
COMPARISON OF SEVERAL MODELS TO ASSESS THE EFFECT OF MICROMIXING PHENOMENA ON THE YIELD OF COMPLEX CHEMICAL REACTIONS IN STIRRED TANK

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Abstract

The effect of micromixing phenomena on the course of chemical reactions occurring in continuous stirred tank reactor was very important especially for fast reactions case. Previous workers have analyzed the effect using several models. The aim of this work is to make comparative study of several models to assess the effect of micro mixing phenomena on the course of two competitive fast parallel chemical reactions carried out in continuous stirred tank reactors. The reaction system investigated was the same as that studied by Baldyga (2001), neutralization of sodium hydroxide and hydrolysis of ethyl chloroacetate. Two closure approaches based on CFD available in FLUENT, Generalized Finite Rate Formulation (GFRF) and Probability Density Function (PDF), and a mechanistic model, Packet Diffusion Model, have been compared in assessing the effect of turbulent mixing on the yield of reaction. Baldyga's experimental data have been used to validate the computational results. The study concludes that a simple mechanistic model, Packet Diffusion Model, can predict fairly well (comparable in accuracy to the more sophisticated model such as PDF and GFRF model) the effect of micromixing phenomena on the course of parallel competitive chemical reactions in the continuous stirred tank reactor.

Key Words : Micromixing, Continuous Stirred Tank Reactor, CFD, PDM, GFRF, PDF

Abstrak

Pengaruh fenomena micromixing terhadap berlangsungnya reaksi kimia yang terjadi di dalam reaktor kimia tangki berpengaduk menjadi sangat penting terutama untuk kasus reaksi-reaksi cepat. Beberapa penelitian terdahulu telah menganalisa secara teoritis pengaruh fenomena ini terhadap kinerja reaktor tangki teraduk menggunakan beberapa model. Tujuan penelitian ini adalah untuk membandingkan beberapa model untuk menganalisa pengaruh fenomena micromixing pada jalannya dua reaksi paralel kompetitif cepat yang terjadi di dalam reaktor alir tangki teraduk menggunakan beberapa model. Sistem reaktor yang diselidiki adalah sama dengan yang dipelajari Baldyga (2001), yaitu reaksi netralisasi sodium hidroksida dan hidrolisa ethyl chloroacetat. Dua pendekatan closure berbasis CFD yang tersedia pada FLUENT, yaitu Generalized Finite Rate Formulation (GFRF) dan Probability Density Function (PDF), dan suatu model mekanistik, yaitu Packet Diffusion Model, telah dibandingkan dalam menganalisa pengaruh pencampuran turbulen pada yield reaksi. Data eksperimen Baldyga telah digunakan untuk mengvalidasi hasil-hasil perhitungan. Penelitian ini menyimpulkan bahwa model mekanistik yang sederhana, yaitu Packet Diffusion Model, dapat memprediksi cukup baik, sebanding dari segi ketelitiannya dengan model berbasis CFD yang lebih rumit, pengaruh fenomena micromixing terhadap jalannya dua reaksi paralel kompetitif di dalam reaktor tangki teraduk.

Kata Kunci : Micromixing, Reaktor Alir Tangki Teraduk, CFD, PDM, GFRF, PDF

1. Introduction

Stirred tanks have been widely used in many chemicals, pharmacologicals and oil refinery industries for mixing and chemical reactions such as producing specialty chemicals, medicines, polymer and for neutralization and crystallization process. Generally the processes comprise multiple reactions. For multiple reactions the distribution of reaction products become an important reactor performance indicator. The role of agitation generated by impeller is to develop turbulent flow field in order to minimize temperature and concentration gradient, and to enhance the contact among the reacting species. Transformation of the reacting species occurred at molecular scale, therefore this process depend on the contact among the reacting species and next, the contact was influenced by mixing phenomena in molecular scale called micromixing. Micromixing was becoming an important factor especially for multiple reactions, which is relatively faster than mixing rate. In this case, micromixing phenomena affect reaction product distribution. The effect of mixing phenomena on reactor performance was also observed in bioreactor where microorganism was grown. Several methods and equipments have been developed to create high mixing quality. However, there is no single mixing device, which is suitable for all processes. Therefore comprehensive research work was needed to study the effect of mixing on the course of multiple reactions in stirred tank.

Many research works have been carried out to study experimentally the effect of the mixing on multiple reaction in stirred tank (Fournier et. al., 1996; Guichardon et. al., 2001; Assirelli et. al., 2002; Baldyga, et.al., 2001). These studies developed chemical test to analyze the effect of mixing on chemical reaction in stirred tank and to apply the test procedure to study the effect of process variables relating with mixing phenomena on reactor performance. The weakness of the experimental research is that the result of study cannot be used for other tank geometry. A turbulent mixing model was required to evaluate the effect of mixing phenomena on the course of chemical reaction in the tank. Several researchers have carried theoretical studies developing micro mixing phenomena modeling to predict the effect of turbulent mixing phenomena on the course of chemical reaction in stirred tank (Villermaux et. al., 1992; Baldyga, 1994; Baldyga et. al., 2001; Bourne & Yu, 1994; and Fournier et. Al., 1996). Most of these research works were carried out batch and semi batch wise. Practically, continuous stirred tank reactor systems were widely operated

in process industries comprising fast multiple chemical reactions. Yield and selectivity of these reactions will be affected by turbulent mixing. It is the aim of the present research to study the effect of turbulent mixing on the selectivity and yield of product of fast competitive parallel chemical reactions in continuous stirred tank reactor system. The effect of impeller rotation speed and mean residence time on product distribution was studied in this work using theoretical approach with several models.

2. Fundamental

Basically there are two approaches to assess the effect of turbulent mixing on the course of chemical reaction in the chemical reactor system: Closure approach based on CFD and phenomenological approach one of which is Packet Diffusion Model. For the Closure approach, the equation to be solved is: time average continuity and momentum equations with turbulent model as closure. Besides these equations we have to consider the flow generated by impeller to model the movement of fluid in the stirred tank. There are several method to handle this impeller problem: Multiple Reference Frame (MRF) method and Black box method. The flow system with chemical reaction needs additional model one of which is Generalized Finite Rate Formulation. The equation to be solved was the continuity equation for each species,

$$\frac{\partial}{\partial t}(\rho m_{i'}) + \nabla \cdot (\rho \bar{u} m_{i'}) = -\nabla \cdot \bar{J}_{i'} + R_{i'} + S_{i'} \quad (1)$$

Where $J_{i',i}$ is diffusion flux of species i' which can be expressed as flows for turbulent flow,

$$J_{i',i} = -(\rho D_{i',m} + \frac{\mu_t}{Sc_t}) \nabla m_{i'} \quad (2)$$

$R_{i'}$ is generation term caused by chemical reaction, while $S_{i'}$ is generation term caused by other sources. $R_{i'}$ can be predicted by considering the effect of mixing phenomena on the course of chemical reaction. Using Generalized Finite Formulation Model, molar rate of formation/consumption of species i' in reaction k was given by the following equation,

$$R_{i',k} = (v_{i',k}^n - v_{i',k}^r) \left(k_{f,k} \prod_{j=1}^N [C_{j'}]^{\eta_{j',k}} - k_{b,k} \prod_{j=1}^N [C_{j'}]^{\eta_{j',k}} \right) \quad (3)$$

And the effect of turbulence on the reaction rate was calculated using Magnussen and Hjertager model. Using this model, reaction rate $R_{i',k}$ was

given by smaller value from the following equations,

$$R_{i',k} = v_{i',k} M_{i'} AB\rho \frac{\varepsilon}{k} \frac{m_R}{v_{R,k} M_R} \quad (4)$$

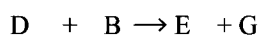
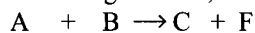
$$R_{i',k} = v_{i',k} M_{i'} AB\rho \frac{\varepsilon}{k} \frac{\sum_P m_P}{\sum_{j'} v_{j',k} M_{j'}} \quad (5)$$

The generation term in Equation (1) was obtained as follows,

$$R_{i'} = \sum_k R_{i',k} \quad (6)$$

Where $R_{i',k}$ was obtained from the smaller value that is predicted from Arrhenius equation and that is predicted by Magnussen and Hjertager model.

Packet diffusion model was first introduced by Nauman (1975). In the application of Packet Diffusion Model, we consider the following reaction,



Where A=HCl, B=NaOH, C= NaCl, D=CH₂ClCOOC₂H₅, E= CH₂ClCOONa, F=H₂O, dan G=C₂H₅OH. The diffusion equation for packet I which is rich in A and packet II which is rich in B were as follows,

Packet I:

$$\frac{\partial \bar{C}_A^I}{\partial \tau} = \frac{1}{\pi^2 Sg_A} \left[\frac{\partial^2 \bar{C}_A^I}{\partial r^2} + \frac{2}{r} \frac{\partial \bar{C}_A^I}{\partial r} \right] - k_1 \bar{C}_A^I \bar{C}_B^I \quad (7)$$

$$\frac{\partial \bar{C}_B^I}{\partial \tau} = \frac{1}{\pi^2 Sg_B} \left[\frac{\partial^2 \bar{C}_B^I}{\partial r^2} + \frac{2}{r} \frac{\partial \bar{C}_B^I}{\partial r} \right] - k_1 \bar{C}_A^I \bar{C}_B^I - k_2 \bar{C}_A^I \bar{C}_D^I \quad (8)$$

$$\frac{\partial \bar{C}_D^I}{\partial \tau} = \frac{1}{\pi^2 Sg_D} \left[\frac{\partial^2 \bar{C}_D^I}{\partial r^2} + \frac{2}{r} \frac{\partial \bar{C}_D^I}{\partial r} \right] - k_2 \bar{C}_A^I \bar{C}_D^I \quad (9)$$

$$\frac{\partial \bar{C}_E^I}{\partial \tau} = \frac{1}{\pi^2 Sg_E} \left[\frac{\partial^2 \bar{C}_E^I}{\partial r^2} + \frac{2}{r} \frac{\partial \bar{C}_E^I}{\partial r} \right] + k_2 \bar{C}_A^I \bar{C}_D^I \quad (10)$$

$$\frac{\partial \bar{C}_F^I}{\partial \tau} = \frac{1}{\pi^2 Sg_F} \left[\frac{\partial^2 \bar{C}_F^I}{\partial r^2} + \frac{2}{r} \frac{\partial \bar{C}_F^I}{\partial r} \right] + k_1 \bar{C}_A^I \bar{C}_B^I \quad (11)$$

The initial conditions were,

$$\bar{C}_A^I(r,0)=1; \bar{C}_B^I(r,0)=0; \bar{C}_D^I(r,0)=C_{D,In}/C_{A,In}; \bar{C}_E^I(r,0)=0 \quad (12)$$

and the boundary conditions were,

$$\bar{C}_i^I(1,\tau) = C_{i,mm}/C_{A,In} \quad (i=A,B,D,E) \quad (13)$$

$$\text{At } r=0 \rightarrow \frac{\partial \bar{C}_i^I}{\partial r} = 0 \quad (i=A,B,D,E) \quad (14)$$

Where,

$$\bar{C}_i^I = C_i^I / C_{A,In} \quad (i=A,B,D,E) \quad (15)$$

Packet II:

$$\frac{\partial \bar{C}_A^{II}}{\partial \tau} = \frac{1}{\pi^2 Sg_A} \left[\frac{\partial^2 \bar{C}_A^{II}}{\partial r^2} + \frac{2}{r} \frac{\partial \bar{C}_A^{II}}{\partial r} \right] - k_1 \bar{C}_A^{II} \bar{C}_B^{II} \quad (16)$$

$$\frac{\partial \bar{C}_B^{II}}{\partial \tau} = \frac{1}{\pi^2 Sg_B} \left[\frac{\partial^2 \bar{C}_B^{II}}{\partial r^2} + \frac{2}{r} \frac{\partial \bar{C}_B^{II}}{\partial r} \right] - k_1 \bar{C}_A^{II} \bar{C}_B^{II} - k_2 \bar{C}_A^{II} \bar{C}_D^{II} \quad (17)$$

$$\frac{\partial \bar{C}_D^{II}}{\partial \tau} = \frac{1}{\pi^2 Sg_D} \left[\frac{\partial^2 \bar{C}_D^{II}}{\partial r^2} + \frac{2}{r} \frac{\partial \bar{C}_D^{II}}{\partial r} \right] - k_2 \bar{C}_A^{II} \bar{C}_D^{II} \quad (18)$$

$$\frac{\partial \bar{C}_E^{II}}{\partial \tau} = \frac{1}{\pi^2 Sg_E} \left[\frac{\partial^2 \bar{C}_E^{II}}{\partial r^2} + \frac{2}{r} \frac{\partial \bar{C}_E^{II}}{\partial r} \right] + k_2 \bar{C}_A^{II} \bar{C}_D^{II} \quad (19)$$

The initial conditions were,

$$\bar{C}_A^{\text{II}}(r,0)=0; \bar{C}_B^{\text{II}}(r,0)=1; \bar{C}_D^{\text{II}}(r,0)=0; \bar{C}_E^{\text{II}}(r,0)=0 \quad (19)$$

And the boundary conditions were,

$$\bar{C}_k^{\text{II}}(1,\tau) = C_{i\text{mm}} / C_{B,\text{In}} \quad (i = A, B, D, E) \quad (20)$$

$$\text{At } \bar{r}=0 \rightarrow \frac{\partial \bar{C}_A^{\text{II}}}{\partial \bar{r}}=0; \frac{\partial \bar{C}_B^{\text{II}}}{\partial \bar{r}}=0; \frac{\partial \bar{C}_D^{\text{II}}}{\partial \bar{r}}=0; \frac{\partial \bar{C}_E^{\text{II}}}{\partial \bar{r}}=0 \quad (21)$$

$$\bar{C}_i^{\text{II}} = C_i^{\text{II}} / C_{A,\text{In}} \quad (i = A, B, D, E) \quad (22)$$

Note that these boundary conditions contain quantities: $C_{A\text{mm}}, C_{B\text{mm}}, C_{D\text{mm}}$ and $C_{E\text{mm}}$ which is the concentration of A, B, D, and E in maximum mixedness reactor. For the degree of macromixing which is equivalent to one mixed flow reactor, the condition of maximum mixedness is the same as perfect mixer. In this case, $C_{A\text{mm}}, C_{B\text{mm}}, C_{D\text{mm}}$ and $C_{E\text{mm}}$ were calculated from component material balance in the reactor,

$$qC_{A,\text{In}} - qC_{A\text{mm}} - k_1 C_{A\text{mm}} C_{B\text{mm}} V = 0 \quad (23)$$

$$qC_{B,\text{In}} - qC_{B\text{mm}} - k_1 C_{A\text{mm}} C_{B\text{mm}} - k_2 C_{D\text{mm}} C_{B\text{mm}} V = 0 \quad (24)$$

$$qC_{D,\text{In}} - qC_{D\text{mm}} - k_2 C_{D\text{mm}} C_{B\text{mm}} V = 0 \quad (25)$$

$$qC_{E,\text{In}} - qC_{E\text{mm}} + k_2 C_{D\text{mm}} C_{B\text{mm}} V = 0 \quad (26)$$

To simplify the computation, we used the concept of reaction progress. In this case, we define reaction progress for the first and second reaction, ϵ_1 and ϵ_2 , as moles of B component reacted per second for the first reaction and the second reaction respectively. Using these quantities, Equation (23) and (25) can be written as follows,

$$\epsilon_1 - k_1 \left[C_{A,\text{In}} \frac{\epsilon_1}{q} \right] \left[C_{B,\text{In}} \frac{\epsilon_1 + \epsilon_2}{q} \right] V = 0 \quad (27)$$

$$\epsilon_2 - k_2 \left[C_{D,\text{In}} \frac{\epsilon_2}{q} \right] \left[C_{B,\text{In}} \frac{\epsilon_1 + \epsilon_2}{q} \right] V = 0 \quad (28)$$

By making the equations to be dimensionless, we obtain,

$$E_1 - \kappa_1 [1 - E_1] [MB - E_1 - E_2] = 0 \quad (29)$$

$$E_2 - \kappa_2 [MD - E_2] [MB - E_1 - E_2] = 0 \quad (30)$$

where :

$$E_1 = \frac{\epsilon_1}{qC_{A,\text{In}}}, E_2 = \frac{\epsilon_2}{qC_{A,\text{In}}}, MB = \frac{C_{B,\text{In}}}{C_{A,\text{In}}}, MD = \frac{C_{D,\text{In}}}{C_{A,\text{In}}}$$

$$\kappa_1 = k_1 t C_{A,\text{In}}, \kappa_2 = k_2 t C_{A,\text{In}} \quad (31)$$

Equation (29) and (30) can be solved numerically to obtain E_1 and E_2 . Finally $C_{A\text{mm}}, C_{B\text{mm}}, C_{D\text{mm}}$ and $C_{E\text{mm}}$ can be obtained as follows,

$$C_{A\text{mm}} = C_{A,\text{In}} [1 - E_1] \quad (32)$$

$$C_{B\text{mm}} = C_{A,\text{In}} [MB - E_1 - E_2] \quad (33)$$

$$C_{D\text{mm}} = C_{A,\text{In}} [MD - E_2] \quad (34)$$

$$C_{E\text{mm}} = C_{A,\text{In}} E_2 \quad (35)$$

3. Methodology

This work was carried out using computational approach. We applied two different methods: Closure method based on CFD using Generalized Finite Rate Formulation (GFRF) and Phenomenological method using a mechanistic model, Packet Diffusion Model (PDM). For the Closure Approach we used CFD commercial code FLUENT sixth version, k- model was used to characterize the turbulent behavior of the flow in the tank and MRF method was used to handle the impeller problem of the flow system. The governing equations were solved using finite volume method with power law differencing scheme, and we used SIMPLE method to handle pressure-velocity coupling problem. For phenomenological approach we developed our own program. In this case successive approximation method was used to solve equations

(29) and (30). The system of partial differential equations with appropriate initial and boundary condition that have been developed for packet I and II were solved using the combination of Runge-Kutta and Finite Difference method with time increment $\Delta\tau=0.0000005$ and radial distance increment $\Delta r=0.05$. The solutions give the concentration of components in packet I and packet II as a function of radial position and time. The average concentration of components in packet I and II as function of time was obtained as follows,

$$\overline{C_k^I}(\tau) = \frac{1}{0} \int_0^1 \overline{C_k^I}(\tau, r) 4\pi r^{-2} dr \quad (36)$$

$$\overline{C_k^{II}}(\tau) = \frac{1}{0} \int_0^1 \overline{C_k^{II}}(\tau, r) 4\pi r^{-2} dr \quad (37)$$

And the concentration of components in packet I and II in reactor effluent was determined from Equation (38) and (39).

$$C_{k,OUT}^I = C_{A,IN} \int_0^\infty \overline{C_k^I}(\tau) f(\tau) d\tau \quad (38)$$

$$C_{k,OUT}^{II} = C_{B,IN} \int_0^\infty \overline{C_k^{II}}(\tau) f(\tau) d\tau \quad (39)$$

The integration in Equations (36) through (39) were done numerically using quadrature method. The concentration of components in the reactor effluent was obtained as follows,

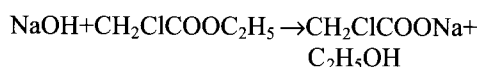
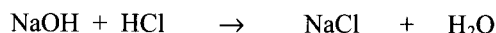
$$C_{k,OUT} = \frac{q_I}{q_I + q_{II}} C_{k,OUT}^I + \frac{q_{II}}{q_I + q_{II}} C_{k,OUT}^{II} \quad (40)$$

$$X = \frac{C_{B,IN}^M - C_{B,OUT}^M}{C_{B,IN}^M} \quad (41)$$

$$S = \frac{C_{P,OUT}^M}{C_{B,IN}^M} \quad (42)$$

The simulation results using these different methods were compared. The comparison was also made with Baldyga's simulation results using PDF model available in FLUENT and Baldyga's experimental data.

System investigated consists of flat bottom cylindrical tank with double inlet pipe equipped with four baffles and six blades Rushton disc turbine. The height of the liquid in the tank is the same as the tank diameter. The clearance between the impeller and tank bottom was 1/3 tank diameter. The tank was used to study the effect of micro mixing on the following reaction system,



with reaction rate constant for the second reaction: $k_2 = 2.0 \cdot 10^5 \exp(-3,891 \cdot 10^4 / \text{RT})$. The first reaction was much faster than the second reaction. Natrium Hydroxide (1000 mol/m³) was fed into the tank through inlet pipe 0.001 m in diameter and mixture of hydrochloric acid and ethyl chloroacetate solution (20 mol/m³) was fed into the tank through 0.003 m diameter inlet pipe. The flow rate of Natrium Hydroxide solution was varied from 0.046 to 0.021 liter/minute and for the mixture of hydrochloric acid and ethyl chloroacetate solution, the flow rate was varied from 2.309 to 1.039 liter/minute corresponding to the mean residence time of fluid in the tank from 9 to 20 minutes.

4. Results and Discussion

In this work, the effect of micromixing phenomena on the course of parallel competitive reaction was analyzed by Closure approach (GFRF) using CFD commercial Code FLUENT and mechanistic model (PDM) with our own program written in PASCAL.

The simulation results using FLUENT was presented as contour of NaOH concentration in the tank shown in Figure 1. It can be shown that NaOH was concentrated in a zone around NaOH inlet pipe. There is no chance for Natrium hydroxide to be mixed homogeneously in the tank, however it was directly reacted with hydrochloric acid and ethyl chloroacetate in the region around the feed inlet. This fact is due to the chemical reaction rate, which is much faster than mixing rate. Figure 2 presented the simulation result showing effect of mean residence time and impeller rotation speed on the selectivity of Sodium ethyl chloroacetate. It is seen that the selectivity of sodium ethyl Chloroacetate increase with increasing mean residence time but decrease with increasing impeller rotation speed.

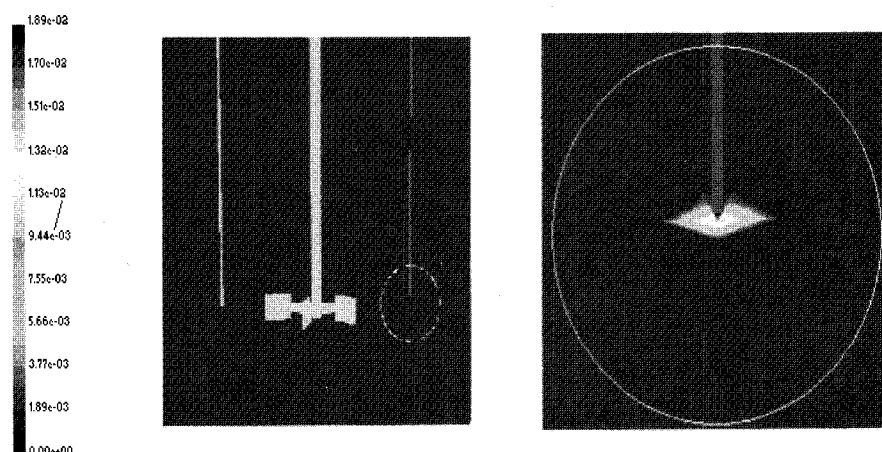


Figure 1. The Contour of NaOH Concentration in The Tank and Reaction Zone

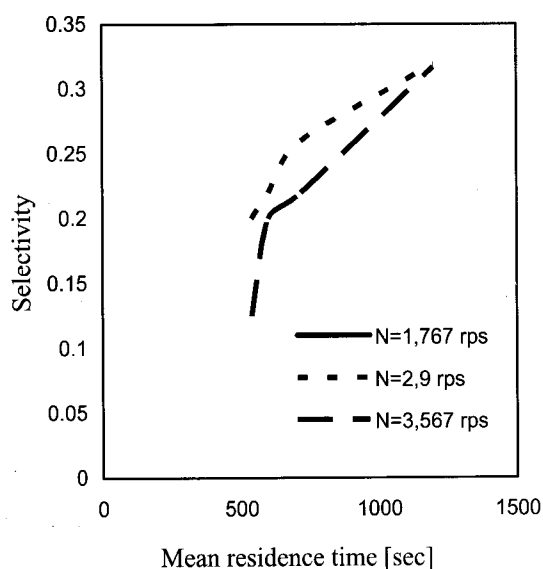


Figure 2. Relation between Mean Residence Time With the Selectivity of Sodium Ethyl Chloroacetate

Increasing the impeller rotation speed causes the flow of fluid in the tank more turbulent and increases the concentration of hydrochloric acid and ethyl chloroacetate in the reaction zone. This phenomena increases Arrhenius reaction rate and Turbulent reaction rate for each reaction. However the increase of the first reaction is higher than the second reaction, that is why the selectivity of sodium ethyl chloroacetate decrease with increasing impeller rotation speed. Increasing mean residence time or decreasing the feed flowrate causes the level of turbulence in the reaction zone decrease which in turn decreases the

reaction rate for each reaction. The decrease is more significant for the first reaction than for the second reaction, therefore the selectivity of sodium ethyl chloroacetate increase.

The selectivity of Sodium ethyl chloroacetate was also predicted using Packet Diffusion Model (PDM). Macromixing level of the reactor was expressed as the number of equivalent continuous stirred tank reactors. Figure 3 through Figure 5 show the effect of impeller rotation speed and mean residence time on the selectivity of Sodium ethyl chloroacetate using this model for the number of equivalent CSTR (NR) of 1,2, and 3 respectively. It is seen that the pattern of effect of mean residence time and impeller rotation speed shown by these figures is the same. Figure 6 depicts the effect of macro mixing level which is expressed as the number of equivalent CSTR on the selectivity of Sodium Ethyl Chloroacetate. It can be seen that NR=1 gives the lowest selectivity. In condition of long enough mean residence time (above 600 seconds), the selectivity for NR=2 and NR=3 almost the same. The trend for the effect of mean residence time on the selectivity predicted by this model is in opposite direction with that predicted by GFRF. Using GFRF, the selectivity of Sodium Ethyl Chloroacetate increase with increasing mean residence time, while using PDM, the selectivity decrease with increasing mean residence time. The simulation results were compared with the experimental data by Baldyga (2001) for impeller rotation speed of 1.767 rps shown in Figure 7. The prediction using PDM agree very well with Baldyga's experimental data and Baldyga's simulation data using PDF. However, the prediction using GFRF deviate significantly with

Baldyga's experimental data. Figure 8 presents the comparison between the prediction results and Baldyga's experimental data for the impeller rotation speed of 2.9 rps. This figure also shows that the prediction using GFRF gives the largest deviation compared to the experimental data. The prediction using PDF by Baldyga agree better with the experimental data than our prediction using PDM.

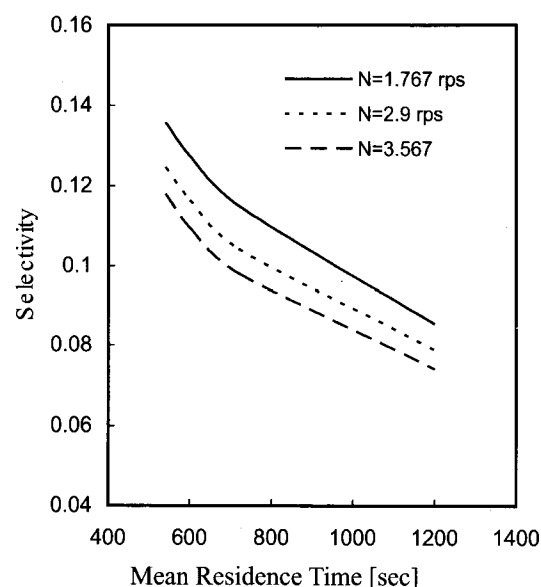


Figure 3. The Effect of Mean Residence Time and Impeller Rotation Speed on The Selectivity of Sodium Ethyl Chloroacetate Using Packet Diffusion Model for NR=1

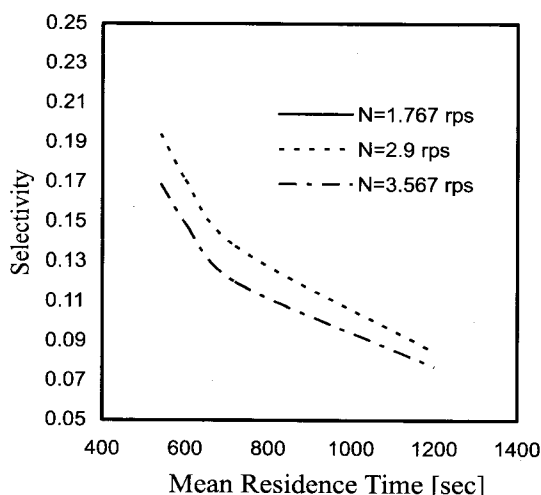


Figure 4. The Effect of Mean Residence Time and Impeller Rotation Speed on The Selectivity of Sodium Ethyl Chloroacetate Using Packet Diffusion Model for NR=2

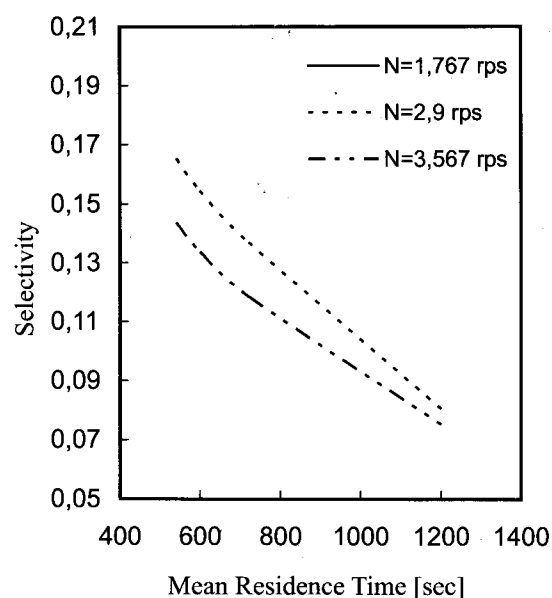


Figure 5. The Effect of Mean Residence Time and Impeller Rotation Speed on The Selectivity of Sodium Ethyl Chloroacetate Using PDM for NR=3

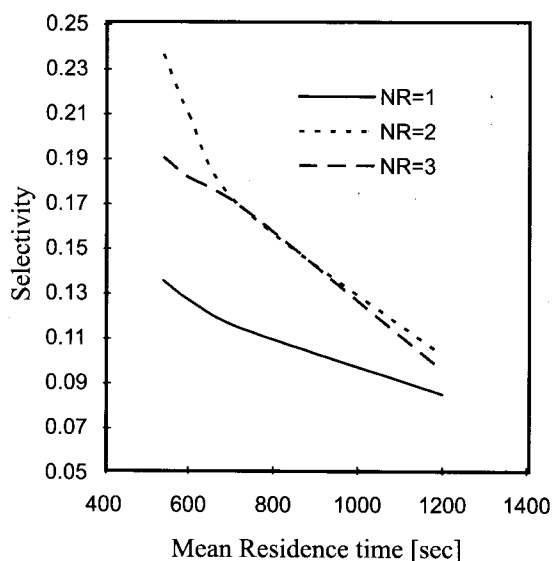


Figure 6. The Effect of Macromixing (Represented as The Number of Equivalent CSTR) on The Selectivity of Sodium Ethyl Chloroacetate

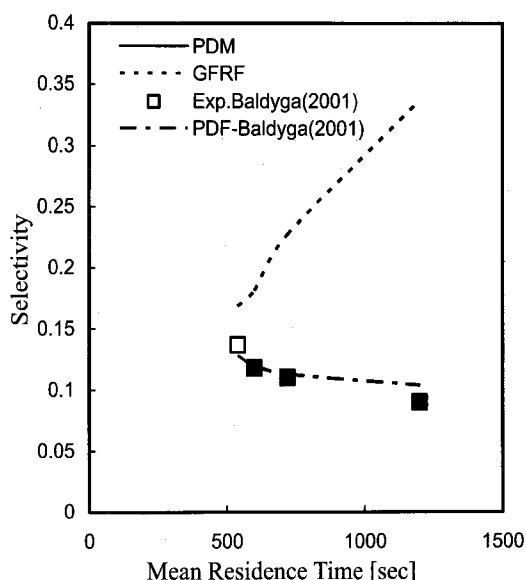


Figure 7. The Comparison The Prediction Results of Sodium Ethyl Chloroacetate Selectivity with Baldyga's Experimental Data for Impeller Rotation Speed of 1.767 rps

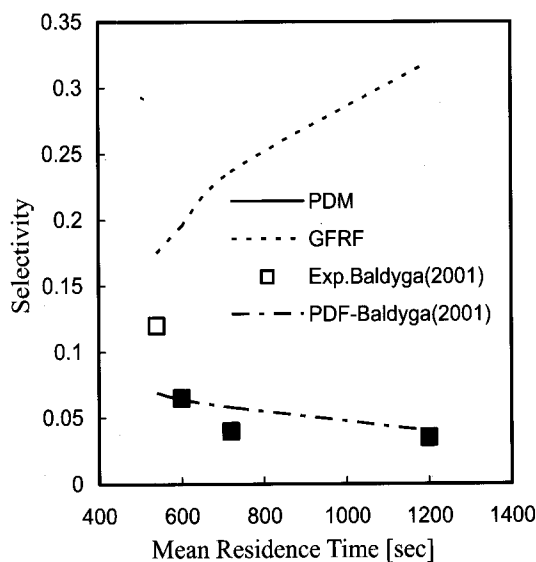


Figure 8. The Comparison The Prediction Results of Sodium Ethyl Chloroacetate Selectivity with Baldyga's Experimental Data for Impeller Rotation Speed of 2.9 rps

5. Conclusions

These works has predicted the effect of two process variables, impeller rotation speed and mean residence time, on product distribution of parallel reaction in continuous stirred tank reactor using two approaches. Closure approach using

GFRF model based on CFD available in FLUENT using PDF model especially and phenomenological approach using Packet Diffusion Model based on computer program written in PASCAL. The reaction system studied is the system concerning netralization of HCl and hydrolyzation of Ethyl Chloroacetate by NaOH producing Sodium Ethyl Chloroacetate as by product. The simulation results either using GFRF or PDM show that the selectivity of Sodium Ethyl Chloroacetate decreases with increasing impeller rotation speed. The selectivity of Sodium Ethyl Chloroacetate increases for GFRF and decreases for PDM with increasing mean residence time in the tank. The prediction of the selectivity of Sodium Chloroacetate using PDM is smaller than that predicted by GFRF model. The prediction using PDM agree very well with the experimental data by Baldyga (2001) and Baldyga's prediction for rotation speed of 1.767 rps.

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Notations

- A,B = Empirical constant equal to 4 dan 0,5 in Equation(4) and (5), -
- b = Baffle width
- C_{k,IN} = The concentration of species k in reactorinfluent, kmole/ m³
- C_{k,m} = The concentration of species k in maximum mixedness reactor, kmole/m³
- C_{kI} = The concentration of species k in packet I , Kmole/ m³
- C_{kII} = The concentration of species k in packet II, kmole/ m³
- $\bar{C}(\tau)$ = Average species concentration packet /m³ as function of, kmole/ m³
- \bar{C}_k^I = Dimensionless concentration of species k in packet I defined as:

$$\bar{C}_k^I = C_k^I / C_{A.In} , \text{Kmole/ m}^3$$
- \bar{C}_k^{II} = Dimensionless concentration of species k in packet II defined as:

$$\bar{C}_k^{II} = C_k^{II} / C_{B.In} , \text{kmole/ m}^3$$
- C_{j'} = Molar concentration of each reactant or product j', kmole/ m³

D	=	impeller diameter, m
D_k	=	Diffusion coefficient of species k in diffusion packet, m ² /s
$D_{i,m}$	=	Diffusion coefficient of species i' in mixture, m ² /s
E ₁	=	Defined as $E_1 = \varepsilon_1 / (q C_{A,In})$
E ₂	=	Defined as $E_2 = \varepsilon_2 / (q C_{A,In})$
f(t)	=	Residence Time Distribution Function
H	=	Tank height
$J^{i,i}$	=	Diffusion flux of species i', kmole/m ² s
k	=	Turbulent kinetic energy, m ² /s ²
k ₁	=	Reaction rate constant for the first reaction, m ³ / kmol.s
k ₂	=	Reaction rate constant for the second reaction, m ³ / kmol.s
k _{b,k}	=	Backward reaction rate constant in k-reaction, m ³ / kmol.s
k _{f,k}	=	Forward reaction rate constant in k-reaction, m ³ / kmol.s
m _{i'}	=	Local mass fraction of species i'
M	=	Molecular weight, kg/kmol
M _{i'}	=	Molecular weight of species i', kg/kmol
M _R	=	Molecular weight of reactant, kg/kmol
m _p	=	Mass fraction of product
m _R	=	Mass fraction of reactant
MB	=	Defined as $MB = C_{B,In} / C_{A,In}$
MD	=	Defined as $D = C_{D,In} / C_{A,In}$
N	=	Impeller rotation speed, s ⁻¹
N _C	=	Number of components
NR	=	Number of reactors
q _I	=	Volumetric flow rate of total feed stream into reactor, m ³ /s
q _{II}	=	Volumetric flow rate of first feed stream into reactor, m ³ /s
r	=	Radial position in diffusion packet, m
\bar{r}	=	Defined as $\bar{r} = r / r_p$
R _{i'}	=	Molar rate of species i' formation, kmole/m ³ s
R _{i,k}	=	Molar rate of species i' formation in reaction k, kmole/m ³ s
Sc _t	=	Turbulent Schmidt number
S _{i'}	=	Rate of formation by the addition from dispersed phase and other sources, kmole/m ³ s
S _{i'}	=	Rate of formation by the addition from dispersed phase and other sources, kmole/m ³ s

D	=	impeller diameter, m
Sg_k	=	Segregation number of species k defined as $Sg_k = \frac{\mu^{3/2}}{4\pi^2 \rho^{3/2} \varepsilon^{1/2} D_k \bar{t}}$
t	=	Time, s
\bar{t}	=	Mean residence time, s
T	=	Tank diameter, m
u _i	=	Velocity in i direction, m s ⁻¹
V	=	Reactor volume, m ³
W	=	Blade width, m
X	=	Conversion
S	=	Selectivity of reaction product

Greek Letters

ε	=	Energy dissipation rate, m ² s ⁻³
ε_1	=	Reaction progress for the first reaction, kmol s ⁻¹
ε^2	=	Reaction progress for the second reaction, kmol s ⁻¹
K ₁	=	Defined as $K_1 = k_1 \bar{t} C_{A,In}$
K ₂	=	Defined as $K_2 = k_2 \bar{t} C_{A,In}$
$v_{i',k}$	=	Reactant Stoichiometric coefficient of reaction k for species i'
$v_{i',k}''$	=	Product stoichiometric coefficient of reaction k for species i'
ρ	=	Density, kg m ⁻³
τ	=	Dimensionless time defined as $\tau = t / \bar{t}$
η', η''	=	Reaction order

References

- [1] Assirelli, M., W. Bujalski, W. Nienow, and A. Eagleshaw (2002), "Study of Micro mixing in Stirred Tank Using a Rushton Turbine: Comparison of Feed Position and Other Mixing Devices", *CHISA 2002*.
- [2] Baldyga, J. (1994), "A Closure Model for Homogeneous Chemical Reactions", *Chem. Eng. Sci.*, Vol. 49, No. 1, pp. 1985-2003.
- [3] Baldyga, J., M. Henczka dan L. Makowski (2001), "Effects of Mixing on Parallel Chemical Reactions in a Continuous-Flow Stirred tank reactor", *Trans. Ichem E*, Vol. 79 Part A, pp. 895-900.

- [4] Bourne, J. R. dan Shengyao Yu (1994)," Investigation of Micromixing in Stirred Tank Reactors Using Parallel Reactions", *Ind. Eng Chem. Res.*, Vol.33, pp.41-55.
- [5] Fournier, M.C., Falk, J.Villermux (1996)," A New Parallel Competing Reactions System For Assessing Micromixing Efficiency. Determination of Micromixing time by A simple mixing model", *Chem Eng Sci.*, Vol. 51, No. 23, pp.5187-5192.
- [6] Guichardon, P, L. Falk, M.Anddrieu(2001),"Experimental Comparison of the Iodide-Iodate and the Diazo Coupling Micromixing Test Reactions in Stirred Reactors", *Trans IChemE.*, Vol.79, Part A, pp.906-914.
- [7] Nauman,E.B. (1975)."Droplets Diffusion Model for Micromixing", *ChemEng.Sci.*, Vol.30, pp.1135-1140.
- [8] Villermux,J., Falk, L., dan Fournier, M.C. (1992),"Use of Parallel Competing Reactions to Characterize Micromixing Efficiency", *AIChE Journal, Symp.Ser.* Vol.286, No. 88, pp. 610.