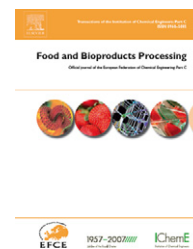


available at www.sciencedirect.comjournal homepage: www.elsevier.com/locate/fbp

External mass transfer model for the hydrolysis of palm olein using immobilized lipase

Yin Hoon Chew^a, Chew Tin Lee^{a,*}, Mohamad Roji Sarmidi^b, Ramlan Abdul Aziz^b, Firdausi Razali^a

^a Department of Bioprocess Engineering, Faculty of Chemical and Natural Resources Engineering, Universiti Teknologi Malaysia, 81310 Skudai, Johor, Malaysia

^b Chemical Engineering Pilot Plant, Universiti Teknologi Malaysia, 81310 Skudai, Johor, Malaysia

ARTICLE INFO

Article history:

Received 10 July 2007

Accepted 7 February 2008

Keywords:

Mass transfer model
Immobilized lipase
Packed-bed reactor
Palm olein

ABSTRACT

The application of immobilized enzyme in packed-bed reactor is gaining interest in the industry as it offers advantages over conventional chemical reactions. However, external mass transfer limitation is significant in immobilized enzyme packed-bed reactors, especially at large scales. This study aimed to develop an external mass transfer model for immobilized lipase (EC 3.1.1.3) during the hydrolysis of palm olein. A mass transfer correlation model of the form $J_D = KRe^{n-1}$ was developed based on the literatures. The Colburn factor, J_D , which is a function of Reynolds and Schmidt numbers, can be related to the external mass transfer coefficient, k_m . The values of K and n were determined by conducting experimental work at different mass flow rates. It was found that the values of K and n are 0.093 and 0.5, respectively. Since the average mass transfer coefficients can be correlated in terms of dimensionless groups which characterize the flow conditions, this model can be used for reactor scale-up design.

© 2008 The Institution of Chemical Engineers. Published by Elsevier B.V. All rights reserved.

1. Introduction

Mass transfer limitation is one of the major concerns in the utilization of immobilized enzymes for industrial processes. In most cases, the phenomenon of mass transfer affects the overall reaction rate of a system. Therefore, it is necessary to consider this aspect during the design and operation of immobilized enzyme reactors.

Apart from understanding the qualitative effects of mass transfer in enzymatic system, a quantitative study is also important. A mathematical model describing the diffusional restriction and the parameters which affect this restriction is not only useful for model systems, but would also be beneficial for systems of direct industrial application.

In a reactor packed with immobilized enzyme particles, two transport processes occur. The first process is the transfer of the substrate from the bulk liquid phase to the surface of the immobilized biocatalyst due to the formation of a fic-

titious laminar film. The second process is the simultaneous diffusion and reaction of the substrate within the biocatalyst particles. External mass transfer limitations occur if the rate of diffusional transport through the laminar film is rate limiting. On the other hand, internal diffusional limitations within porous carriers indicate that the slowest step is the penetration of the substrate into the interior of the catalyst particle.

The roles and effects of both internal and external mass transfer limitations have been extensively studied (Bailey and Ollis, 1986). Numerous analytical approaches and dimensionless numbers have been generated to simplify the assessment of these limitations. For internal mass transfer, solutions such as the plots of effectiveness factors (Fig. 4.21 in Bailey and Ollis, 1986) can be used. The plots are general for immobilized enzyme catalysts and developed based on some simple manipulations to eliminate uncertainties in the intrinsic parameters.

External mass transfer coefficients, on the other hand, are usually evaluated from available correlations in the chemical

* Corresponding author. Tel.: +60 16 2320865; fax: +60 7 5535538.

E-mail address: ctlee@fkkksa.utm.my (C.T. Lee).

Nomenclature

a_c	cross-sectional area of column
a_m	surface area per unit weight of immobilized enzyme for external mass transfer
C	concentration of substrate (palm olein) in the bulk liquid
C_{in}	column inlet substrate (palm olein) concentration
C_{out}	column outlet substrate (palm olein) concentration
C_s	substrate (palm olein) concentration on the outer surface of the immobilized particle
C_0	initial palm olein concentration in the reservoir
C_1	concentration of palm olein in the reservoir
C_2	concentration of palm olein at the outlet of the packed-bed column to be circulated back to the reservoir
d_p	enzyme particle diameter
D_f	diffusivity
G	mass flux
H	height of the column
J_D	Colburn factor
k	'Surface' first-order reaction rate constant
k_m	external mass transfer coefficient
k_p	observed first-order reaction rate constant
K	mass transfer correlation constant
n	exponential factor in mass transfer correlation
N	group of parameters
N_{Sc}	Schmidt number
r	reaction (substrate consumption) rate
r_m	external mass transfer rate
Re	Reynolds number
t	time
V_{res}	volume of the reacting solution in the reservoir
W	total amount of immobilized enzyme particles
z	distance from the bottom of the packing in a column
Greek letters	
ε	void fraction in a packed-bed
μ	fluid viscosity
ρ	fluid density
ρ_p	enzyme particle density
τ	residence time in the reservoir

engineering literatures. According to [Rovito and Kittrell \(1973\)](#), the existing chemical engineering principles for mass transfer problems in heterogeneous catalysis should also be applicable for analysis of data on immobilized enzyme system. However, these applications have been empirical in nature. Thus, it is necessary to evaluate each immobilized enzyme system individually.

In this study, the external mass transfer model for immobilized lipase (EC 3.1.1.3) during the hydrolysis of palm olein in a recirculated packed-bed batch reactor (RPBBR) was developed. An RPBBR is shown in [Fig. 1](#) ([Mutlu and Gökmen, 1998](#)). It consists of a packed-bed reactor with recycling system.

1.1. Development of an external mass transfer model

The external mass transfer model presented here is developed based on the approach used by [Rovito and Kittrell \(1973\)](#). A few

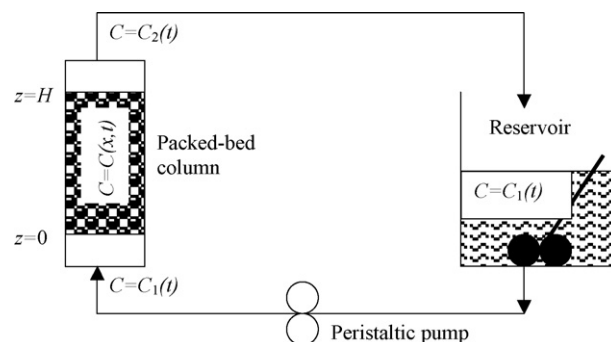


Fig. 1 – Schematic representation of a recirculated packed-bed batch reactor (RPBBR).

assumptions have been made during the development of this model as follows:

- The reaction follows a first-order rate (this is especially true at low substrate concentrations).
- The immobilized enzyme particles are spherical.
- The packed-bed column has a plug flow with no axial dispersion.
- The enzyme activity throughout the particle is uniform.

1.2. Apparent reaction rate

A material balance for palm olein (substrate) in the packed-bed column was first developed as shown in the following equation

$$\left(\frac{HQ}{W}\right) \frac{dC}{dz} \times 6 \times 10^{-2} = -r \quad (1)$$

where r is the reaction (substrate consumption) rate ($\text{mg g}^{-1} \text{h}^{-1}$), Q the volumetric flow rate (ml min^{-1}), H the height of the column (cm), W the total amount of immobilized enzyme particles (g), and dC/dz is the concentration gradient along the column length ($\text{mg l}^{-1} \text{cm}^{-1}$).

Since a first-order reaction rate was assumed, the relation between the apparent reaction rate and bulk substrate (palm olein) concentration in the column is given as

$$r = k_p C \quad (2)$$

where k_p is the apparent first-order reaction rate constant ($\text{l g}^{-1} \text{h}^{-1}$) or the observed reaction rate constant and C is the bulk substrate concentration (mg l^{-1}). Combining Eqs. (1) and (2) gives

$$\left(\frac{HQ}{W}\right) \frac{dC}{dz} \times 6 \times 10^{-2} = -k_p C \quad (3)$$

Eq. (4) is found by integrating Eq. (3) using boundary conditions at $z=0$ of $C=C_{in}$, and at $z=H$ of $C=C_{out}$

$$\ln \left(\frac{C_{in}}{C_{out}} \right) = \frac{W}{Q} k_p \times (10^3/60) \quad (4)$$

where C_{in} is the column inlet substrate (palm olein) concentration (mg l^{-1}) and C_{out} is the column outlet substrate (palm olein) concentration (mg l^{-1}). The concentration at the outlet

of the packed-bed is therefore given by

$$C_{\text{out}} = C_{\text{in}} e^{-N} \quad (5)$$

with N defined as

$$N = \frac{W}{Q} k_p \times (10^3/60) \quad (6)$$

Eq. (5) only gives the relation between the inlet and outlet concentration of palm olein in the packed-bed column every time the fluid flows through the column. Since a recycling system is involved, the inlet concentration to the column changes for every cycle. Therefore, an overall mass balance for an RPBBR is as follows. Referring to Fig. 1, if the reservoir is a perfectly mixed tank, the total mass balance gives

$$\frac{dV}{dt} = 0 \quad (7)$$

where V is the volume of the reacting solution in the reservoir (ml). The component balance in the reservoir gives

$$\frac{dC_1}{dt} = -\frac{1}{\tau} (C_2 - C_1) \quad (8)$$

where τ is the residence time (min) in the reservoir (V/Q), C_1 the concentration of palm olein (mg l^{-1}) in the reservoir, and C_2 is the concentration (mg l^{-1}) at the outlet of the packed-bed column to be circulated back to the reservoir. Based on Eq. (5), C_2 can be defined as follows:

$$C_2 = C_1 e^{-N} \quad (9)$$

Substituting Eq. (9) into Eq. (8) gives

$$\frac{dC_1}{dt} = -\frac{1}{\tau} (C_1 e^{-N} - C_1) \quad (10)$$

Integrating Eq. (10) using boundary conditions of $V = V_{\text{res}}$ and $C_1 = C_0$ when $t = 0$ gives the change of palm olein concentration in the reservoir with time as

$$C_1 = C_0 \exp \left[\frac{-(e^{-N} - 1)t}{\tau} \right] \quad (11)$$

Based on Eq. (11), a plot of $\ln (C_1/C_0)$ versus time will give a slope term as follows:

$$\text{slope} = -\frac{e^{-N} - 1}{\tau} \quad (12)$$

If a constant quantity of immobilized enzyme particles is used, the apparent reaction rate constant, k_p for each flow rate can be found from Eq. (6) when the value of N is known (from the slope as shown in Eq. (12)). k_p is the apparent rate constant which takes into account both the reaction and mass transfer phenomena.

1.3. Apparent reaction rate as a function of external mass transfer limitation

The mass transfer rate of the palm olein from the bulk liquid to the outer surface of the immobilized beads is proportional to the external mass transfer coefficient, area of external mass transfer and the concentration difference between the bulk

and the external surface of immobilized bead:

$$r_m = k_m a_m (C - C_s) \quad (13)$$

where r_m is the external mass transfer rate ($\text{mg g}^{-1} \text{h}^{-1}$), k_m is the external mass transfer coefficient (cm h^{-1}), and a_m is the surface area per unit weight of immobilized particle for external mass transfer ($\text{cm}^2 \text{mg}^{-1}$), while C and C_s is the substrate concentration in the bulk liquid and on the outer surface of the immobilized particle (mg l^{-1}), respectively. The value of a_m can be determined using the following equation

$$a_m = \frac{6}{\rho_p d_p} \quad (14)$$

with d_p as the particle diameter (cm) and ρ_p the particle density (mg cm^{-3}).

A first-order reaction rate is derived to account for the overall rate of substrate utilization of each enzyme particle. The reaction rate is written to take the form of first-order kinetics towards C_s as follows:

$$r = k C_s \quad (15)$$

k is the 'surface' first-order reaction rate constant ($\text{l g}^{-1} \text{h}^{-1}$) which takes into account both the effective internal mass transfer and the intrinsic reaction. According to Özdural (1994), at low substrate concentration, the effective internal mass transfer coefficient can be assumed constant. As the substrate concentrations used in this study were low, Eq. (15) is valid throughout this study.

Since the rates of external mass transfer and overall substrate utilization by the particle will be the same at steady state, Eqs. (13) and (15) are equated and rearranged to give

$$C_s = \frac{k_m a_m C}{k + k_m a_m} \quad (16)$$

Substituting Eq. (16) into Eq. (15) and equating with Eq. (2), the effects of external mass transfer on the apparent reaction rate constant, k_p is shown in Eq. (17).

$$k_p = \frac{k k_m a_m}{k + k_m a_m} \quad (17)$$

or

$$\frac{1}{k_p} = \frac{1}{k} + \frac{1}{k_m a_m} \quad (18)$$

The terms $(1/k)$ and $(1/k_m a_m)$ show the contributions of overall substrate utilization by particle and external mass transfer resistance on the k_p , respectively, at constant temperature.

1.4. External mass transfer correlation model

The value of k is constant as far as this particular reaction is concerned and is independent of the operating parameter, particularly the mass flow rate and the scale of the system. However, the external mass transfer coefficient, k_m changes with parameters such as flow rate, reactor diameter and fluid properties. This in turn changes the apparent reaction rate. Therefore, a correlation is needed so that the mass transfer coefficient can be estimated at different operating parameters and during scale-up.

Average mass transfer coefficients between the bulk fluid and particle surface in the packed-bed reactor can be correlated in terms of dimensionless groups which characterize the flow conditions (Satterfield, 1970; Bailey and Ollis, 1986; Nath and Chand, 1996). The correlation of the external mass transfer coefficient, k_m , with variables such as flow rate, reactor diameter and fluid properties can be obtained by defining a dimensionless group as follows:

$$J_D = \frac{k_m \rho}{G} \left(\frac{\mu}{\rho D_f} \right)^{2/3} \quad (19)$$

where J_D is the Colburn factor, defined in terms of Schmidt number and Reynolds number.

The Schmidt number is the term in parentheses in Eq. (19) as follows:

$$N_{Sc} = \frac{\mu}{\rho D_f} \quad (20)$$

The symbols μ , ρ and D_f are the fluid viscosity ($\text{g cm}^{-1} \text{ min}^{-1}$), density (g ml^{-1}) and diffusivity (cm min^{-1}), respectively.

The Reynolds number can be defined as

$$Re = \frac{d_p G}{\mu} \quad (21)$$

where d_p is the particle diameter (cm).

G is the mass flux ($\text{g cm}^{-2} \text{ min}^{-1}$) and it can be calculated using Eq. (22) as follows:

$$G = \frac{Q\rho}{a_c \varepsilon} \quad (22)$$

where Q is the volumetric flow rate (ml min^{-1}), a_c the cross-sectional area of column (cm^2) and ε is the void fraction in a packed-bed.

A few correlations for mass flow rates are available, varying in the dependence of the Colburn factor, J_D , on Re . This correlation was suggested by Chilton and Colburn (1934) as follows:

$$J_D = K Re^{(n-1)} \quad (23)$$

This general correlation is not only applicable in immobilized enzyme systems, but it can also correlate data in immobilized cell systems (Nath and Chand, 1996; Aksu and Bülbül, 1998; Murugesan and Sheeja, 2005).

Different mass transfer conditions have different values of K and n . The value of n varies from 0.1 to 1.0. Equating Eqs. (19) and (23) and solving for the external mass transfer coefficient gives

$$k_m = \left(\frac{K}{\rho} \right) \left(\frac{\mu}{\rho D_f} \right)^{-2/3} \left(\frac{d_p}{\mu} \right)^{n-1} G^n \quad (24)$$

or

$$k_m = A G^n \quad (25)$$

where

$$A = \left(\frac{K}{\rho} \right) \left(\frac{\mu}{\rho D_f} \right)^{-2/3} \left(\frac{d_p}{\mu} \right)^{n-1}$$

Substituting Eq. (25) into Eq. (18) and rearranging it leads to the following equation:

$$\left(\frac{1}{k_p} \right) = \left(\frac{1}{A a_m} \right) \left(\frac{1}{G^n} \right) + \left(\frac{1}{k} \right) \quad (26)$$

Eq. (26) can be analyzed for different values of n ranging from 0.1 to 1.0. A straight line of slope $1/(A a_m)$ and intercept $1/k$ should be obtained if the experimentally measured values of $1/k_p$ versus $1/G^n$ for each value of n is plotted. The calculated values of A and k (the 'surface' first-order reaction rate constant) from the graph are then used to get the values of k_m (using Eq. (25)) and an estimated k_p (using Eq. (18)). A trial-and-error procedure is repeated for all n values until the estimated value of k_p matches well with the experimental k_p .

2. Materials and methods

2.1. Materials

The commercial immobilized lipase, Lipozyme TL IM (bead size 0.3–1.0 mm, wet bulk density 415 kg/m^3), was obtained from Novozyme. A commercial cooking oil Seri Murni which is distributed by FFM Marketing Sdn Bhd Malaysia, was used as the source of palm olein. The oil contains 44.3 weight % saturated fats, 12.1% poly-unsaturated fats and 43.6% mono-unsaturated fats. Oleic acid, palmitic acid and linoleic acid (GC standard) were purchased from Sigma-Aldrich. *n*-Hexane was of analytical grade and purchased from Fluka Chemie AG, Switzerland.

2.2. Hydrolysis of palm olein in an RPBBR

The batch stirred-tank reactor consisted of a water-jacketed vessel with a maximum capacity of 50 ml and a magnetic stirrer. A water bath (Grant Instruments, Cambridge, England) was used to maintain the temperature of the reaction mixture in the vessel. A peristaltic pump (Masterflex, Cole-Parmer) and a thermostat XK 16/20 (16 mm i.d. \times 20 cm length) jacketed column from Pharmacia Biotech, Sweden was installed to the batch reactor to form a recirculated packed-bed batch reactor (RPBBR).

The reaction mixture (15 ml of palm olein, 23.8 ml of *n*-hexane, 1.2 ml of water) was first prepared and incubated at 55°C and 200 rpm. The amount of substrates used were decided based on a kinetic study (data not shown) to ensure the reaction was in first-order. Two grams of immobilized lipase was then packed into the jacketed column. A time zero-sample was taken. Samples were taken at different time intervals and analysed for fatty acids. Experiments were repeated at four different flow rates (2, 6, 10, and 20 ml min^{-1}). Duplicate was made for each experiment to ensure the reproducibility and accuracy of the data.

2.3. Sample analysis

All the samples were analysed using gas chromatography. A Shimadzu GC-17A Version 3 (Kyoto, Japan) equipped with a flame-ionization detector (FID) was used. A Nukol fused-silica capillary column (15 m length \times 0.53 mm i.d. \times 0.5 μm film thickness, Supelco, USA) was used with nitrogen as the carrier gas. The injector and detector were set at 220°C . The column temperature was programmed to rise from 180 to

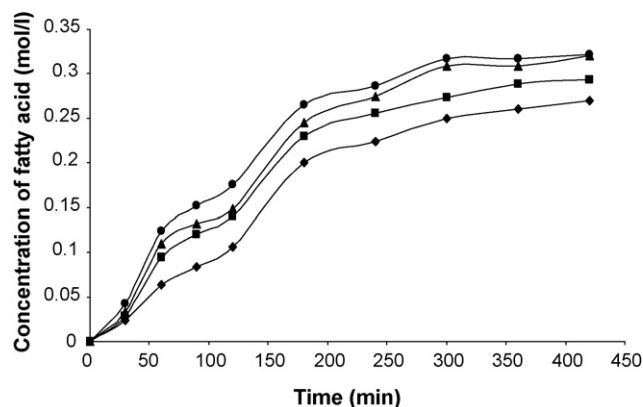


Fig. 2 – Hydrolysis profiles at different liquid flow rates: (a) 2 ml min⁻¹ (♦); (b) 6 ml min⁻¹ (■); (c) 10 ml min⁻¹ (▲); (d) 20 ml min⁻¹ (●).

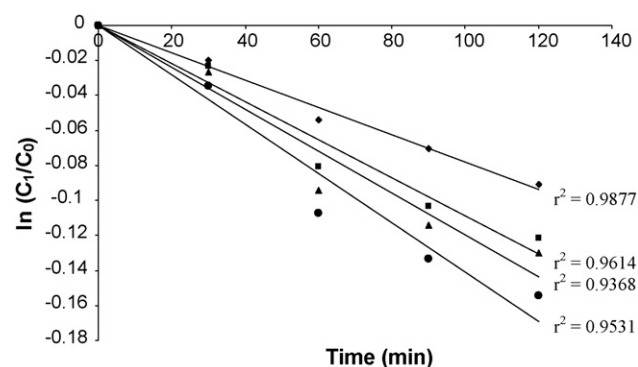


Fig. 3 – Overall rate of reaction at different flow rates: (a) 2 ml min⁻¹ (♦); (b) 6 ml min⁻¹ (■); (c) 10 ml min⁻¹ (▲); (d) 20 ml min⁻¹ (●).

215 °C at 12 °C/min and maintain for 4 min before rising again at 12 °C/min until it reaches 220 °C and stay for 2 min. The gas chromatography column was connected to Shimadzu CLASS-VP Chromatography Data System software (Columbia, USA). Calibration curves for the fatty acids were first prepared using the external GC standard.

3. Results and discussion

The hydrolysis profile in the recirculated packed-bed batch reactor at various flow rates for the mass transfer study is presented in Fig. 2. Results reported were the average values of the duplicate data with both sets of experiment giving very similar results. Experimental data in Fig. 2 were analyzed to determine the overall reaction rate at each flow rate. Only the data in the first 2 h were used. This was because as the reaction proceeded further, the reaction rate started to decrease until it achieved its maximum conversion. During this period, the reaction rate was governed by the equilibrium thermodynamics and not the intrinsic reaction or mass transfer. Thus, experimental data after 2 h were not included in this analysis.

Based on the data in Fig. 2, the concentration of palm olein at each time interval was calculated based on the concentration of fatty acids formed. Plots of $\ln C_1/C_0$ as a function of time at different flow rates are shown in Fig. 3.

The slope of each line in Fig. 3 was determined and used to calculate the value of N (Eq. (12)). After that, the value of k_p , the observed first-order reaction rate constant, was obtained

Table 1 – Observed first-order reaction rate constants, k_p , at different flow rates

Flow rate (ml min ⁻¹)	Residence time (min)	Slope $\times 10^3$ (min ⁻¹)	$k_p \times 10^4$ (lg ⁻¹ h ⁻¹)
2	20	0.8	9.68
6	6.67	1.1	13.25
10	4	1.2	14.43
20	2	1.4	16.82

for each flow rate using Eq. (6). The calculated values of k_p are listed in Table 1.

Table 1 shows that as the flow rate increases, the value of k_p also increases. This means that the overall reaction rate is higher at higher flow rate. This is the expected effect in a film diffusion-controlled system as reported by Bihari (1985). According to the report, increasing the linear velocity of the feed would decrease the film thickness surrounding the immobilized enzyme particles and hence reduce the external mass transfer resistances.

Referring to Eq. (26), a plot of the experimentally measurable quantity of $1/k_p$ against $1/G^n$ should yield a straight line of slope $1/Aa_m$ and intercept $1/k$. This was done for the data in Table 1 using values of n from 0.1 to 1.0. Rovito and Kittrell (1973) reported that this range of value encompassed all the exponential values in the Colburn–Chilton factor that have been presented in the chemical engineering literature. A few plots of $1/k_p$ against $1/G^n$ (for n values of 0.1, 0.3, 0.5 and 0.7) are shown in Fig. 4. It was found that all values of n between 0.3 and 1.0 give satisfactory straight lines (not all plots are shown). These results are in agreement with the ones discovered by Rovito and Kittrell (1973) using immobilized glucose oxidase.

In order to determine the value of n that gives the best film diffusional model in predicting mass transfer limitations, all n values between 0.3 and 1.0 were analyzed further. The values of n at 0.1 and 0.2 were not used for further analysis as the straight lines displayed negative intercepts. The slope and intercept of each plot were used to calculate the values of k and A . After that, the value of k_m at each flow rate was estimated. Based on the calculated k and k_m , a value of k_p was recalculated and compared with the k_p found experimentally. The n value which provides the closest k_p as compared to the experimental results would be the most satisfactory model. The percent deviation of the calculated values and the experimental results for all flow rates are shown in Table 2 for $n = 0.3$, $n = 0.5$ and $n = 0.7$.

According to Table 2, a model having an exponent of 0.5 ($n = 0.5$) has the lowest percent deviation, with each less than 2%. All the other values of n (not all data are shown) give deviations more than 2%. Therefore, a model having an exponent of 0.5 would provide satisfactory predictions of the external mass

Table 2 – The percent deviation of calculated values of k_p from the experimental values at different n

Q (ml/min)	Experimental $k_p \times 10^4$ (lg ⁻¹ h ⁻¹)	Percent deviation (%)		
		$n = 0.3$	$n = 0.5$	$n = 0.7$
2	9.68	0.85	0.12	-0.36
6	13.25	-2.92	-1.18	0.50
10	14.43	0.76	1.69	2.36
20	16.82	1.50	-0.62	-2.62

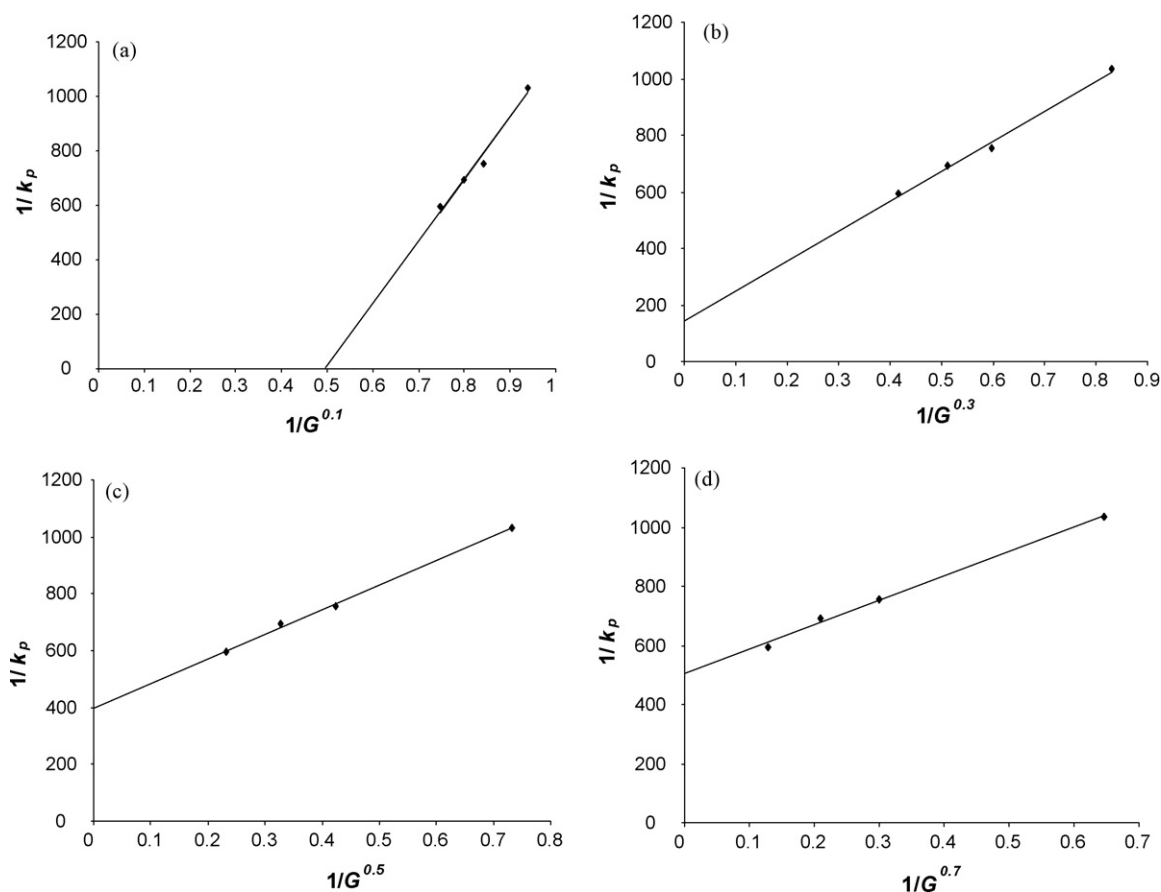


Fig. 4 – Plots of $1/k_p$ against $1/G^n$ for (a) $n = 0.1$; (b) $n = 0.3$; (c) $n = 0.5$; (d) $n = 0.7$.

transfer coefficients in immobilized lipase system. Rovito and Kittrell (1973) reported the same result in their film diffusion studies with immobilized glucose oxidase. They found that models with values of n other than 0.5 could not represent their reactor bed. The model that they used was the McCune–Wilhelm model (McCune and Wilhelm, 1949), which has an n value of 0.493. Therefore, the value of n found in this study, 0.5, is comparable to the value in the McCune–Wilhelm model.

At n equals to 0.5, the value of k was found to be $2.516 \times 10^{-3} \text{ l g}^{-1} \text{ h}^{-1}$ while the value of A was 0.005193. Using these values, K was calculated and found to be 0.093. Therefore, the external mass transfer correlation model for the immobilized lipase system in this study can be written as

$$J_D = 0.093 \text{Re}^{-0.5} \quad (27)$$

An analysis on the data in Table 2 could give a clearer picture on the effects of external mass transfer limitations on the overall observed reaction rate. The percent contribution

of external mass transfer and overall utilization by particle are presented in Table 3.

As can be seen from Table 3, the apparent reaction rate is affected by both the film diffusion of the substrate and the overall substrate utilization of immobilized particle (which accounts for both intrinsic reaction and internal mass transfer). Both steps have significant contributions to the apparent reaction rate. At low mass fluxes, external mass transfer limitations dominate, with a contribution of 61.5% when the mass flux is $1.86 \text{ g cm}^{-2} \text{ min}^{-1}$. Higher contributions mean that the related step has a higher effect and therefore a higher limitation on the apparent reaction rate. As the mass flux goes higher, however, the contribution of external mass transfer decreases while the contribution of overall substrate utilization rate rises. At a mass flux of $18.61 \text{ g cm}^{-2} \text{ min}^{-1}$, external mass transfer only contributes 33.6% while overall utilization rate contributes 66.4%. This shows that the system is moving away from the film diffusion-limited regime. Even though the contribution of external mass transfer drops when the flow rate increases, its effects are still significant, as shown by the considerable percentage it

Table 3 – Effects of external mass transfer and overall substrate utilization rate in enzyme particle on the apparent reaction rate

$G \text{ (g cm}^{-2} \text{ min}^{-1})$	$1/k_p \text{ (l}^{-1} \text{ g h)}$	$1/k \text{ (l}^{-1} \text{ g h)}$	$1/k_m a_m \text{ (l}^{-1} \text{ g h)}$	% contribution of k	% contribution of $k_m a_m$
1.86	1032.06	397.44	634.62	38.5	61.5
5.58	763.84	397.44	366.40	52.0	48.0
9.30	681.25	397.44	283.81	58.3	41.7
18.61	598.13	397.44	200.68	66.4	33.6

holds. Therefore, it must not be neglected, especially at large scales.

4. Conclusion

Based on the results of this study, a few conclusions can be drawn. They are as follows:

- (a) The observed first-order reaction rate constant, k_p , increases when the mass flow rate of the reaction media increases. This is due to reduction in the film thickness at high flow rates in the RPBBR.
- (b) Both the film diffusion, $k_m a_m$, and overall utilization rate of particle, k , influence the apparent reaction rate constant, k_p . However, the effects of film diffusion are significant especially at low flow rates and therefore should not be ignored in any engineering analysis.
- (c) The external mass transfer correlation model of the form $J_D = 0.093Re^{-0.5}$ predicts the experimental data in this study accurately. This model is valid for Reynolds number in the range of 0.53–5.3. Its usage is recommended for the quantification of mass transfer coefficient in immobilized lipase packed-bed reactors within this range. Further studies are recommended to validate the model beyond this range.

Acknowledgements

The authors wish to thank the Short Term Grant No. 75131 provided by Research Management Centre, Universiti Teknologi Malaysia for the financial support and also Chemical Engineering Pilot Plant (CEPP), UTM for its technical support.

REFERENCES

- Aksu, Z. and Bülbül, G., 1998, Investigation of the combined effects of external mass transfer and biodegradation rates on phenol removal using immobilized *P. putida* in a packed-bed column reactor. *Enzyme Microb Technol*, 22: 397–403.
- Bailey, J.E. and Ollis, D.F., (1986). *Biochemical Engineering Fundamentals*. (McGraw-Hill, New York).
- Bihari, V., 1985, Diffusional behaviour of immobilized glucose oxidase. *J Chem Technol Biotechnol*, 35B: 83–93.
- Chilton, T.H. and Colburn, A.P., 1934, Mass transfer (absorption) coefficients predictions from data on heat transfer and fluid friction. *Ind Eng Chem*, 26: 1183–1187.
- McCune, L.K. and Wilhelm, R.H., 1949, Mass and momentum transfer in a solid–liquid system. *Ind Eng Chem*, 41: 1124–1134.
- Murugesan, T. and Sheeja, R.Y., 2005, A correlation for the mass transfer coefficients during the biodegradation of phenolic effluents in a packed-bed reactor. *Sep Purif Technol*, 42: 103–110.
- Mutlu, M. and Gökmen, V., 1998, Determination of effective mass transfer coefficient (K_c) of patulin adsorption on activated carbon packed bed columns with recycling. *J Food Eng*, 35: 259–266.
- Nath, S. and Chand, S., 1996, Mass transfer and biochemical reaction in immobilized cell packed-bed reactors: correlation of experiment with theory. *J Chem Technol Biotechnol*, 66: 286–292.
- Özdural, A.R., 1994, Determination of overall mass transfer coefficients in fixed bed sorption columns. *Chem Eng Technol*, 17: 285–289.
- Rovito, B.J. and Kittrell, J.R., 1973, Film and pore diffusion studies with immobilized glucose oxidase. *Biotechnol Bioeng*, 15: 143–161.
- Satterfield, C.N., (1970). *Mass Transfer in Heterogeneous Catalysis*. (M.I.T. Press, London, England).