.35$ |
| $nC_6^{**}$  | Concentration for $nC_6 \leftrightarrow 2,3$-DMB at $x=0.1$ |
| CA           | Concentration of n-paraffin present at time $t$ |
| CAo          | Concentration of inlet LSRN stream (mole/m$^3$) |
| Ciso         | Concentration of i-paraffin |
| CN           | Concentration of n-olefin |
| CH           | Cyclohexane |
| D            | Top stream of DIH shown in Fig. 1 |
| DIH          | Deisohexanizer (DIH) |
| $E_{a_1}$    | Activation energy for n-paraffin $\rightarrow$ Olefin |
| $E_{a_2}$    | Activation energy for Olefin $\rightarrow$ i-paraffin |
| F1           | Reactor feed shown in Fig. 1 |
| F2           | Hydrogen feed shown in Fig. 1 |
| F3           | Stabilizer feed shown in Fig. 1 |
| F4           | Scrubber feed shown in Fig. 1 |
| F5           | DIH feed shown in Fig. 1 |
| FAo          | Mole rate of inlet component LSRN |
| Fiso         | Mole rate of isomerate product |
| HC           | Hydrocarbon |
| i-C$_4$      | Iso-butane |
| i-C$_5$      | Iso-pentane |
| i-C$_6$      | Iso-hexane |
| K1           | Rate Constant for Paraffins (hr$^{-1}$) |
| K2           | Rate Constant for Olefins (hr$^{-1}$) |
| Ko           | Frequency Factor |
| LHSV         | Liquid Hourly Space Velocity |
| LSRN         | Light straight run naphtha |
| MCP          | Methyl cyclopentane |
| nC$_5$       | Normal pentane |
| nC$_6$       | Normal hexane |
| Pt           | Platinum |
| Pt/ZrO$_2$   | Sulfated zirconia |
| SO$_4$       | Chlorinated alumina |
| R            | DIH sidedraw shown in Fig. 1 |
| R           | Gas Constant (8.314 J/K mole) |
| RON          | Research Octane Number |
| t            | Residence time (hr) |
| S1           | NaOH solution feed to scrubber shown in Fig. 1 |
| S2           | Bottom of scrubber shown in Fig. 1 |
| S3           | Off gas outlet from the scrubber shown in Fig. 1 |
| T            | Temperature (K) |
| To           | Initial temperature (K) |
| V            | Volume of reactor (m$^3$) |
| W            | Bottom stream of DIH shown in Fig. 1 |
| $x$          | reaction conversion |
| $Z_t$        | Length of reactor (Cm) |
| $\varepsilon$| Voidage |

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