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Chemical Engineering Journal 84 (2001) 475–483

Chemical Engineering Journal

www.elsevier.com/locate/cej

Theoretical analysis of a packed bed membrane reactor

V.K. Jayaraman, B.D. Kulkarni*, Ashwin Rao

Chemical Engineering Division, National Chemical Laboratory, Pune 411 008, India

Received 14 January 2000; received in revised form 2 October 2000; accepted 4 October 2000

Abstract

An annular reactor packed with matrices in which the catalysts/enzyme/microorganisms are immobilized, has been simulated and the results indicate that the overall resistance and hence the conversion depend upon the Thiele modulus and another parameter, α. This parameter characterizes the ratios of the diffusion times and the ratios of length scales of the bulk liquid phase and the solid phase in the reactor shell. Analytical solutions can be obtained for linear reactions and a simplified semi-analytic method has been used for obtaining concentration profiles for nonlinear reactions.

Keywords: Reactor; Catalyst; Membrane

1. Introduction

Great interest has recently been shown in the development of newer types of reactor configurations with a view to optimizing the yield of desired products and reducing the overall costs. Membrane based reactors [1–22] have shown enormous potential in fulfilling this requirement. This is mainly because these reactors integrate the desirable properties of both synthetic membranes and catalysts. A large variety of catalytic reactions and bio reactions, viz. enzymatic reactions, microbial fermentations and plant and animal cell culture are routinely being conducted in annular reactors. Tsotsis et al. [9] presented a mathematical study of ethane dehydrogenation and methane steam reforming reactions. Their general steady state mathematical model considered temperature and concentration gradients in the tube side, membrane and shell side of the reactor. Gallaher et al. [10] experimentally evaluated industrially important dehydrogenation reactions in catalytic membrane reactors. Koska et al. [13] developed a mathematical model of protein transport in packed bed ultra filtration hollow fiber bioreactors. Their model treated hydrodynamic equations as quasi-steady state while transient solute mass balance governed the redistribution in extra capillary space (ECS). Pena et al. [16] studied the performance of ethylene epoxidation in catalytic packed bed membrane reactors with two different configurations. Their study was oriented towards increasing the ethylene oxide selectivity and they found out that the membrane reactors offer a high level of operational flexibility. Recently, Langhendries et al. [19] studied the liquid phase hydrocarbon oxidation in a packed bed membrane reactor. They developed a mathematical model and interpreted the reactor performance using conversion, peroxide efficiency and oxidation product separation factor. Their membrane reactor configuration gave a substantially higher peroxide efficiency as compared to the conventional packed bed configuration. In the present communication we have used a much-simplified approach (in contrast to the more rigorous approaches of some of the above-mentioned previous work) to simulate the performance of a membrane reactor arrangement as shown schematically in Fig. 1. The reactant entering from the tube side diffuses through the membrane to enter the shell side. The shell is packed with matrices in the pores of which catalysts/enzymes/cells are immobilized. The reactant entering the shell diffuses into the pores of the matrices (which will henceforth be denoted as the solid phase) and reacts in presence of the catalysts/enzymes/cell therein. The reactant also diffuses radially through the bulk liquid medium (this will henceforth be denoted as the bulk phase) in the reactor shell. Such an arrangement can prove useful for conducting chemical and biochemical reactions. In these reactors we can (1) maintain a very high throughput; (2) control flow rates of different reactants by suitable selection of membrane component. This can be achieved by using membranes, which selectively pass species based on size, shape or interaction with them [23]; and (3) integrate separation with reaction.

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* Corresponding author. Tel./fax: +91-20-5893041.
E-mail address: bdk@ems.ncl.res.in (B.D. Kulkarni).

1385-8947/01/$ – see front matter
PII: S1385-8947(00)00272-2

Nomenclature

a	central tube radius
A_{1N}	eigen constants given by Eq. (42)
b	distance from tube center to screen outer wall
B_{1n}	eigen constant defined by Eq. (64)
c	concentration of the reactant
c_0	concentration at tube inlet
C	dimensionless concentration of the reactant
C_w	tube wall concentration
$\bar{C}(m)$	tube side mixing cup concentration
d	distance from the tube center to the annulus wall
D	diffusion coefficient of the reactant
F_1	function given by Eq. (51)
F_2	function given by Eq. (45)
H	parameter defined by Eq. (26)
I_0	modified Bessel function of the first kind, zero order
I_1	modified Bessel function of the first kind, first order
k_1	partition function at $r = a$
k_2	partition function at $r = b$
K_0	modified Bessel function of the second kind, zero order
K_1	modified Bessel function of the second kind, first order
K_m	Michaelis–Menten constant
$\bar{K}_\mathrm{m}$	dimensionless M–M constant
$M(a, b, c)$	Kummer function
Pe	Peclet number defined by Eq. (20)
r	radial coordinate
V	maximum reaction rate in the M–M kinetic rate expression
V_0	centerline laminar flow velocity for tube side
X	dimensionless radial coordinate defined by Eqs. (16a) and (16b)
y	matrix length variable
y_s	radius of the spherical matrix
Y	dimensionless matrix length variable
Y_n	eigenfunctions
z	axial distance variable
Z	dimensionless axial distance variable

Greek symbols

α	parameter defined by Eq. (24)
β	d/b parameter defined by Eq. (25)
ε	bed porosity
η	parameter defined by Eq. (58)
κ	k_2/k_1
Λ_{1N}	eigenvalues
σ	overall mass transfer resistance defined by Eqs. (47)–(49)
Φ	Thiele modulus defined by Eq. (23)
Φ_m	Thiele modulus defined by Eq. (42)
χ	parameter defined by Eq. (21)

Subscripts

1, 2, 3	tube side, membrane and annulus, respectively
b	bulk liquid phase in the shell
s	solid phase in the shell

2. Theoretical development

The following assumptions have been made in formulating the design equations:

1. The reactor is assumed to be of tubular geometry and steady state prevails in the reactor.
2. The reactant movement through the tube side is by laminar convection in the axial direction and by diffusion in the radial direction.

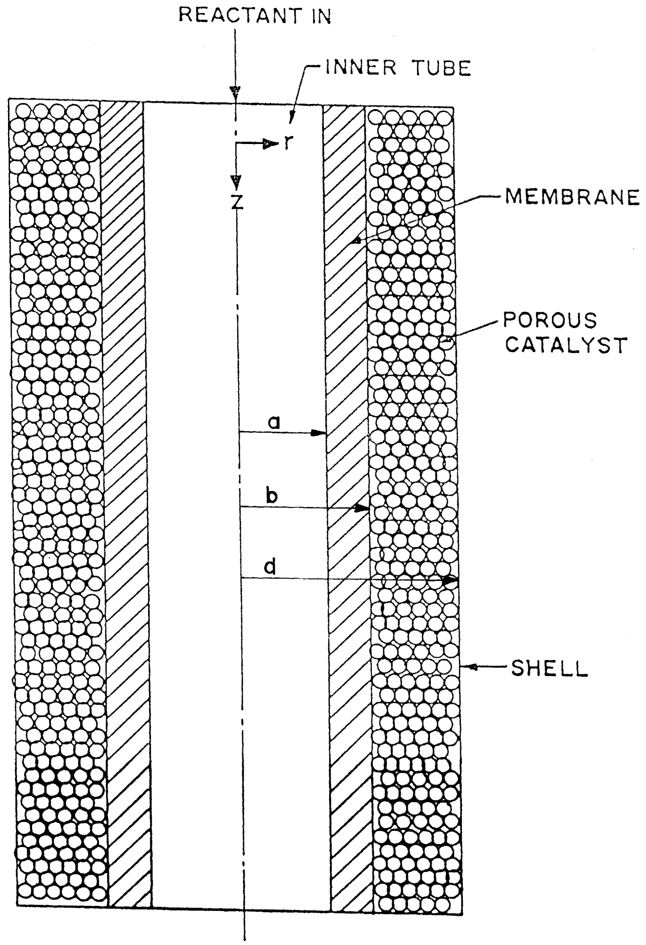

Fig. 1. Schematic of a membrane fixed bed reactor.

3. There is no convection in the membrane or shell.
4. The reactor is assumed to be isothermal and the temperature gradients are neglected.
5. The radial diffusion coefficients in the tube side, membrane bulk phase in the shell and through the pores of the matrices are constants and are independent of concentration.
6. The matrices are assumed to be of spherical shape with a uniform radius throughout the bed.
7. As a representative example, reaction rate is assumed to follow Michaelis–Menten kinetics.

With these assumptions the balance equations for the different regions can be written as follows.

Tube side:

$$\frac{1}{r}\frac{\partial}{\partial r}\left(rD_1\frac{\partial c_1}{\partial r}\right) = V_0\left(1-\frac{r^2}{a^2}\right)\frac{\partial c_1}{\partial z} \tag{1}$$

Membrane:

$$\frac{1}{r}\frac{\partial}{\partial r}\left(rD_2\frac{\partial c_2}{\partial r}\right) = 0 \tag{2}$$

Shell side bulk phase:

$$\frac{1}{r}\frac{\partial}{\partial r}\left(rD_{3\mathrm{b}}\frac{\partial c_{3\mathrm{b}}}{\partial r}\right) = \frac{3(1-\varepsilon)}{y_{\mathrm{s}}}D_{3\mathrm{s}}\left(\frac{\partial c_{3\mathrm{s}}}{\partial y}\right)_{y=y_{\mathrm{s}}} \tag{3}$$

Shell side solid phase:

$$\frac{1}{y^2}\frac{\partial}{\partial y}\left(y^2D_{3\mathrm{s}}\frac{\partial c_{3\mathrm{s}}}{\partial y}\right) = \frac{V_{\max}c_{3\mathrm{s}}}{K_{\mathrm{m}}+c_{3\mathrm{s}}} \tag{4}$$

The appropriate boundary conditions are

$$c_1(0,0) = c_0 \tag{5}$$

$$\left(\frac{\partial c_1}{\partial r}\right)_{r=0} = 0 \tag{6}$$

$$D_1\left(\frac{\partial c_1}{\partial r}\right) = D_2\left(\frac{\partial c_2}{\partial r}\right) \quad \text{at } r=a \tag{7}$$

$$\frac{c_2}{c_1} = k_1 \tag{8}$$

$$D_2\left(\frac{\partial c_2}{\partial r}\right) = D_{3\mathrm{b}}\left(\frac{\partial c_{3\mathrm{b}}}{\partial r}\right) \quad \text{at } r=b \tag{9}$$

$$\left(\frac{\partial c_{3\mathrm{b}}}{\partial r}\right) = 0 \quad \text{at } r=d \tag{10}$$

$$\frac{c_2}{c_{3\mathrm{b}}} = k_2 \tag{11}$$

$$c_{3\mathrm{b}} = c_{3\mathrm{s}} \quad \text{at } y=y_{\mathrm{s}} \tag{12}$$

$$\left(\frac{\partial c_{3\mathrm{s}}}{\partial y}\right)_{y=0} = 0 \tag{13}$$

The membrane equation (Eq. (2)) can be eliminated by integrating it and applying b.c.'s (7) and (9) to get

$$D_1\left(\frac{\partial c_1}{\partial r}\right)_{r=a} = \frac{D_2k_1}{a\ln(b/a)}(\kappa c_3 - c_1) \quad \text{at } r=a \tag{14}$$

$$D_1\left(\frac{\partial c_1}{\partial r}\right)_{r=a} = \frac{bD_{3\mathrm{b}}}{aD_1}\left(\frac{\partial c_3}{\partial r}\right)_{r=b} \tag{15}$$

Defining,

$$C_i = \frac{c_i}{c_0}, \quad i=1,2,3 \tag{16a}$$

$$C_{3\mathrm{s}} = \frac{c_{3\mathrm{s}}}{c_0} \tag{16b}$$

$$X = \frac{r}{a} \quad \text{shell side} \tag{17}$$

$$X = \frac{r}{b} \quad \text{tube side} \tag{18}$$

$$Y = \frac{y}{y_{\mathrm{s}}} \tag{19}$$

$$Pe = \frac{aV_0}{D_1} \tag{20}$$

$$K = \frac{k_2}{k_1} \tag{21}$$

$$Z = \frac{z}{a\,Pe} \tag{22}$$

Eqs. (1), (3) and (4) can be made dimensionless as follows.

Tube side:

$$\Phi^2 = \frac{y_{\mathrm{s}}^2V_{\max}}{D_{3\mathrm{s}}K_{\mathrm{m}}} \tag{23}$$

$$\alpha = 3(1-\varepsilon)\frac{D_{3\mathrm{s}}b^2}{D_{3\mathrm{b}}y_{\mathrm{s}}^2} \tag{24}$$

$$\gamma = \frac{D_{3\mathrm{b}}b}{D_1a} \tag{25}$$

$$H = \frac{K_1D_2}{D_1a\ln(a/b)} \tag{26}$$

$$\beta = \frac{d}{b} \tag{27}$$

$$\bar{K}_{\mathrm{m}} = \frac{K_{\mathrm{m}}}{c_0} \tag{28}$$

Tube side:

$$\frac{1}{X}\frac{\partial}{\partial X}\left(X\frac{\partial C_1}{\partial X}\right) = (1-X^2)\frac{\partial C_1}{\partial Z} \tag{29}$$

Shell side bulk phase:

$$\frac{1}{X}\frac{\partial}{\partial X}\left(X\frac{\partial C_{3\mathrm{b}}}{\partial X}\right) = \alpha\left(\frac{\partial C_{3\mathrm{s}}}{\partial X}\right)_{Y=1} \tag{30}$$

Shell side solid phase:

$$\frac{1}{Y^2}\frac{\partial}{\partial Y}\left(Y^2\frac{\partial C_{3\mathrm{s}}}{\partial Y}\right) = \left(\frac{\Phi^2C_{3\mathrm{s}}\bar{K}_{\mathrm{m}}}{C_{3\mathrm{s}}+\bar{K}_{\mathrm{m}}}\right) = f(C_{3\mathrm{s}}) \tag{31}$$

$$f(C_{3\mathrm{s}}) = \Phi^2C_{3\mathrm{s}} \tag{32}$$

The boundary conditions in the dimensionless form are

$$(C_1)_{Z=0} = 1 \tag{33}$$

$$\left(\frac{\partial C_1}{\partial X}\right)_{X=0} = 0 \tag{34}$$

$$\left(\frac{\partial C_1}{\partial X}\right)_{X=1} = \gamma \left(\frac{\partial C_{3\mathrm{b}}}{\partial X}\right)_{X=1} \tag{35}$$

$$\left(\frac{\partial C_1}{\partial X}\right)_{X=1} = H(kC_3 - C_1) \tag{36}$$

$$\left(\frac{\partial C_{3\mathrm{b}}}{\partial X}\right)_{X=\beta} = 0 \tag{37}$$

$$C_{3\mathrm{s}} = C_{3\mathrm{b}} \quad \text{at} \quad Y = 1 \tag{38}$$

$$\left(\frac{\partial C_{3\mathrm{s}}}{\partial Y}\right)_{Y=0} = 0 \tag{39}$$

The dimensionless parameter "Φ" is the well-known Thiele modulus which characterizes the ratio of the diffusion time scale to the reaction time scale in the catalyst phase. The parameter "α" characterizes the ratio of the radial diffusion coefficient of the reactant in the bulk phase in the shell side and the diffusion coefficient of the reactant within the spherical catalyst pores. "H" is the dimensionless membrane parameter. For constant reactor configurations the parameter "γ" indicates the ratio of the radial diffusion coefficients of the shell side and the tube side, respectively. "β" is a geometric parameter.

3. Solution procedure

3.1. First order limit of the Michaelis–Menten kinetics

For the first order limit the kinetic expression becomes linear and analytical solution is possible. The solution procedure is similar to the one adopted by Kim and Cooney [3] and Jayaraman [8] for a reactor configuration similar to the present study. Thus the shell side solid phase equation (Eq. (31)) temperature can be integrated with Eqs. (38) and (39) to obtain

$$C_{3\mathrm{s}} = C_{3\mathrm{b}} \frac{\sinh(\Phi Y)}{Y \sinh(\Phi)} \tag{40}$$

$$\left(\frac{\partial C_{3\mathrm{s}}}{\partial Y}\right)_{Y=1} = C_{3\mathrm{s}} \left(\frac{\Phi}{\tanh(\Phi)} - 1\right) \tag{41}$$

Defining,

$$\Phi_{\mathrm{m}}^2 = \alpha \left(\frac{\Phi}{\tanh(\Phi)} - 1\right) \tag{42}$$

The parameter Φ_{m} is a modified modulus, which incorporates information about the intra-particle diffusion–reaction interaction as well as the information about the ratio of the diffusion times of the bulk shell side to that of the intra-particle diffusion times. The shell side bulk liquid phase equation (Eq. (30)) can be written as

$$\frac{1}{X}\frac{\partial}{\partial X}\left(X \frac{\partial C_{3\mathrm{b}}}{\partial X}\right) = \Phi_{\mathrm{m}}^2 C_{3\mathrm{b}} \tag{43}$$

$$C_{3\mathrm{b}} = a_3(Z) F_3 \tag{44}$$

Eq. (43) can be integrated separately, subjecting to boundary conditions given by Eqs. (36) and (37) to yield

$$F_3 = I_0(\Phi_{\mathrm{m}} X) + I_1(\Phi_{\mathrm{m}}\beta) \frac{K_0(\Phi_{\mathrm{m}} X)}{K_1(\Phi_{\mathrm{m}}\beta)} \tag{45}$$

$$a_3(Z) = \left. \frac{HC_1}{HkF_3 - \gamma(\mathrm{d}F_3/\mathrm{d}X)} \right|_{X=1} \tag{46}$$

Eq. (35) can be combined with Eqs. (44)–(46) to give

$$\left. \frac{\partial C_1}{\partial X} \right|_{X=1} = -\frac{C_1}{\sigma} \tag{47}$$

$$\sigma = \frac{1}{H} - \frac{k}{\gamma} \left. \frac{F_3}{(\partial F_3/\partial X)} \right|_{X=1} \tag{48}$$

The expression for σ can be written in terms of modified Bessel function as

$$\sigma = \frac{1}{H} + \frac{\kappa}{\gamma} \frac{I_0(\Phi_{\mathrm{m}}) K_1(\Phi_{\mathrm{m}}\beta) + I_1(\Phi_{\mathrm{m}}\beta) K_1(\Phi_{\mathrm{m}})}{\Phi_{\mathrm{m}}[I_1(\Phi_{\mathrm{m}}\beta) K_1 \beta - I_1(\Phi_{\mathrm{m}}) K_1(\Phi_{\mathrm{m}}\beta)]} \tag{49}$$

The problem has now been reduced to solving the tube side equation (Eq. (29)) subject to boundary condition equation (33) and the modified boundary condition equation (47). The solution of this problem in terms of hyper geometric series is available in literature [1] and is given by

$$C_1(X, Z) = \sum_{n=1}^{\infty} A_{1n} \exp(-\Lambda_{1N}^2 Z) F_1 \tag{50}$$

where

$$F_1 = \exp(-\Lambda_{1N} \tfrac{1}{2} X^2) M(\tfrac{1}{4}(2 - \Lambda_{1N}), 1, \Lambda_{1N} X^2) \tag{51}$$

$$M(a, b, c) = 1 + \frac{a}{b} c + \frac{a}{b} \frac{a+b}{(b+1)2!} c^2 + \cdots \tag{52}$$

The eigenvalues Λ_{1N} are solutions of

$$\left(\sigma \frac{\mathrm{d}F_1}{\mathrm{d}X} + F_1\right)_{X=1} = 0 \tag{53}$$

The eigen constants can be obtained from

$$A_{1N} = -\frac{2}{\Lambda_{1N}} \left. \frac{\partial F_1/\partial X}{-(\partial^2 F_1/\partial X^2) F_1 + (\partial F_1/\partial \lambda)(\partial F_1/\partial X)} \right|_{X=1}, \quad n = 1, \infty \tag{54}$$

Once the inner tube concentration profiles are obtained the mixing cup concentration can be found out using

$$\bar{C}(m) = \frac{\int_0^1 C(X, Z)(1 - X^2) X \, \mathrm{d}X}{\int_0^1 (1 - X^2) X \, \mathrm{d}X} \tag{55}$$

which can be simplified to

$$C_1(Z) = \sum_{N=1}^{\infty} -4A_{1N}\left(\frac{\partial F_1}{\partial X}\right)_{X=1}\frac{\exp(-\Lambda_{1N}^2 Z)}{\Lambda_{1N}} \quad (56)$$

3.2. Rigorous Michaelis–Menten kinetics

For nonlinear reactions it is not possible to obtain analytical solutions. The shell side equations have to be solved separately at each axial step using a shooting procedure with Newton correction to obtain the effectiveness factor. From the effectiveness factor, which is a function of wall concentration, a dimensionless expression for the flux $(\partial C_3/\partial X)|_{X=1}$ can be obtained in terms of the nonlinear reaction term:

$$\left.\frac{\partial C_{3\mathrm{b}}}{\partial X}\right|_{X=1} \left.\frac{\eta(Z)\alpha\Phi^2(\beta^2-1)C_1\bar{K}_\mathrm{m}}{45(\bar{K}_\mathrm{m}+C_1)}\right|_{X=1} = f(C_\mathrm{w}) \quad (57)$$

where

$$\eta(Z) = \left.\frac{4.5(\partial C_{3\mathrm{b}}/\partial X)(\bar{K}_\mathrm{m}+C_1)}{\alpha\Phi^2(\beta^2-1)C_1\bar{K}_\mathrm{m}}\right|_{X=1} \quad (58)$$

The above simplification decouples the shell side equations whose functions (diffusion terms and reaction terms) now appear in Eq. (57). The tube side equation (Eq. (29)) along with the boundary conditions represented by Eqs. (33) and (57) can be solved by using Strum–Lioiville approach and Duhamel's formula. Details of solution procedure for these equations can be found in Rudisill and Levan [7] and Jayaraman and Kulkarni [12]. The solution can be written as

$$C_1(X,Z)=C_\mathrm{w}(Z)-\int \frac{\partial C_\mathrm{w}(Z')}{\partial Z'}C_\infty(X,Z-Z')\,\mathrm{d}Z' \quad (59)$$

with

$$C_\infty(X,Z-Z')=\sum_{n=1}^{\infty} A_n Y_n(X)\exp(-\Lambda_{1N}^2(Z-Z')) \quad (60)$$

where $C_\infty(X, Z-Z')$ is the solution to the tube side equation with the wall boundary condition represented by

$C_1 = 0$ at $X = 1$

Differentiating Eq. (59), we can obtain

$$-\int_0^Z \frac{\mathrm{d}C_\mathrm{w}(Z')}{\mathrm{d}Z'}\left.\frac{\partial C_\infty}{\partial Z'}\right|_{X=1}(X,Z-Z')\,\mathrm{d}Z' = f(C_\mathrm{w},\eta(Z)) \quad (61)$$

where the RHS of the equation is given by Eq. (57). The above equation is a Volterra integral equation and can be solved by discretizing Eq. (61) as follows:

$$-\sum_{j=1}^{m}\frac{C_{\mathrm{w}j}-C_{\mathrm{w}j-1}}{\Delta Z}\int_0^{Z'}\left.\frac{\partial C_\infty}{\partial Z'}\right|_{X=1}(X,Z-Z')\,\mathrm{d}Z' = f(C_\mathrm{w}(m),Z_\mathrm{m},\eta(Z_\mathrm{m})) \quad (62)$$

$$\sum_{j=1}^{m}\frac{C_{\mathrm{w}j}-C_{\mathrm{w}j-1}}{\Delta Z} \times\left(\sum_{n=1}^{\infty} B_n\exp(\Lambda_{1N}^2 Z_\mathrm{m})(\exp(\Lambda_{1N}^2 Z_j)-\exp(\Lambda_{1N}^2 Z_{j-1}))\right) = f(C_\mathrm{w}(m),\eta(m)) \quad (63)$$

where

$$B_n = -\frac{A_n(\mathrm{d}Y_n/\mathrm{d}X)|_{X=1}}{\Lambda_{1N}^2} \quad (64)$$

The above equation is a nonlinear algebraic equation and can be solved by a standard Newton–Raphson routine.

The corresponding mixing cup concentration is given by

$$\bar{C}(m) = C_\mathrm{w}(m) - \sum_{j=1}^{m}\left(\frac{C_{\mathrm{w}j}-C_{\mathrm{w}j-1}}{\Delta Z}\right) \times\left(\sum_{n=1}^{\infty}\frac{4B_n}{\Lambda_{1N}^2}\exp(-\Lambda_{1N}^2 Z_\mathrm{m})\right)(\exp(-\Lambda_{1N}^2 Z_j) -\exp(Z_{j-1})) \quad (65)$$

4. Results and discussions

4.1. First order limit of the Michaelis–Menten kinetics

An inspection of the dimensionless equations (Eqs. (24)–(33)) reveals that the concentration profiles and conversion in the reactor depend essentially on three parameters, viz., σ, Φ and α. The parameter σ, can be thought of as an overall mass transfer resistance, and its significance has been discussed in detail by Kim and Cooney [3] and Jayaraman [8]. For the reactor set-up proposed in this work, this parameter

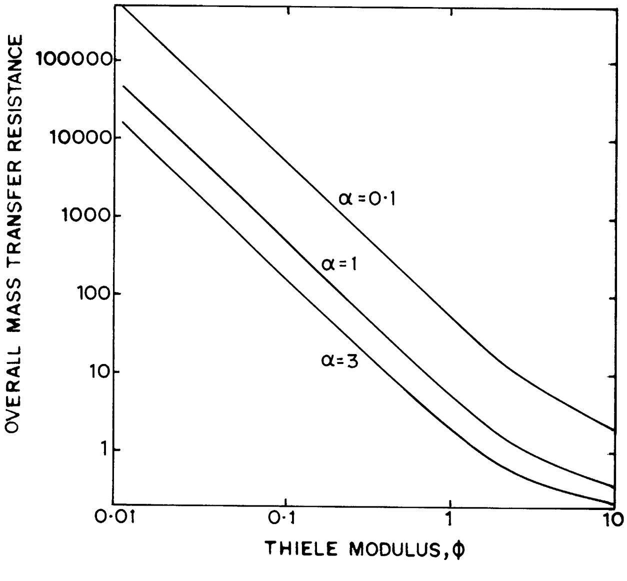

Fig. 2. Variation of overall mass transfer resistance with Thiele modulus.

represents the combined resistance of membrane, resistance offered for diffusion in the bulk liquid phase in the reactor annulus, and for diffusion and reaction in the pores of the matrices. The value of σ and hence the overall conversion would depend upon the membrane characteristics and the shell side parameters. To understand the significance of the parameters Φ and α, it is necessary to recapitulate the physical processes occurring in the annulus of the reactor. The transport of the reactant into the pores of matrices and subsequent reaction with catalysts/enzymes/cells localized there depends on the diffusion coefficient $D_{3\mathrm{s}}$, the length scale of the matrices and the reaction parameters. All these physical

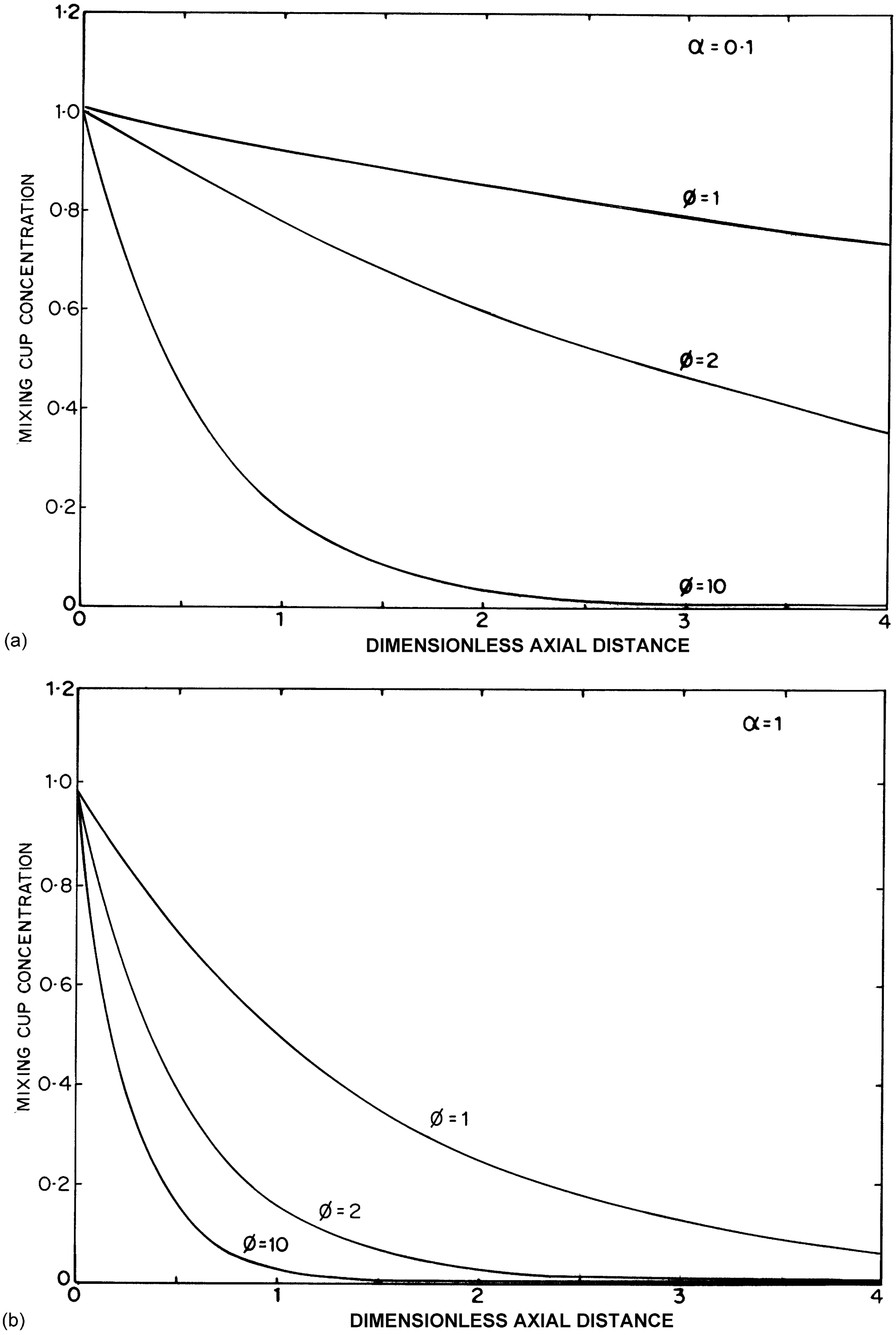

Fig. 3. (a)–(c) Mixing cup concentration profile in the reactor (first-order kinetics).

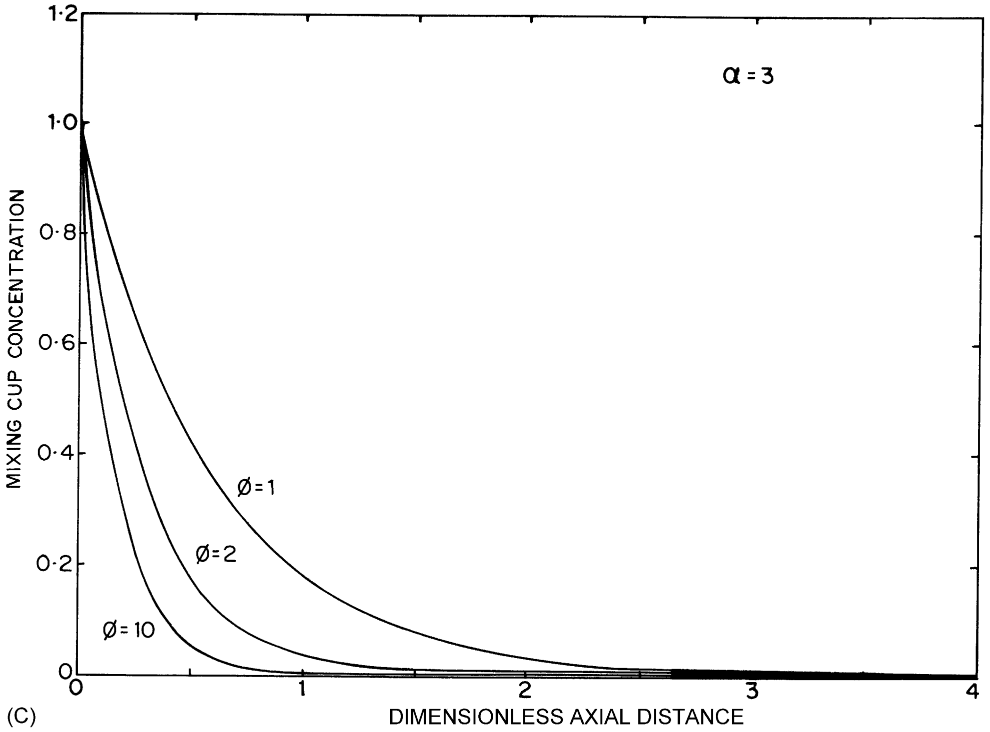

Fig. 3. (*Continued*).

parameters are contained in the dimensionless parameter Φ, the Thiele modulus. This parameter characterizes the ratio between the diffusion time and reaction time. High values of this parameter indicate diffusion-controlled regime and low values indicate reaction-controlled regime. The movement of the substrate molecules in the bulk liquid phase in the annulus is solely by radial diffusion. The substrate flux in the bulk liquid phase is controlled by the magnitude of the diffusion coefficient $D_{3\mathrm{b}}$ and the annulus length scale. Thus, the overall physical processes occurring in the reactor shell side are more complex than in conventional reactors. Due to the existence of porous matrices on the shell side, there are two distinct length scales with widely differing magnitudes. In the light of the above discussion, it is clear that the extent of reaction depends on one additional parameter α apart from the Thiele modulus. This parameter characterizes the ratio of diffusion coefficients and length scales of the bulk phase in the shell and the solid phase in the shell, respectively. Thus, the overall mass transfer resistance σ and the conversion depend significantly on both the Thiele modulus and α. This dependence is depicted in Fig. 2. It can be seen from the figure that, with an increase in Thiele modulus, the overall mass transfer resistance decreases for any value of α. This is due to the fact that with an increase in the Thiele modulus the overall reaction rate in the reactor increases thereby decreasing the mass transfer resistance. Similarly, for fixed values of the Thiele modulus, the overall mass transfer resistance decreases and conversion increases with an increase in α. Thus, the effect of α is similar to that of the Thiele modulus. This is because of the overall flux in the shell side increases with an increase in the Thiele modulus Φ.

Fig. 3 shows the mixing cup concentration profile as a function of Thiele modulus for two different axial locations in the reactor. For a given value of α, with an increase in the Thiele modulus (or vice versa), the flux of the substrate increases and the mixing cup concentration decreases (and hence conversion increases). The influence of the membrane resistance on the overall performance can be gauged by suitably varying the membrane parameters, “H”, γ and b/a. Similarly, the influence of changing the total amount of catalytic material on reactor performance can be found out by suitably varying the dimensionless parameter d/a.

Data can be collected from different experimental runs with varying values of catalytic material, membrane thickness, reactor length and diameter of tube side and shell side. The simple mode proposed in the work can be used to verify the experimental results. This can help in elucidation of different governing mechanisms and in obtaining a range of optimum parameters.

4.2. Michaelis–Menten kinetics

For this nonlinear kinetic expression analytical solutions are not possible. This is due to the fact that the flux of the substrate/reactant to the shell side (Eq. (58)) contains a nonlinear term. Similar problems have already been solved by

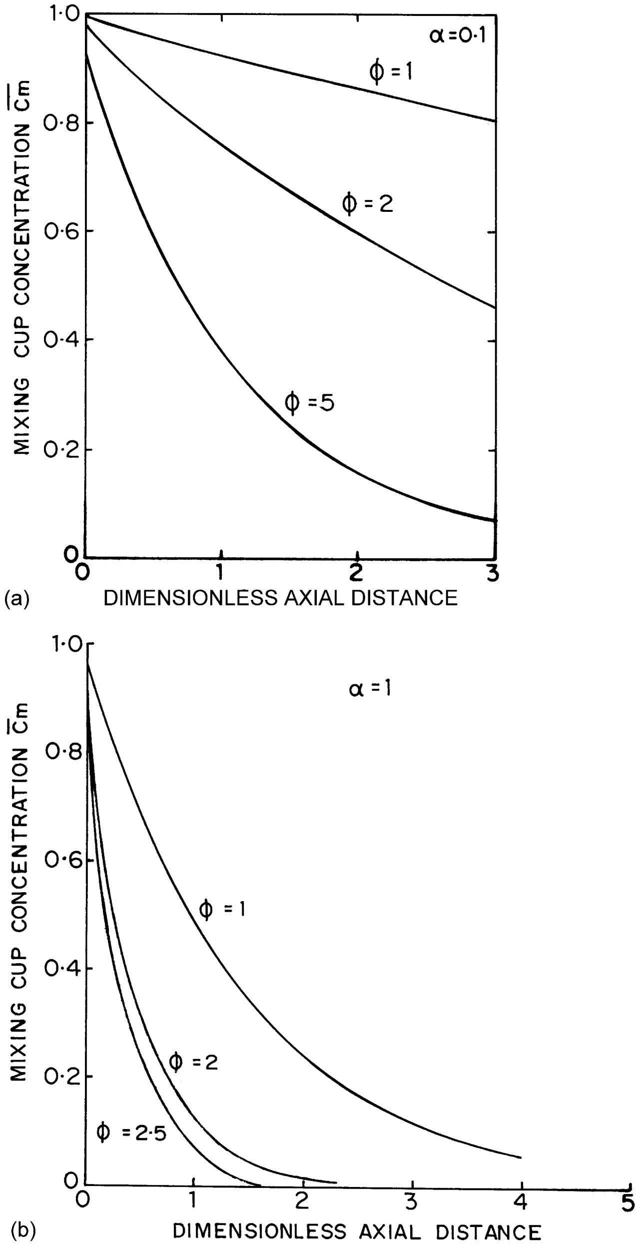

Fig. 4. (a) and (b) Mixing cup concentration profile in the reactor (M–M kinetics).

Rudisill and Levan [7] and Jayaraman and Kulkarni [12], while working with other reactor configurations. Using this approach the tube wall concentration is obtained in terms of a Volterra integral equation (Eq. (61)), which can be further discretized to give a nonlinear algebraic equation (Eq. (63)). Starting at the reactor inlet the effectiveness factor for the inlet substrate concentration can be obtained by solving the shell side equation. Using this value of the effectiveness factor the tube side nonlinear algebraic equation (Eq. (61)) can be solved by Newton–Raphson method to yield the tube wall concentration at the next axial location (this can be decided by a suitable choice of the step size in the axial direction). With this wall concentration the shell side equations are again solved to give the value of the effectiveness factor at the next axial location. This value of the effectiveness factor can now be used to update the tube wall concentration in the axial direction. By repeatedly switching between the tube side equation and the shell side equation the concentration profiles for the entire reactor length can be obtained. A representative plot of the mixing cup concentration profile for two values of α are shown in Fig. 4a and b. Such a procedure can be used to simulate the reactor for different values of the Thiele modulus and α, the parameter governing the ratio of the diffusion coefficients of the bulk liquid phase and the matrix phase. The algorithm developed in this work can be readily extended to other nonlinear kinetic expressions like substrate inhibition, product inhibition and for multi-substrate reactions.

5. Conclusions

Theoretical analysis of a membrane reactor packed with porous matrices, in which enzymes/cells have been localized, has been presented with a view to simulate the performance of the reactor. The results indicate that the overall performance of the reactor depends on the diffusion time scales and length scales of the bulk phase and the solid matrix phase in the reactor shell. Analytical solutions are possible for the first order reaction occurring in the reactor shell side, whereas, a formalism using Duhamel's theorem has been used to obtain solutions for Michaelis–Menten kinetics. Such an approach can be extended to solve other kinetic expressions. This novel reactor configuration can be advantageously used for conducting a large variety of chemical and biochemical reactions.

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