

# Optimization of the Pyrolysis Gasoline Hydrogenation Reactor Considering the Hydrogen Network Integration

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A mathematical model is developed considering the pyrolysis gasoline (pygas) hydrogenation kinetic, the temperature of the second-stage pygas reactor, hydrogen network integration, the variation of catalyst activity and product quality along time. The industrial data is employed to fit the linear equation about the average temperature of the second-stage reactor and operation time of catalyst. The relationship between catalyst activity and operation time is deduced, and the temperature of second-stage reactor is optimized within a certain range with the product quality requirements satisfied, as well as the economic benefit. Case study shows that this model can provide guidance for the design and improvement of pygas hydrogenation process.

## 1. Introduction

Pygas is an important by-product of ethylene industry with high content of aromatic hydrocarbon (benzene, toluene and xylene). Besides that, olefins, diolefins and organic compounds containing sulphur, nitrogen and oxygen also exist and these components make it unstable (Sun et al., 1999). Because of this, pygas needs to be hydrogenated before further processing. In industry, pygas is processed with a two-stage hydrogenation reactor. The diolefins are selectively hydrogenated in the first-stage, while olefins and compounds containing nitrogen, sulfur, and oxygen are hydrogenated in the second-stage. Both stages consume large amount of hydrogen. When the operating parameter of the reactor changes, the hydrogen consumption of the hydrogen network will change correspondingly. The reason is that the hydrogen separated from the effluent of this reactor is a source of the hydrogen network, while its inlet hydrogen is a sink. Therefore, the hydrogen consumption should be considered in the optimization of the pygas hydrogenation reactor.

Navid et al. (2005) developed a two-stage model for the hydrogenation of pygas and illustrated that the prediction of this model is in satisfactory agreement with the industrial unit. Qian et al. (2011) evaluated the introduction of Zn and Mo into the Ni-based catalyst and proved that this catalyst has excellent catalytic activity for the hydrogenation of pygas. Zhou et al. (2007) proposed a Langmuir-Hinshelwood-type kinetic model for the selective hydrogenation of pygas, and the predicted results show that competitive adsorption of diolefins and mono-olefins exists over the active sites of the catalyst. Later, this model was applied in liquid-phase selective hydrogenation of pygas, and a diffusion-reaction mathematical model was developed (Zhou et al., 2010).

Catalysts play a critical role in pygas hydrogenation, and its activity decreases along time. To compensate its deactivation and keep the reaction rate stable, the inlet temperature of the reactor is generally raised. This will have different effects on different reactions and should be considered in the optimization of pygas hydrogenation reactor. Castaño (2007) evaluated the effect of five different Pt-supported catalysts on conversion, selectivity and deactivation and provided a guidance for optimal catalyst design. Zhao (2006) analyzed the nickel-based catalyst applied in the first-stage pygas hydrogenation and reached a conclusion that CS<sub>2</sub> leads to its deactivation. Similarly, for the palladium-based catalyst, the main cause of deactivation is also CS<sub>2</sub> (Xue et al., 2014). Deng et al. (2014) located the flowrate targets of interplant hydrogen conservation network via the improved problem table (IPT). Mao et al. (2015) integrated the hydrogen network of a refinery with the hydrogen consumption of Vacuum Gas Oil (VGO) hydro-cracking reactor considered. However, no research considered the hydrogen network integration together with the catalyst activity.

In this work, the pygas hydrogenation reactor of a refinery is optimized with both the hydrogen network integration and the variation of the catalyst activity considered. The mathematical model will be built to analyze

the influence of the second-stage reactor's temperature on the hydrogen consumption of whole hydrogen network and the product quality, and identify its optimal value. A case is studied to show the validity of the proposed model.

## 2. Problem Statement

In refinery, the hydrogen stream separated from the effluent of the second-stage pygas hydrogenation reactor is generally the source (SR<sub>p</sub>) of hydrogen network, while the inlet hydrogen of this reactor is a sink (SK<sub>q</sub>). For this reactor, the conversion of each component is  $X_i$ . There are  $N_m$  hydrogen sources and  $N_n$  hydrogen sinks in a hydrogen network, the flow rate and hydrogen concentration of each hydrogen source are  $F_{SR_i}$  and  $C_{SR_i}$ , respectively; those of each hydrogen sink are  $F_{SK_j}$  and  $C_{SK_j}$ , respectively. The purity of fresh hydrogen is 99.99 %.

The objective is to identify the optimal parameters of the second-stage pygas hydrogenation reactor considering the hydrogen network integration.

## 3. Mathematical model

In the pygas hydrogenation reactor, the catalyst activity will decrease along time. To compensate the deactivation of the catalyst and keep the reaction rate stable, the inlet temperature is generally raised. However, the upper limit of temperature exists. If the reactor is operated at the temperature greater than the upper limit, the catalyst might deactivate rapidly, and the reaction rate of side reactions increases significantly, or more side reactions occur. This will affect the product and properties of the desired product (Li et al., 2016). Because of this, the catalysts should be changed or regenerated once the temperature reaches the upper limit. For the second-stage reactor of the pygas hydrogenation, the average temperature of catalyst bed can be adjusted along the operation time according the fitting equation shown by Eq. (1) (Zhang and Wang, 1996).

$$T_a = e + ft \quad (1)$$

Where  $T_a$  is the average temperature of catalyst bed, °C;  $t$  is the operation time, day;  $e$  and  $f$  are constants.

### 3.2 Relation between the conversion and operating temperature

In the second-stage reactor, diolefins unreacted in the first-reactor will be hydrogenated to mono-olefines, and the latter can be further hydrogenated. For the second-stage reactor, Eq. (2) shows the relation between the conversion( $X$ ) and the operating temperature.

$$X_i = 1 - \left[ 1 - \frac{Y \omega k_i \exp[-E_{ai}/R(T + 273.15)] p_i^{a_i} p_{H_2}^{b_i} (1 - a_i)}{F_{i,in}} \right]^{\frac{1}{1-a_i}} \quad (2)$$

Where  $X_i$  is the conversion of component  $i$ , %;  $Y$  is the catalyst activity in the second-stage reactor;  $\omega$  is the weight of catalyst, g;  $k_i$  is the pre-exponential factor;  $E_{ai}$  is the activation energy, J·mol<sup>-1</sup>;  $T$  is the reactor temperature, °C;  $p_i$  is the pressure of component  $i$ , Pa;  $P_{H_2}$  is the pressure of  $H_2$ , Pa;  $a_i$  and  $b_i$  are reaction orders;  $F_{i,in}$  is inlet flow of component  $i$ , mol·h<sup>-1</sup>.

When  $X_i$  is fixed, the relationship between  $Y$  and  $t$  can be deduced with Eq(1) substituted into Eq(2), as shown by to Eq(3).

$$Y = F_{i,in} \left( \frac{1 - (1 - X_i)^{1-a_i}}{1 - a_i} \right) / \omega k_i \exp \left( -\frac{E_{ai}}{R(e + ft + 273.15)} \right) p_i^{a_i} p_{H_2}^{b_i} \quad (3)$$

### 3.3 The effect of reactants conversion on the product price and hydrogen consumption

If the reactor is operated at the temperature different from that identified according Eq(1),  $X_i$  will change, and this will affect the product purity, as well as the product price. The relationship between the product purity and conversion is shown by Eq(4), while that between the product price and conversion is shown by Eq(5).

$$C = \sum_{i=1}^n g_i X_i + h_i \quad (4)$$

$$P = mC + n \quad (5)$$

Where  $P$  is the product price,  $\text{RMB}\cdot\text{t}^{-1}$ ;  $C$  is the total mass fraction of benzene, methylbenzene and dimethylbenzene in the product, %;  $g_i$  and  $h_i$  are constants related to reactants' stoichiometric factors;  $m$  and  $n$  are constants.

In the second-stage reactor,  $H_r$  is composed by two parts, the hydrogen consumption of diolefins hydrogenation ( $\sum_{i=1}^n \varphi_i F_i X_i$ ) and that of mono-olefins hydrogenation ( $H_m$ ), as shown by Eq(6). When the reactor's parameters change, the hydrogen consumption of second-stage reactor changes to  $H_r'$ . And the variation of hydrogen consumption can be calculated by Eq(7) (Mao et al., 2016).

$$H_r = \sum_{i=1}^n \varphi_i F_i X_i + H_m \quad (6)$$

$$\Delta H_r = H_r' - H_r \quad (7)$$

Where  $\varphi_i$  is the stoichiometric ratio of  $H_2$  to reactant  $i$  in the second-stage reactor;  $F_i$  is the flow of reactant  $i$ . According to the work of Mao et al. (2015), the hydrogen utility adjustment (HUA), which denotes the variation of the hydrogen consumption of the hydrogen network, is also affected by  $X_i$ . When the pinch appears at the sink-tie-line lying above  $SK_q$ , HUA can be calculated by Eq(8). And when it appears at the sink-tie-line lying between  $SK_q$  and  $SR_p$ , and below  $SR_p$ , HUA can be calculated by Eq(9).

$$\Delta F_u = - \frac{H_i}{c_u - c_{SR_i}} \quad (8)$$

$$\Delta F_u = \frac{1 - c_{SR_i} / c_{SR_p}}{c_u - c_{SR_i}} \Delta H_r - \frac{H_i}{c_u - c_{SR_i}} \quad (9)$$

Where  $c$  is the hydrogen concentration of each stream.

### 3.4 Objective of the optimization

With the price of product calculated by Eq(5) and that of hydrogen given by Li et al. (2016), the economic benefit can be calculated according to Eq(10). Based on this, the reactor parameters, including the operation temperature, can be optimized.

$$S = \Delta PM - P_{H_2} \Delta F_u \quad (10)$$

Where  $\Delta P$  is the variation of product price,  $\text{RMB}\cdot\text{t}^{-1}$ ;  $M$  is the weight of product,  $\text{t}$ ;  $P_{H_2}$  is the price of hydrogen,  $\text{RMB}\cdot\text{Nm}^{-3}$ .

## 4. Case study

The pygas hydrogenation reactor and hydrogen network of a refinery will be studied in this section. In the hydrogen network of this refinery, there are 26 sources and 14 sinks, and their data are shown in Table 1.  $SR_8$  and  $SK_4$  are connected to the second-stage pygas hydrogenation reactor. The initial temperature of second-stage reactor is  $280\text{ }^\circ\text{C}$ ; the hydrogen pressure is  $3\text{ MPa}$ ; pygas flow rate is  $29.9\text{ t}\cdot\text{h}^{-1}$  and its density is  $0.804\text{ g}\cdot\text{cm}^{-3}$ ; hydrogen to oil mole ratio is 3.6. In this reactor, there are the hydrogenation reactions of cyclopentene, styrene, hexene and thiophene, and the corresponding hydrogenation reaction kinetics are shown Table 2 (Mao et al., 2015), as well as the inlet flow rates and  $d_i$ , which is the amount of hydrogen required for hydrogenating per mole of component  $i$ .

The catalyst activity will decline along operation time, and reaction temperature is generally raised to keep the reaction rate stable. The fitted relationship between average temperature and operation time is shown by Eq(11). And the upper limit on the operating temperature of this reactor is  $300\text{ }^\circ\text{C}$  (Zhang and Wang, 1996).

$$T = 0.078t + 280 \quad (11)$$

Table 1: The data of sources and sinks

Streams	Purity/%	Flowrate /Nm <sup>3</sup> ·h <sup>-1</sup>	Streams	Purity/%	Flowrate /Nm <sup>3</sup> ·h <sup>-1</sup>	Streams	Purity/%	Flowrate /Nm <sup>3</sup> ·h <sup>-1</sup>
<b>Sources</b>								
SR <sub>0</sub>	97.60	29,800.00	SR <sub>9</sub>	90.00	8,650.00	SR <sub>18</sub>	91.97	5,314.00
SR <sub>1</sub>	99.80	14,000.00	SR <sub>10</sub>	90.00	54,500.00	SR <sub>19</sub>	74.99	300.00
SR <sub>2</sub>	95.00	1,000.00	SR <sub>11</sub>	88.00	63,000.00	SR <sub>20</sub>	69.67	170.00
SR <sub>3</sub>	95.00	2,700.00	SR <sub>12</sub>	89.70	67,000.00	SR <sub>21</sub>	65.80	500.00
SR <sub>4</sub>	93.50	640.00	SR <sub>13</sub>	88.00	31,000.00	SR <sub>22</sub>	63.96	474.00
SR <sub>5</sub>	93.00	22,800.00	SR <sub>14</sub>	88.00	49,000.00	SR <sub>23</sub>	58.60	300.00
SR <sub>6</sub>	92.00	26,500.00	SR <sub>15</sub>	87.00	6,500.00	SR <sub>24</sub>	50.00	3,000.00
SR <sub>7</sub>	91.97	500.00	SR <sub>16</sub>	82.00	200.00	SR <sub>25</sub>	55.00	200.00
SR <sub>8</sub>	91.00	18,000.00	SR <sub>17</sub>	82.00	2,300.00	SR <sub>26</sub>	55.00	50.00
<b>Sinks</b>								
SK <sub>1</sub>	99.78	1,400.00	SK <sub>6</sub>	92.41	28,000.00	SK <sub>11</sub>	87.97	55,000.00
SK <sub>2</sub>	99.60	4,000.00	SK <sub>7</sub>	91.70	9,100.00	SK <sub>12</sub>	89.40	67,000.00
SK <sub>3</sub>	99.60	5,000.00	SK <sub>8</sub>	88.21	73,000.00	SK <sub>13</sub>	87.45	7,650.00
SK <sub>4</sub>	96.00	25,000.00	SK <sub>9</sub>	90.26	56,000.00	SK <sub>14</sub>	92.10	5,954.00
SK <sub>5</sub>	93.58	25,000.00	SK <sub>10</sub>	89.74	38,000.00			

Table 2: Reaction kinetics of all the hydrogen consumption component

Components	Hydrogenation reaction kinetics	Inlet flow rate/mol·h <sup>-1</sup>	<i>d</i>
Cyclopentene	$r_1 = 1.422 \times 10^{-4} e^{-\frac{64845}{RT}} p_1^{0.724} p_{H_2}^{0.682}$	54,810	1
Styrene	$r_2 = 6.022 \times 10^{-5} e^{-\frac{65545}{RT}} p_2^{0.775} p_{H_2}^{0.475}$	1,524.38	4
Hexene	$r_3 = 4.022 \times 10^{-5} e^{-\frac{59845}{RT}} p_3^{0.764} p_{H_2}^{0.685}$	18,090	1
Thiophene	$r_4 = 7.251 \times 10^{-6} e^{-\frac{71855}{RT}} p_4^{0.73} p_{H_2}^{0.84}$	106.11	4
Methylthiophene	$r_5 = 8.311 \times 10^{-6} e^{-\frac{75955}{RT}} p_5^{0.83} p_{H_2}^{0.835}$	151.2	4
Dromethylthiophene	$r_6 = 9.629 \times 10^{-6} e^{-\frac{64855}{RT}} p_6^{0.87} p_{H_2}^{0.832}$	31.86	4

Table 3: The HUA of each sink-tie-lines

Sink-tie-lines	HUA / Nm <sup>3</sup> ·h <sup>-1</sup>	Sink-tie-lines	HUA / Nm <sup>3</sup> ·h <sup>-1</sup>
1	$\Delta F_{u,1} = -4,305.22$	5	$\Delta F_{u,5} = 0.181 \times \Delta H_{f_2} - 2,756.2$
2	$\Delta F_{u,2} = -2,805.73$	6	$\Delta F_{u,6} = 0.343 \times \Delta H_{f_2}$
3	$\Delta F_{u,3} = -2,348.94$	7	$\Delta F_{u,7} = 0.634 \times \Delta H_{f_2} - 337.87$
4	$\Delta F_{u,4} = 0.145 \times \Delta H_{f_2} - 2,181.45$		

In this hydrogen network, seven sink-tie-lines can intersect the sources and are numbered from 1 to 7; the Pinch can only appear at these sink-tie-lines (Mao et al., 2015). By the hydrogen surplus method, it is identified that the initial Pinch purity is 88 % (volume fraction) and the hydrogen utility is 27,235.63 Nm<sup>3</sup>·h<sup>-1</sup>. At the minimum utility consumption, the hydrogen surpluses of sink-tie-lines 1 to 7 are 198.04 mol·h<sup>-1</sup>, 157.12 mol·h<sup>-1</sup>, 155.03 mol·h<sup>-1</sup>, 165.79 mol·h<sup>-1</sup>, 217.74 mol·h<sup>-1</sup>, 0, and 52.71 mol·h<sup>-1</sup>, respectively. Sink-tie-lines 1, 2 and 3 lie above SK<sub>4</sub>, the corresponding T versus HUA relationship can be determined according to Eq(8). Sink-tie-lines 4, 5, 6 and 7 lie between SK<sub>4</sub> and SR<sub>8</sub>, the corresponding T versus HUA can be determined according to Eq(9). The results are shown in Table 3. The conversion of each components can be calculated based on Eq(2) and Table 2, as shown in Table 4. Substituting the conversions into Eq(6), Eq. (12) is obtained. According to Eq(3), Eq(11),

the data in Table 2 and the operating parameters, the relationship between catalyst activity about different reactants and operation time can be deduced respectively, as shown in Table 4.

$$H_r^i = 54,810X_{cyc} + 6,097.52X_{sty} + 10,890X_{hex} + 424.44X_{thi} + 604.8X_{met} + 127.44X_{dro} + 1,178.07 \quad (12)$$

Table 4: The conversion and catalyst activity of each components

Components	Conversion	Catalyst Activity
Cyclopentene	$X_{cyc} = 1 - \left[ 1 - 445,000Y \times \exp\left(-\frac{7,799.49}{T_2 + 273.15}\right) \right]^{3.623}$	$Y_{cyc} = 7.533 \times 10^{-7} / \exp\left(-\frac{7,799.49}{0.078t + 553.15}\right)$
Styrene	$X_{sty} = 1 - \left[ 1 - 30,082.64Y \times \exp\left(-\frac{7,883.69}{T_2 + 273.15}\right) \right]^{4.44}$	$Y_{sty} = 6.458 \times 10^{-7} / \exp\left(-\frac{7,883.69}{0.078t + 553.15}\right)$
Hexene	$X_{hex} = 1 - \left[ 1 - 236,788.62Y \times \exp\left(-\frac{7,198.1}{T_2 + 273.15}\right) \right]^{4.24}$	$Y_{hex} = 2.23 \times 10^{-6} / \exp\left(-\frac{7,198.1}{0.078t + 553.15}\right)$
Thiophene	$X_{thi} = 1 - \left[ 1 - 1,303,014Y \times \exp\left(-\frac{8,642.65}{T_2 + 273.15}\right) \right]^{3.7}$	$Y_{thi} = 1.57 \times 10^{-7} / \exp\left(-\frac{8,642.651}{0.078t + 553.15}\right)$
Methyl-thiophene	$X_{met} = 1 - \left[ 1 - 1,422,164Y \times \exp\left(-\frac{9,135.80}{T_2 + 273.15}\right) \right]^{5.88}$	$Y_{met} = 1.225 \times 10^{-7} / \exp\left(-\frac{9,135.795}{0.078t + 553.15}\right)$
Dromethyl-thiophene	$X_{dro} = 1 - \left[ 1 - 1,860,975Y \times \exp\left(-\frac{7,800.70}{T_2 + 273.15}\right) \right]^{7.69}$	$Y_{dro} = 3.078 \times 10^{-7} / \exp\left(-\frac{7,804.306}{0.078t + 553.15}\right)$

If the product quality that affects product price is allowed to change within a given interval, based on the hydrogen network integration, the average temperature can be optimized with HUA and product price taken into consideration, as shown by Eq(13). The price of hydrogen is taken as 1.52 RMB·Nm<sup>-3</sup> (Li et al., 2016), and the mass flow rate of the aromatics product is 28 t·h<sup>-1</sup> (Hao, 2007).  $\Delta F_u$  is identified according to Table 3 and the method presented in Mao (2016). And  $P$  can be calculated by Eq(14) (Hao, 2007). According to the mass balance, the purity of benzene, methylbenzene and dimethyl in the benzene aromatic product,  $S$  can be calculated by Eq(15). Based on Tables 3 and 4, Eqs(12) and (13), the  $S$ - $T$ - $t$  surface is plotted in the three-dimensional diagram, as shown by Figure 1a. This diagram clearly shows the variation of the economic benefit along time and operating temperature.

$$S = 28P - 1.52\Delta F_u \quad (13)$$

$$P = 9,400C - 5075.9 \quad (14)$$

$$S = 0.63 + 0.203X_{cyc} + 0.0057X_{sty} + 0.067X_{hex} \quad (15)$$

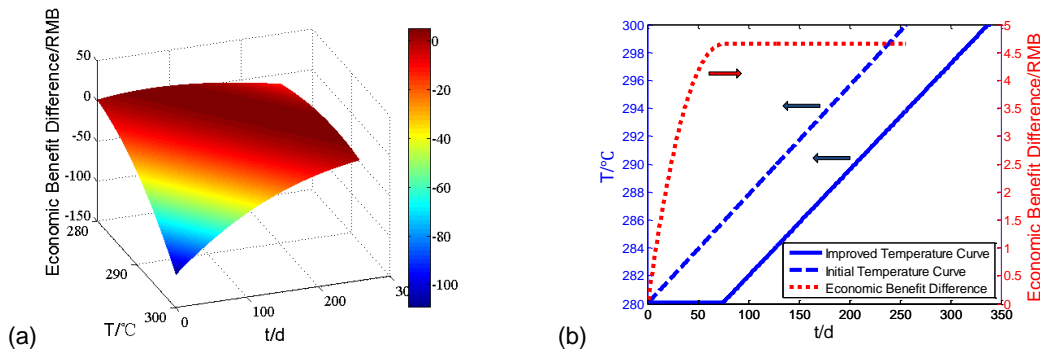


Figure 1: (a) The  $S$ - $T$ - $t$  three-dimensional diagram. (b) The temperature against operation time with maximum economic benefit

According to Eq(11), the initial temperature curve is drawn in Figure 1b. The optimal temperature at different operation time also can be calculated according to Table 4, as shown by the improved temperature curve. Furthermore, the variation of the economic benefit along operation time is illustrated by the economic benefit curve. According to the initial temperature curve, when  $t = 256$ d, the temperature reaches to the upper limit. If the reactor is operated according the improved temperature curve, the operation time of the catalyst can be raised to 337days, as shown in Figure 1b.

## 5. Conclusion

A model is presented for optimizing the temperature of the second-stage reactor of pyrolysis gasoline hydrogenation unit based on hydrogen network. This model can give a significant guidance for pyrolysis gasoline hydrogenation unit design and improvement, and the optimization results is more meaningful as the catalyst activity, product quality and hydrogen network integration are considered. The case study shows that this model clearly shows the effect of operation temperature variation on economic benefit at different time, optimal temperature can be identified according to the initial and improved temperature curves, and the service life of catalyst can be prolonged effectively and the economic benefit is increased by adjusting the temperature. In the proposed model, the numerical relationship between reactor temperature and operation time is represented by a linear fit, this will affect the accuracy of the optimization results. In the future, more accuracy relation should be deduced and applied. Furthermore, more aspects should be taken into consideration to calculate the economic benefit over reaction temperature. This will be studied in the future.

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