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Model of Steady State Cyclohexane Oxidation for Ketone-Alcohol (K-A) Oil Production

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Model of Steady State Cyclohexane Oxidation for Ketone-Alcohol (K-A) Oil Production

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Abstract

Cylohexane oxidation is of great industrial importance in the production of intermediates for the manufacture of nylon-6 and nylon-6,6. Most cyclohexane is commercially converted into a cyclohexanone–cyclohexanol mixture (known as K-A oil) by catalytic oxidation. K-A (Ketone-Alcohol) oil is then used as a raw material for adipic acid and caprolactam production. Practically, if the cyclohexanol content of KA oil is higher than that of cyclohexanone, it is more profitable to convert it into adipic acid; otherwise caprolactam production is more likely. The steady state cyclohexane oxidation reaction model in a stirred tank reactor for K-A oil production is presented and solved in this paper. The model was derived based on the mass balance and mass transfer equations using the kinetic equation. The set of algebraic equations was solved using non linear programming. The advantage of this method is that the relationship among variables can be better understood and an appropriate solution to the equation set can be obtained more quickly. Simulation results are particularly useful for process design such as in determining reactor dimensions and operating conditions.

Abstrak

Model Oksidasi Sikloheksana Tunak untuk Produksi Minyak Ketone-Alcohol (K-A). Oksidasi sikloheksana adalah salah satu reaksi penting dalam industri petrokimia dimana produk utamanya adalah senyawa intermediate untuk produksi nylon-6 dan nylon-6,6. Sebagian besar hasil produksi sikloheksana dikonversi secara komersial menjadi campuran sikloheksanon dan sikloheksanol (dikenal dengan nama minyak K-A) dengan oksidasi katalitik. Minyak K-A adalah bahan baku untuk produksi asam adipat dan kaprolaktam. Bila kandungan sikloheksanol dalam minyak K-A lebih tinggi daripada sikloheksanon, maka akan lebih menguntungkan kalau dikonversi menjadi asam adipat. Bila sebaliknya, maka konversi lebih lanjut menjadi kaprolaktam akan lebih disukai. Model reaksi oksidasi sikloheksana kondisi tunak dalam reaktor tangki berpengaduk telah dikembangkan dan diselesaikan dalam penelitian ini. Model tersebut didasarkan pada neraca massa dan perpindahan massa dengan memanfaatkan persamaan kinetika. Model dalam persamaan aljabar non-linear kemudian diselesaikan dengan metode optimisasi pemrograman non-linear. Keuntungan metode ini yaitu bahwa hubungan antar variabel dapat lebih dipahami dan penyelesaian terhadap model akan lebih cepat diperoleh. Manfaat hasil penelitian dapat dimanfaatkan untuk tahapan perancangan proses seperti penentuan dimensi reaktor maupun penentuan kondisi operasi.

Keywords: cyclohexane, non linear programming, oxidation optimization

1. Introduction

Many organic chemicals are produced from the oxidation reaction of hydrocarbon. Most of the industrial interests are carried out at pressure of 10-30 bar and temperatures of 353-523 K [1]. The liquid oxidation of cyclohexane is one example of the hydrocarbon oxidation reaction. This reaction is very important as it produces cyclohexanol and cyclohexanone as raw materials for the nylon-6 and nylon-6,6 industries. However the oxidation reaction of cyclohexane is very complex where cyclohexanol and cyclohexanone are intermediates of the reaction scheme. Several reaction schemes and kinetics, both catalytic and autocatalytic, have been proposed previously [1-6].

Understanding this oxidation process is very important for economical plant operations and to ensure adequate safety procedures, which are necessary due to its high temperature and pressure characteristic. The Flixborough disaster [7] in a cyclohexane oxidation plant in the United Kingdom presents an excellent example for why this process should be designed and operated carefully.

The cyclohexane-oxidation-reaction scheme is illustrated in Figure 1, based on the general hydrocarbon oxidation reaction.

The oxidation of cyclohexane is carried out at elevated temperatures and pressures and is usually catalyzed homogeneously under mild conditions using soluble catalysts, such as cobalt octoate/oleate/naphthenate. The conversion is limited to 4-15% to promote K-A yield in the range 85-90% [6].

There have been numerous studies on the reaction mechanisms of cyclohexane oxidation. In this work, the reaction scheme and kinetics of Alagy *et al*. [1] were adopted. The reaction scheme is described in Figure 2.

Figure 2 illustrates the great complexity of the reaction. The ratios of kinetic parameters (k_2/k_1, k_3/k_1 and k_4/k_1) were obtained by Alagy *et al.* [1]. The ratios showed that the oxidation of cyclohexanol and cyclohexanone was faster than that of cyclohexane. Therefore, further cyclohexane conversion may lead to the formation of by-products.

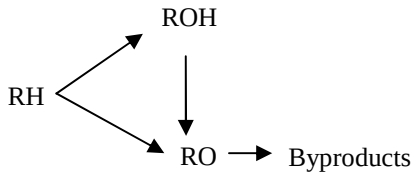

Figure 1. Cyclohexane Oxidation Scheme in General

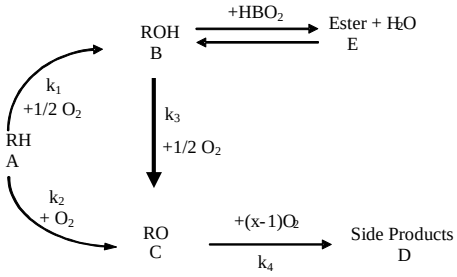

Figure 2. Cyclohexane Oxidation Mechanism (A: cyclo-hexane, B: cyclohexanol, C: cyclohexanone, D: byproduct, E: ester) [1]

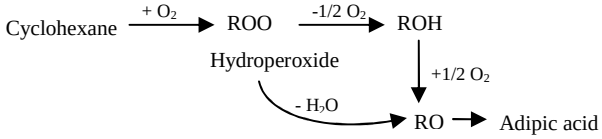

Figure 3. Cyclohexane Oxidation Mechanism Proposed by Spielman [9]

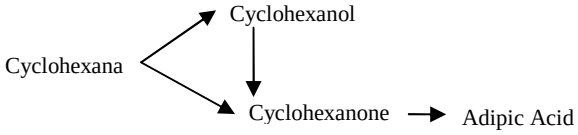

Figure 4. Cyclohexane Oxidation with Cobalt Acetate

This scheme and its kinetic parameters (shown in Table 1) were specifically valid for boric acid promoted processes.

A mechanism proposed by Spielman [9] is shown in Figure 3, while Suresh *et al.* [2] presented the autocatalytic oxidation of cyclohexane which include initiation, propagation and termination steps as follows:

Initiation:

$$RH \rightarrow R'^*$$

$$PH \rightarrow R'^*$$

Propagation:

$$R'^* + O_2 \xrightarrow{kp} R'O_2^*$$

$$R'O_2^* + RH \xrightarrow{kp1} PH + R'^*$$

$$R'O_2^* + PH \xrightarrow{kp2} PH + R'^*$$

Termination:

$$R'^* \rightarrow PH$$

$$R'O_2^* \rightarrow PH$$

Rao [10] proposed a reaction scheme (Figure 4) and kinetic model of cyclohexane oxidation for acetic acid and cobalt acetate as catalysts.

Significant industrial demand for K-A oil in recent years has stimulated various studies aiming to find milder, lower cost, and more environmentally friendly conditions; namely, to find greener and more efficient catalytic processes for the classic industrial method [11-15].

The aim of this research was to develop and solve the mathematical model of cyclohexane oxidation in single stirred tank reactor as a basis for further investigation in a series of reactors. The model was then simulated to study the effects of the gas-to-liquid molar-rate ratio (R_G), impeller speed (N_p), and space-time (θ) on the performance of the single reactor

2. Methods

Cyclohexane oxidation in a single reactor. The system studied in this paper is described in Figure 5. Cyclohexane in the form of liquid and air are introduced to the reactor which operate at 9.5 atm and 167 °C. The gas exiting the reactor is passed through condenser, where cyclohexane vapor condenses and is recycled back to the reactor. The other components such as CO, CO_2, water vapor, oxygen, and nitrogen exit in the blow-down gas stream.

The operating conditions of the reactor are shown in Table 1.

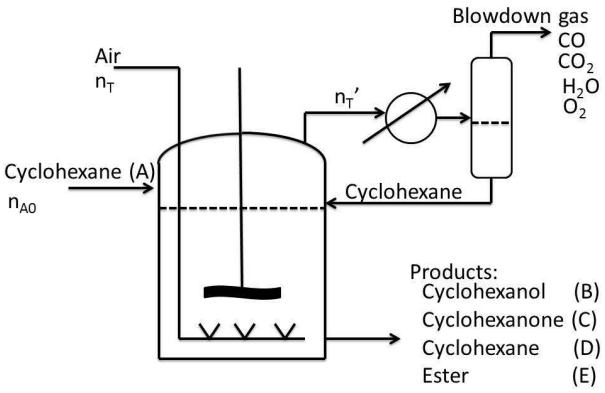

Figure 5. Cyclohexane Oxidation in Single Reactor

Table 1. Operating Condition and Parameter Values

	Values [unit]
Operating Conditions	
Pressure, P	9.5 atm
Temperature, T	167 °C
Reactor diameter, D	2 m
Impeller type	Disk Turbine
Impeller diameter, Da	0.667 m
Kinetic Parameters	
k_1	23 (cc/mol.sec)
$\alpha(k_2/k_1)$	0.2
β (k_3/k_1)	19
γ (k_4/k_1)	32.75
λ	0.6
σ	2.7
τ	14.57

Mathematical model. The mass balance equations which represented the balance for each component in the reaction system (cyclohexane, cyclohexanol, cyclohexanone, by-product, oxygen, water, CO, and CO_2) were developed. This resulted in the non linear algebraic equation system as follows:

$$1 - N_A = \frac{1}{G}(1+\alpha)N_A \tag{1}$$

$$N_B = \frac{\lambda P_{H2O}}{(1+\lambda P_{H2O})}\frac{1}{G}(N_A - \beta N_B) \tag{2}$$

$$N_C = \frac{1}{G}(\alpha N_A + \beta N_B - \gamma N_C) \tag{3}$$

$$\frac{P_{H2O}\,0.79R_G}{[(P-P_A)(1-\eta-\varphi)-P_{H2O}]} = \frac{1}{G}\left[\alpha N_A + \beta N_B + \gamma N_C + \frac{N_A - \beta N_B}{1+\lambda P_{H2O}}\right] \tag{4}$$

$$\eta = \frac{N_D[(P-P_A)(1-\eta-\varphi)-P_{H2O}]}{0.79R_G(P-P_A)} \tag{5}$$

$$\varphi = \frac{\left(0.21 - \frac{K}{G.R_G}\right)}{0.79(P-P_A)} \times [(P-P_A)(1-\eta-\varphi)-P_{H2O}] \tag{6}$$

$$K = (0.5+\alpha)N_A + 0.5\beta N_B + (x_1 - 1)\gamma N_C \tag{7}$$

$$G = \frac{K}{0.21R_G} + \left(\frac{K\rho_L}{k_L a} + \frac{1}{k_1}\right)\frac{He'}{\theta} \times \frac{0.79P}{0.21[(P-P_A)(1-\eta-\varphi)-P_{H2O}]} \tag{8}$$

$$N_D = \frac{1}{G}\gamma N_C \tag{9}$$

$$N_E = \frac{1}{G}\frac{(N_A - \beta N_B)}{(1+\lambda P_{H2O})} \tag{10}$$

where:

N_A = Ratio of cyclohexane molar rate in the product stream to that of cyclohexane in the feed stream.

N_B = Ratio of cyclohexanol molar rate in the product stream to that of cyclohexane in the feed stream.

N_C = Ratio of cyclohexanone molar rate in the product stream to that of cyclohexane in the feed stream.

P_{H2O} = Water partial pressure in the gas exiting the reactor.

P_A = Cyclohexane vapor pressure at the reactor temperature.

φ = Oxygen mole fraction in the blow down gas stream.

η = CO and CO_2 mole fraction in the blow down gas stream.

N_D = Ratio of byproducts molar rate in the product stream to that of cyclohexane in the feed stream.

N_E = Ratio of cyclohexanone molar rate in the product stream to that of cyclohexane in the feed stream

K and *G* are defined as follows:

$$K \equiv \frac{D}{k_1 n_{A0}} \tag{11}$$

$$G \equiv \frac{1}{[O_2]_L \theta k_1} \tag{12}$$

The mass transfer coefficient correlation used here is the Hickman equation [8]:

$$k_L a = 0.046\left(\frac{P_g}{V}\right)^{0.54} V_s^{0.68} \quad (13)$$

where:

$$\frac{P_g}{V} = 706.3 N_p^{3.01} Q^{-0.45}\left(\frac{Da}{D}\right)^{5.38} \quad (14)$$

$k_L a$ = Liquid side mass transfer coefficient, sec^{-1}

P_g = Gassed power, Watt

N_p = Impeller speed, rps

Q = Volumetric gas flowrate, m^3/sec

Da = Impeller diameter, m

D = Tank diameter, m

V = Liquid volume, m^3

V_s = Gas superfisial velocity, m/s

The data needed to solve the equations above are:

$$He' = \frac{He}{\rho_G} \quad (15)$$

$$P_A = 11.43 - 0.1826T + 0.00095T^2 \quad (16)$$

where:

He = Henry's constant for oxygen in cyclohexane

ρ_G = Gas density, mol/cm^3

T = Temperature, °C

The overall performance of the oxidation such as conversion, selectivity and the alcohol-ketone ratio are calculated according to the following equations:

$$Conversion = \frac{n_{Ao} - N_A}{n_{Ao}} \quad (17)$$

$$Selectivity = \frac{N_B + N_E + N_C}{n_{Ao} - N_A} \quad (18)$$

$$Alcohol - Ketone \quad Ratio = \frac{N_B + N_E}{N_C} \quad (19)$$

Solution Procedure. If the right sides of equations (1) to (10) are brought to the left side, then there are 10 equations with zero values ($f_i = 0$) and 10 variables that should be obtained (i.e. $N_A, N_B, N_C, P_{H2O}, \varphi, \eta, K, G, N_D, N_E$).

This set of algebraic equations can be solved by considering equation (1) to (10) as the equalities constraints for the optimization problem with the following objective function:

$$\underset{x}{Min}\left[abs(f_4) + abs(f_7) + abs(f_9) + abs(f_{10})\right] \quad (20)$$

The above objective function was chosen because it contains all the variables that must be found.

The following inequalities constraints must be employed:

$$0 \le N_A \le 1 \quad (21)$$

$$0 \le N_B \le 1 \quad (22)$$

$$0 \le N_C \le 1 \quad (23)$$

$$0 \le N_D \le 1 \quad (24)$$

$$0 \le N_E \le 1 \quad (25)$$

Other inequalities constraints must be employed appropriately to obtain feasible solution (i.e. all the resulting variables are positive, real numerical values).

3. Results and Discussion

Figure 6 shows the effect of the gas-to-liquid molar-rate ratio (R_G) on the oxidation performance (conversion, selectivity, and alcohol-ketone ratio). The conversion and alcohol-ketone ratio increased with the gas-to-liquid molar-rate ratio, while the selectivity decreased. According to equations (13) and (14) the increase in the gas molar rate (Q) will increase the value of the liquid mass transfer coefficient, which will then enhance the dissolved oxygen concentration in the liquid phase.

The cyclohexanol-to-cyclohexanone ratio could also increase with higher dissolved oxygen concentration because of the faster by-product formation rate compared to that of cyclohexanone ($k_4 > k_2$) and ($k_4 > k_3$). This means that the oxidation of cyclohexanone is faster than its formation from cyclohexanol and cyclohexane. Despite that, the oxidation of cyclohexane into cyclo-hexanol was faster than into cyclohexanone ($k_1 > k_2$) and the equlibrium esterification with boric acid could shift the esterification toward the right when the cyclohexanol formed. The selectivity decreased because the higher oxygen concentration in the liquid phase favored the oxidation reaction of cyclohexanol into cyclohexanone and cyclohexanone into by-products. It also favored the CO and CO_2 formation in the oxidation reaction.

Figure 7 shows that the impeller speed increased the cyclohexane conversion but decreased the selectivity of cyclohexanol and cyclohexanol. The impeller speed could

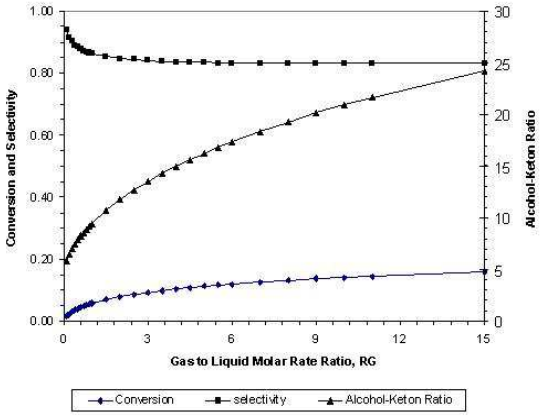

Figure 6. The Effect of R_G on the Oxidation Performance at Impeller Speed = 70 rpm, and θ=3600 sec

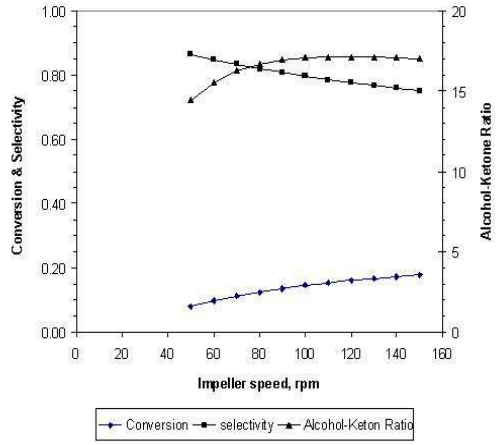

Figure 7. Effect of Impeller Speed, R_G = 5, and θ =3600 s.

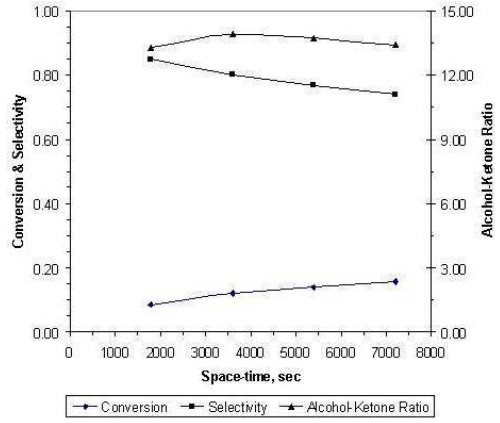

Figure 8. Effect of Space-Time at R_G = 5, and Impeller Speed =100 rpm

could also increase the mass transfer coefficient, and the effects of this increase were the same as increasing the gas molar flow rate.

Figure 8 shows the effect of space-time on the conversion, selectivity and alcohol-to-ketone ratio. The conversion of cyclohexane increased as the space-time increased due to longer reaction time. However, this higher conversion could decrease the selectivity. The alcohol-to-ketone ratio was relatively constant with the increase of space-time.

4. Conclusions

The mathematical model of cyclohexane oxidation in a single stirred tank reactor has been developed and solved using constrained non-linear programming. This model allows further investigation of the effect of process operating condition on the performance of the reactor. The effect of the gas-to-liquid molar-rate ratio (R_G), impeller speed (N_p) and space-time (θ) on the performance of the reactor (conversion, selectivity and alcohol-ketone ratio) are presented. The resulting model and solution procedure, which was derived for single reactor, can be readily extended for reactor train in series arrangement.

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