We are IntechOpen, the world's leading publisher of Open Access books Built by scientists, for scientists

6,900

186,000

200M

Downloads

154
Countries delivered to

Our authors are among the

TOP 1%

most cited scientists

12.2%

Contributors from top 500 universities



WEB OF SCIENCE

Selection of our books indexed in the Book Citation Index in Web of Science™ Core Collection (BKCI)

Interested in publishing with us? Contact book.department@intechopen.com

Numbers displayed above are based on latest data collected.

For more information visit www.intechopen.com



Mass Transfer through Catalytic Membrane Layer

Nagy Endre University of Pannonia, Research Institute of Chemical and Process Engineering Hungary

1. Introduction

The catalytic membrane reactor as a promising novel technology is widely recommended for carrying out heterogeneous reactions. A number of reactions have been investigated by means of this process, such as dehydrogenation of alkanes to alkenes, partial oxidation reactions using inorganic or organic peroxides, as well as partial hydrogenations, hydration, etc. As catalytic membrane reactors for these reactions, intrinsically catalytic membranes can be used (e.g. zeolite or metallic membranes) or membranes that have been made catalytic by dispersion or impregnation of catalytically active particles such as metallic complexes, metallic clusters or activated carbon, zeolite particles, etc. throughout dense polymeric- or inorganic membrane layers (Markano & Tsotsis, 2002). In the majority of the above experiments, the reactants are separated from each other by the catalytic membrane layer. In this case the reactants are absorbed into the catalytic membrane matrix and then transported by diffusion (and in special cases by convection) from the membrane interface into catalyst particles where they react. Mass transport limitation can be experienced with this method, which can also reduce selectivity. The application of a sweep gas on the permeate side dilutes the permeating component, thus increasing the chemical reaction gradient and the driving force for permeation (e.g. see Westermann and Melin, 2009). At the present time, the use of a flow-through catalytic membrane layer is recommended more frequently for catalytic reactions (Westermann and Melin, 2009). If the reactant mixture is forced to flow through the pores of a membrane which has been impregnated with catalyst, the intensive contact allows for high catalytic activity with negligible diffusive mass transport resistance. By means of convective flow the desired concentration level of reactants can be maintained and side reactions can often be avoided (see review by Julbe et al., 2001). When describing catalytic processes in a membrane reactor, therefore, the effect of convective flow should also be taken into account. Yamada et al., (1988) reported isomerization of 1-butene as the first application of a catalytic membrane as a flow-through reactor. This method has been used for a number of gas-phase and liquid-phase catalytic reactions such as VOC decomposition (Saracco & Specchia, 1995), photocatalytic oxidation (Maira et al., 2003), partial oxidation (Kobayashi et al., 2003), partial hydrogenation (Lange et a., 1998; Vincent & Gonzales, 2002; Schmidt et al., 2005) and hydrogenation of nitrate in water (Ilinitch et al.,

From a chemical engineering point of view, it is important to predict the mass transfer rate of the reactant entering the membrane layer from the upstream phase, and also to predict

the downstream mass transfer rate on the permeate side of the catalytic membrane as a function of the physico-chemical parameters. The outlet mass transfer rate should generally be avoided. The mathematical description of the mass transport enables the reader to choose the operating conditions in order to minimize the outlet mass transfer rate. If this transfer (permeation) rate is known as a function of the reaction rate constant, it can be substituted into the boundary conditions of the full-scale differential mass balance equations for the upstream and/or the downstream phases. Such kind of mass transfer equations can not be found in the literature, yet. For their description, two types of membrane reactors should generally be distinguished, namely intrinsically catalytic membrane and membrane layer with dispersed catalyst particle, either nanometer size or micrometer size catalyst particles. Basically, in order to describe the mass transfer rate, a heterogeneous model can be used for larger particles and/or a pseudo-homogeneous one for very fine catalyst particles (Nagy, 2007). Both approaches, namely the heterogeneous model for larger catalyst particles and the homogeneous one for submicron particles, will be applied for mass transfer through a catalytic membrane layer. Mathematical equations have been developed to describe the simultaneous effect of diffusive flow and convective flow and this paper analyzes mass transport and concentration distribution by applying the model developed.

Membrane bioreactor (MBR) technology is advancing rapidly around the world both in research and commercial applications (Strathman et al., 2006; Yang and Cicek, 2006; Giorno and Drioli, 2000; Marcano and Tsotsis, 2002). Integrating the properties of membranes with biological catalyst such as cells or enzymes forms the basis of an important new technology called membrane bioreactor. Membrane layer is especially useful for immobilizing whole cells (bacteria, yeast, mammalian and plant cells) (Brotherton and Chau, 1990; Sheldon and Small, 2005), bioactive molecules such as enzymes (Rios et al., 2007; Charcosset, 2006; Frazeres and Cabral, 2001) to produce wide variety of chemicals and substances. The main advantages of the membrane, especially the hollow fiber, bioreactor are the large specific surface area (internal and external surface of the membrane) for cell adhesion or enzyme immobilization; the ability to grow cells to high density; the possibility for simultaneous reaction and separation; relatively short diffusion path in the membrane layer; the presence of convective velocity through the membrane if it is necessary in order to avoid the nutrient limitation (Belfort, 1989; Piret and Cooney, 1991; Sardonini and DiBiasio, 1992). This work analyzes the mass transport through biocatalytic membrane layer, either live cells or enzymes, inoculated into the shell and immobilized within the membrane matrix or in a thin layer at the membrane matrix matrix-shell interface. Cells are either grown within the fibers with medium flow outside or across the fibers while wastes and desired products are removed or grown in the extracapillary space with medium flow through the fibers and supplied with oxygen and nutrients (Fig. 12 illustrates this situation). The performance of a hollow-fiber or sheet bioreactor is primarily determined by the momentum and mass transport rate (Calabro et al., 2002; Godongwana et al., 2007) of the key nutrients through the bio-catalytic membrane layer. Thus, the operating conditions (trans-membrane pressure, feed velocity), the physical properties of membrane (porosity, wall thickness, lumen radius, matrix structure, etc.) can considerably influence the performance of a bioreactor, the effectiveness of the reaction. The introduction of convective transport is crucial in overcoming diffusive mass transport limitation of nutrients (Nakajima and Cardoso, 1989) especially of the sparingly soluble oxygen. Several investigators modeled the mass transport through this biocatalyst layer, through enzyme membrane layer (Ferreira et al., 2001; Long et al., 2003; Belfort, 1989; Hossain and Do, 1989; Calabro et al., 2002; Waterland et al., 1975;

Salzman et al., 1999; Carvalho et al, 2000) or cell culture membrane layer (Melo and Oliveira, 2001; Brotherton and Chau, 1990, 1996; Piret and Cooney, 1991; Sardonini and Dibiasio, 1992; Lu et al., 2001; Schonberg and Belfort, 1987). These studies analyze both the mass transport through the membrane and the bulk phase concentration change. Against these detailed studies, there are not known mass transfer equations which define the mass transfer rate through a biocatalytic membrane layer, in closed forms as a function of the transport parameters as membrane Peclet number, reaction rate modulus as well as the Peclet number of the concentration boundary layer. These equations could then be replaced in the full-scale mass transfer models in order to predict the concentration distribution in the bulk liquid phase.

When someone knows the mass transfer rate through the membrane, these rate equations now can be put into the full-scale mass balance equation as boundary value to describe the concentration distribution on the lumen side, feed side or on the shell side, permeate side. The full-scale description of flow in crossflow filtration tubular membrane or in flat sheet membrane is also very often the object of investigations (Damak et al., 2004). A fluid dynamic description of free flows is usually easy to perform, and in a great majority of examples, the well known Navier-Stokes equations can be used to coupling Darcy's law and the Navier-Stokes equations (Mondor & Moresoli, 1999; Damak et al., 2004). A steady-state, laminar, incompressible, viscous and isothermal flow in a cylindrical tube with a permeable wall is considered. The Navier-Stokes equation and Darcy's law describe the transfer in the tube and in the porous wall, respectively.

2. Mass transfer through membrane reactor

Six membrane reactor concepts can be considered related to the catalysts location in the membrane modules (Seidel-Morgenstern, 2010). Topics of this paper are the concept when the catalyst particles are dispersed in the membrane matrix (the membrane serves an active contactor) or the membrane layer is intrinsically catalytic. This concept is illustrated in Fig. 1. The reactants are fed into the reactor from different sides and react within the membrane.

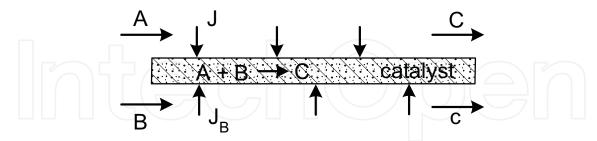


Fig. 1. Schematic illustration of catalytic membrane reactor

Before one can analyze the mass transport in the lumen or shell side of a capillary or on the two sides of a flat membrane, the outlet or inlet mass transfer rate at the membrane interface should be determined. A schematic diagram of the physical model and coordinate system is given in Fig. 2. The mass transfer rate depends strongly on the membrane properties, on the catalyst activity and the mass transfer resistance between the flowing fluid phase and membrane layer. This mass transfer rate should then be taken into account in the mass balance equation for the flowing fluid (liquid or gas) phase, on both sides of membrane reactor. This will be discussed in section 6.

The mass transport through a catalytic membrane layer can be diffusive (there is no transmembrane pressure difference between the two sides of the membrane layer) or diffusive+ convective transport. These two modes of flow will be discussed separately due to its different mathematical treatments in order to get the transfer rate.

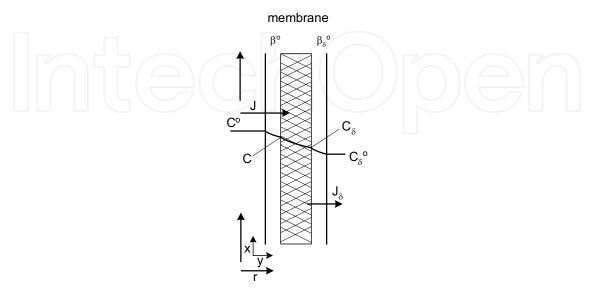


Fig. 2. Illustration of the mass transfer through a membrane reactor

The other important classification of the reactors that, as it was mentioned, the membrane reactor can intrinsically catalytic or it is made catalytic by dispersed catalyst particles distributed uniformly in the membrane matrix. In this latter case two types of mathematical model can be used (Nagy, 2007), namely pseudo-homogeneous or heterogeneous models, depending on the catalyst particle size. It was shown by Nagy (2007) if the size of catalyst particles less than a micron, the simpler homogeneous model can be recommended, in other wise, the heterogeneous model should be applied.

The differential mass balance equation can generally be given by the following equation for the catalytic membrane layer with various geometries, perpendicular to the membrane interface, applying cylindrical coordinate (Ferreira et al., 2001):

$$\frac{\partial}{\partial r} \left(D_m \frac{\partial c}{\partial r} \right) + \frac{(p+1)}{r} \frac{\partial (D_m c)}{\partial r} - \frac{\partial (\upsilon c)}{\partial r} - Q = \frac{\partial c}{\partial t}$$
 (1a)

where p denotes a geometrical factor with values of 0 for cylindrical coordinate and -1 for rectangular membranes. The membrane concentration, C is given here in a unit of measure of gmol/m³. This can be easily obtained by means of the usually applied in the e.g. g/g unit of measure with the equation of C=wp/M, where w concentration in kg/kg, ρ – membrane density, kg/m³, M-molar weight, kg/mol. The most often recommended mass balance equation (Marcano & Tsotsis, 2002), in dimensionless form, for membrane reactor is as (R=r/R_o; C=c/c°):

$$\frac{\upsilon R_o^2}{D_m} \frac{\partial C}{\partial X} = \frac{1}{R} \frac{\partial}{\partial R} \left(\frac{\partial C}{\partial R} \right) - Q^*$$
 (1b)

where Q* reaction term given in dimensionless form. The boundary conditions are as:

$$C=1$$
 at $X=0$, for all R (2a)

$$\frac{\partial C}{\partial R} = 0$$
 at R=0, for all X (2b)

$$\nu C - D \frac{dC}{\partial R} = J$$
 at R=1, for all X (2c)

The value of the mass transfer rate through the membrane, J will be shown in the next sections under different conditions. From eq. 1, the mass balance equation is easy to get for flat sheet membrane.

2.1 Diffusive mass transport with intrinsic catalytic layer or with fine catalytic particles

In both cases the membrane matrix is regarded as a continuous phase for the mass transport. Assumptions, made for expression of the differential mass balance equation to the catalytic membrane layer, are:

- Reaction occurs at every position within the catalyst layer;
- Mass transport through the catalyst layer occurs by diffusion;
- The partitioning of the components (substrate, product) is taken into account (thus, $CH_m=C_m^*$ where C_m^* denotes membrane concentration on the feed interface; see Fig. 2);
- The mass transport parameters (diffusion coefficient, partitioning coefficient) are constant;
- The effect of the external mass transfer resistance should also be taken into account;
- The mass transport is steady-state and one-dimensional;

In case of dispersed catalyst particles they are uniformly distributed and they are very fine particles with size less than 1 µm, i.e. they are nanometer sized particles. It is assumed that catalyst particles are placed in every differential volume element of the membrane reactor. The reactant firstly enters in the membrane layer and from that it enters into the catalyst particles where the reaction of particles is porous as e.g. active carbon, zeolite (Vital et al., 2001) occurs or it enters onto the particle interface and reacts [particle is nonporous as e.g. metal cluster, (Vancelecom & Jacobs, 2000)]. Consequently, the mass transfer rate into the catalyst particles has to be defined first. In this case, the whole amount of the reactant transported in or on the catalyst particle will be reacted. Then this term should be placed into the mass balance equation of the catalytic membrane layer as a source term. Thus, the differential mass balance equation for intrinsic membrane and membrane with dispersed nanosized particles differ only by their source term. The cylindrical effect can only be significant when the thickness of a capillary membrane can be compared to the internal radius of the capillary tube as it was shown by Nagy (2006). On the other hand, the application of cylindrical coordinate hinders the analytical solution for first or zero-order reactions as well. Thus, the basic equations will be shown here for plane interface and in the section 5 an analytical approach will be presented for cylindrical tube as well.

2.1.1 Mass transfer accompanied by first-order reaction

Herewith first the reaction source term will be defined indifferent cases, namely in cases of intrinsically catalytic membrane and membrane with dispersed catalytic particles and the solution of the differential mass balance equation under different boundary conditions.

2.1.1.1 Reaction terms

Intrinsically catalytic membrane; this is well known in literature ($\Phi = \sqrt{k_1 c_1^o}$):

$$Q = k_1 c_1^{\circ} C \equiv \Phi^2 C \tag{3}$$

Catalyst with dispersed particles, reaction takes place inside of the porous particles; For catalytic membrane with dispersed nanometer size particles, the mass transfer rate into the spherical catalyst particle has to be defined. The internal specific mass transfer rate in spherical particles, for steady-state conditions and when the mass transport accompanied by first-order chemical reaction can be given as follows (Nagy & Moser, 1995):

$$j = \beta_p C_p^* \tag{4}$$

where

$$\beta_p = \frac{D_p}{R_p} \left(\frac{Ha_p}{\tanh(Ha_p)} - 1 \right)$$
 (5a)

and

$$Ha_p = \sqrt{\frac{k_1 R_p^2}{D_p}}$$

The external mass transfer resistance, through the catalyst particle depends on the diffusion boundary layer thickness, δ_p . The value of δ_p could be estimated from the distance of particles from each other (Nagy & Moser, 1995). Namely, its value is limited by the neighboring particles, thus, the value of β_p will be slightly higher than that follows from the well known equation of $2 = \beta_p^o d_p / D_m$, where the value of δ_p is supposed to be infinite. Thus, one can obtain (Nagy et al., 1989):

 $\beta_p^o = \frac{2D_m}{d_p} + \frac{D_m}{\delta_p} \tag{5b}$

where

$$\delta_p = \frac{h - d_p}{2}$$

From eqs 4 and 5 one can obtain for the mass transfer rate with the overall mass transfer resistance:

$$j = \beta_{tot}c^{o}C = c^{o}\frac{C}{\frac{1}{\beta_{p}^{o}} + \frac{1}{H\beta_{p}}}$$

$$\tag{6}$$

Accordingly, the Φ value in eq. 3 can be expressed as follows (Nagy et al., 1989):

$$\Phi = \sqrt{\frac{\omega \delta_m^2}{1 - \varepsilon} \beta_{tot}} \tag{7}$$

Reaction occurs on the interface of the catalytic particles (Nagy, 2007). It often might occur that the chemical reaction takes place on the interface of the particles, e.g, in cases of metallic clusters, the diffusion inside the dense particles is negligibly. Assuming the Henry's sorption isotherm of the reacting component onto the spherical catalytic surface (CH_f=q_f), applying $DdC/dr = k_f H_f C$ boundary condition at the catalyst's interface, at r=R_p, the Φ reaction modulus can be given according to eq. (7) with the following β_{sum} value:

$$\beta_{\delta} = \frac{1}{\frac{1}{\beta_p^o} + \frac{1}{k_f H_f}} \tag{8}$$

where k_f is the interface reaction rate constant. The above model is obviously a simplified one.

2.1.1.2 Mass transfer rates

The differential mass balance equation for the reactant entering the catalytic membrane layer is as follows in dimensionless form:

$$\frac{d^2C}{dY^2} - \Phi^2C = 0 \tag{9}$$

Solution of eq. 9 is well known:

$$C = Te^{\Phi Y} + Se^{-\Phi Y} \tag{10}$$

For the sake of generalization, in the boundary conditions you should take into account the external mass transfer resistance on both sides of the membrane, though it should be noted that the role of the β_{δ}^{o} will be gradually diminish with the increase of the reaction rate. At the end of this subsection the limiting cases will also be briefly given. Thus:

$$Y=0 \qquad \beta^{o}(1-C) = -\frac{D_{m}}{\delta_{m}} \frac{dC}{dY}\Big|_{Y=0}$$
 (11)

Y=1
$$\beta_{\delta}^{o}\left(C_{\delta}-C_{\delta}^{o}\right) = -\frac{D_{m}}{\delta_{m}}\frac{dC}{dY}\Big|_{Y=1}$$
 (12)

The mass transfer rate on the upstream side of the membrane can be given as follows (Nagy, 2007):

$$J = \beta_m H_m c^o \left(1 - T C_\delta^o \right) \tag{13}$$

with

$$\beta_{m} = \beta_{m}^{o} \Phi \frac{\left(1 + \frac{\beta_{m}^{o} \Phi H_{m}}{\beta_{\delta}^{o}} \tanh \Phi\right)}{\left(1 + \frac{\left[\beta_{m}^{o} \Phi\right]^{2} H_{m}^{2}}{\beta^{o} \beta_{\delta}^{o}}\right) \tanh \Phi + \beta_{m}^{o} \Phi\left(\frac{H_{m}}{\beta^{o}} + \frac{H_{m}}{\beta_{\delta}^{o}}\right)}$$

$$(14)$$

and

$$T = \frac{1}{\cosh \Phi \left(1 + \frac{\beta_m^o \Phi H_m \tanh \Phi}{\beta_\delta^o} \right)}$$
 (15)

with

$$\beta^o = \frac{D_L}{\delta_L}; \quad \beta_m^o = \frac{D_m}{\delta_m}$$

Similarly, the mass transfer rate for the downstream side of the membrane, at Y=1:

$$J_{\delta} = \beta_{\delta} H_{m} c^{o} \left(1 - \cosh \Phi \left\langle \tanh \Phi + \frac{\beta_{m}^{o} \Phi H_{m}}{\beta^{o}} \right\rangle C_{\delta}^{o} \right)$$
 (14)

with

$$\beta_{\delta} = \frac{\beta_{m}^{o}\Phi}{\cosh\Phi} \frac{1}{\tanh\Phi \left(1 + \frac{\left(\beta_{m}^{o}\Phi\right)^{2}}{\beta^{o}\beta_{\delta}^{o}}H_{m}^{2}\right) + \left(\frac{H_{m}}{\beta^{o}} + \frac{H_{m}}{\beta_{\delta}^{o}}\right)\beta_{m}^{o}\Phi}$$
(15)

Limiting cases; The transfer rate without external mass transfer resistances, namely when $\beta^o \to \infty$ and $\beta^o_\delta \to \infty$, can easily be obtained from eq. 13 as limiting case as:

$$J = \frac{\beta_m^o \Phi c^o}{\tanh \Phi} \left(1 - \frac{C_\delta^o}{\cosh \Phi} \right) \tag{16}$$

Eq. 16 is a well known mass transfer equation for liquid mass transfer accompanied by first-order reaction. The mass transfer can similarly be obtained rate for the case when the outlet concentration is zero, and, $\beta_{\delta}^{o} \rightarrow \infty$:

$$J = \beta_{tot}^o c^o \tag{17}$$

where

$$\beta_{tot}^{o} = \frac{1}{\frac{\tanh \Phi}{H_m \Phi \beta_m^0} + \frac{1}{\beta^0}}$$
(18)

To avoid the outlet flow of reactant is an important requirement for the membrane reactors. For it the operating conditions should be chosen rightly.

2.1.2 Mass transfer accompanied by zero-order reaction

In this case the reaction rate is independent of the concentration of reactant in the membrane layer. The differential mass balance equation can be given as:

$$\frac{d^2C}{dY^2} = \Phi^2 \tag{19}$$

The value of Φ can be given for intrinsically catalytic membrane as:

$$\Phi = \sqrt{\frac{k_0 \delta_m^2}{D_m c^o}} \tag{20}$$

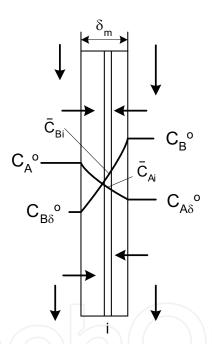


Fig. 3. Illustration of the concentrations for second-order reaction

The case of dispersed catalyst particles in the membrane layer is not discussed here because it unimportance for membrane reactor. For the solution of the eq. 19 let us use the following boundary conditions:

at Y=0
$$\beta^{\circ}(1-C) = -\frac{D_m}{\delta_m} \frac{dC}{dY}\Big|_{Y=0}$$
 (21)

at Y=1
$$C = HC_{\delta}^{o}$$
 (22)

The mass transfer resistance on the outlet side has not importance in that case because the concentration rapidly decreases down to zero, thus does exist outlet mass transfer in a

narrow reaction rate regime, only. After solution, the concentration distribution can be given as:

$$C = \frac{J}{\beta_{m}^{o}} Y + \frac{\Phi^{2}}{2} + B \tag{23}$$

where

$$B = \frac{C_{\delta}^{o} - \Phi^{2} / 2 + \beta^{o} / \beta_{m}^{o}}{1 + \beta^{o} / (\beta_{m}^{o} H)}$$
(24)

The mass transfer rate can be given as:

$$J = \beta_m^o c^o \left(\frac{H - HC_\delta^o + \Phi^2 / 2}{H + \beta^o / \beta_m^o} \right)$$
 (25)

From eq. 25, the well known expression of mass transfer rate without chemical reaction can easily be obtained.

2.1.3. Mass transfer accompanied by second-order reaction

It is assumed that the reagents (component A and B) are fed on the both sides of the membrane reactor and they are diffusing through the membrane layer counter-currently (Fig. 3). The reaction term can be given for intrinsically catalytic membrane as follows:

$$Q = k_2 c_A^0 c_B^0 C_A C_B \tag{26}$$

Substituting the reaction term into eq. (1) for e.g. the A component and plane interface as well as steady-state condition (D_{mA} is constant) one can get:

$$D_{mA} \frac{d^2 C_A}{dy^2} - k_2 c_A^o c_B^o C_A C_B = 0$$
 (27)

This equation can be solved either by numerical method or an analytical approach can be developed. Such an analytical approach is given in details in Appendix. The essential of this method that the membrane layer is divided into N very thin sub-layer and the concentration of one of the two components is considered to be constant in this sub-layer (see Fig. 3 and Fig. 13). Thus, one can get a second-order differential equation with linear source term that can be solved analytically. In dimensionless form it is for the ith sub-layer as:

$$\frac{d^{2}C_{A}}{dY^{2}} - \Phi_{Ai}^{2}C_{A} = 0 \quad \text{for} \quad Y_{i-1} \le Y \le Y_{i}$$
 (28)

where

$$\Phi_{Ai} = \sqrt{\frac{k_2 \delta_m^2 c_A^o c_B^o \overline{C}_B}{D_{mA}}}$$

where \bar{C}_B denotes the average concentration of B component in the ith sub-layer. Solution of eq. 28 is well known (see eq. 10). The general solution for every sub-layer has two parameters that should be determined by the suitable boundary conditions (see Appendix):

at
$$Y=0$$
 C=1 (29)

at
$$Y_{i-1} \le Y \le Y_i$$

$$\frac{dC_A}{dY}\Big|_{Y_i^-} = \frac{dC_A}{dY}\Big|_{Y_i^+} \text{ with } i=1,2,...,N$$
 (30)
at $Y_{i-1} \le Y \le Y_i$
$$C_{A\Big|Y_i^-} = C_{A\Big|Y_i^+} \text{ with } i=1,2,...,N$$

at
$$Y_{i-1} \le Y \le Y_i$$
 $C_{A|Y_i^-} = C_{A|Y_i^+}$ with i=1,2,...,N (31)

at y=1
$$C_A = C_{A\delta}^o \tag{32}$$

After solution of the N differential equation with 2N parameters to be determined the T₁ and S_1 parameters for the first sub-layer can be obtained as (ΔY is the thickness of the sublayers):

$$T_{1} = -\frac{1}{2\xi_{N}^{O}\cosh(\Phi_{A1}\Delta Y)} \left(\xi_{N}^{T} - \frac{C_{A\delta}^{o}}{\prod_{i=2}^{N}\cosh(\Phi_{Ai}\Delta Y)}\right)$$
(33)

and

$$S_{1} = \frac{1}{2\xi_{N}^{O}\cosh(\Phi_{A1}\Delta Y)} \left(\xi_{N}^{S} - \frac{C_{A\delta}^{o}}{\prod_{i=2}^{N}\cosh(\Phi_{Ai}\Delta Y)}\right)$$
(34)

Knowing the T_1 and S_1 the other parameters, namely T_i and S_i (i=2,3,...,N) can be easily be calculated by means of the internal boundary conditions given by eqs. 30 and 31 from starting from T_2 and S_2 up to T_N and S_N .

After differentiating eq. 10 and applying it for the first sub-layer, the mass transfer rate of component A can be expressed as:

$$J = \frac{D_m \Phi_{A1} c_A^o}{\delta_m} \frac{\xi_N^S - \xi_N^T}{2\xi_N^O \cosh(\Phi_{A1} \Delta Y)} \left(1 - \frac{C_{A\delta}^o}{\left(\xi_N^S - \xi_N^T\right) \prod_{j=2}^N \cosh(\Phi_{Aj} \Delta Y)} \right)$$
(35)

where

$$\xi_i^j = \xi_{i-1}^j + \kappa_{i-1}^j \frac{\tanh(\Phi_{Ai}\Delta Y)}{z_i} \text{ for i=2,3,...,N and j=S,T,O}$$
 (36)

and

$$\kappa_i^j = \xi_{i-1}^j \tanh\left(\Phi_{Ai}\Delta Y\right) + \frac{\kappa_{i-1}^j}{z_i} \text{ for i=2,3,...,N and j=S,T,O}$$
(37)

The starting values of ξ_1^j and κ_1^j are as follows:

$$\xi_1^T = e^{-\Phi_{A1}\Delta Y}$$
 $\xi_1^S = e^{\Phi_{A1}\Delta Y}$ $\xi_1^O = \tanh(\Phi_{A1}\Delta Y)$

and

$$\kappa_1^T = -e^{-\Phi_{A1}\Delta Y} \quad \kappa_1^S = e^{\Phi_{A1}\Delta Y} \quad \kappa_1^O = 1$$

Obviously, in order to get the inlet mass transfer rate of component A, the concentration distribution of component B is needed. Thus, for prediction of the J value the concentration of component B has to be known. It is easy to learn that trial-error method should be used to get alternately the component concentrations. Steps of calculation of concentration of both components can be as follows:

- 1. Starting concentration distribution, e.g. for component B should be given and one calculates the concentration distribution of component A;
- 2. The indices of sub-layer of A component have to be changed adjusted them to that of B started from the permeate side of membrane, i.e. at Y=1, thus, i subscript of A_i should be replaced by N+1-i;
- 3. Now applying the previously calculated averaged A_i (\bar{A}_i), one can predict the concentration distribution of component B, using eqs. 33 to 37, adapted them to component B;
- 4. These three steps should be repeated until concentrations do not change anymore;

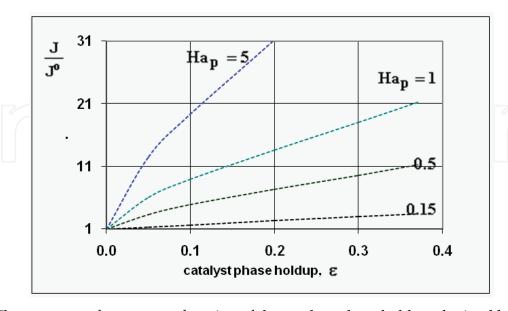


Fig. 4. The mass transfer rate a s a function of the catalyst phase holdup obtained by the pseudo-homogeneous model (H_m=H=1; D_m=1 x 10⁻¹⁰ m²/s; $C_{\delta}^{o}=0$; $\beta^{o}=\beta_{\delta}^{o}\to\infty$; d_p=2 µm; δ_{m} =30 µm)

2.1.4 Analysis of the mass transport

The detailed discussion of the mass transport through a membrane reactor is not a target of this paper. Applying the equations for mass transfer rate or for concentration distribution presented the reactor performance is easy to calculate. Only a typical figure will be shown in this section. Fig. 4 illustrates the effect of the catalyst holdup and the reaction modulus on the mass transfer rate. Detailed analysis is given in Nagy's paper (2007). Similar results can be obtained by zero-order reaction though its effect is somewhat stronger because its independency of concentration (Nagy, 2007). The concentration of the reactor rapidly decreases down to zero even at rather low reaction rate coefficient. Thus the role of the convective velocity should have got careful attention.

Normally, 3-5 recalculations of concentrations are enough to get the correct results.

2.2 Diffuive+convective mass transport with intrinsic catalytic layer or with fine catalytic particles

Convective mass transport can take place if transmembrane pressure difference exists between the two membrane sides. Recently it was proved in the literature (Ilinitch, 2000, Nagy, 2007) that the presence of convective flow can improve the efficiency of the membrane reactor. Thus, the study of the mass transport in presence of convective mass flow can be important in order to predict the reaction process. On the otherwise, the use of convective flow is rather rare, because the aim is mostly to minimize the outlet rate of the reactant on the permeate side. The source terms of this case are the same as it was showed in subsection 2.1.

2.2.1 Mass transport accompanied by first-order reaction

The differential mass balance equation for the polymeric or macroporous ceramic catalytic membrane layer, for steady-state, taking both diffusive and convective flow into account, can be given as:

$$\frac{d^2C}{dY^2} - Pe_m \frac{dC}{dY} - \Phi^2 C = 0 \tag{38}$$

where

$$Pe_m = \frac{\upsilon \delta_m}{D_m}; \quad \Phi = \sqrt{\frac{\omega \delta_m^2}{D_m (1 - \varepsilon)} \beta_{tot}}$$

where υ denotes the convective velocity, D_m is the diffusion coefficient of the membrane, and δ_m is the membrane thickness.

$$\tilde{C} = Ce^{-Pe_m Y/2} \tag{39}$$

Introducing a new variable, \tilde{C} (eq. 39) the following differential equation is obtained from eq. 38):

$$\frac{d^2\tilde{C}}{dY^2} - \Theta^2\tilde{C} = 0 \tag{40}$$

where

$$\Theta = \sqrt{\frac{Pe_m^2}{4} + \Phi^2}$$

The general solution of eq. 40 is well known, so the concentration distribution in the catalytic membrane layer can be given as follows:

The inlet and the outlet mass transfer rate can easily be expressed by means of eq. (41). The overall inlet mass transfer rate, namely the sum of the diffusive and convective mass transfer rates, is given by:

$$J = \nu C \Big|_{Y=0} - \frac{D_m}{\delta_m} \frac{dC}{dY} \Big|_{Y=0} = \beta_m^o \left(\lambda T + \tilde{\lambda} S \right)$$
 (42)

The outlet mass transfer rate is obtained in a similar way to eq. (42) for X=1:

$$J_{\delta} = \beta_m^o \left(\tilde{\lambda} T e^{\lambda} + \lambda S e^{\tilde{\lambda}} \right) \tag{43}$$

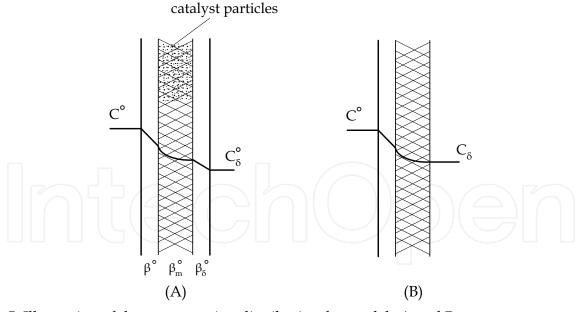


Fig. 5. Illustration of the concentration distribution for models A and B.

The value of parameters T and S can be determined from the boundary conditions. For the sake of generality, two models, namely model A and model B, will be distinguished according to Figure 5 (for details see Nagy, 2010). The essential difference between the models is that, in case of model A, there is a sweeping phase that can remove the transported component from the downstream side providing the low concentration of the reacted component in the outlet phase and due to it, high diffusive mass transfer rate. There

is no sweep phase in case of model B, thus the outlet phase is moving from the membrane due to the lower pressure on the permeate side.

Model A. In this case, due to the effect of the sweeping phase, the external mass transfer resistance on both sides of the membrane should be taken into account in the boundary conditions, though the role of β^o_δ is gradually diminished as the catalytic reaction rate increases. The concentration distribution in the catalytic membrane when applying a sweep phase on the two sides of the membrane, as well is illustrated in Fig. 5a. On the upper part of the catalytic membrane layer, in Fig. 5a, the fine catalyst particles are illustrated with black dots. It is assumed that these particles are homogeneously distributed in the membrane matrix. Due to sweeping phase, the concentration of the bulk phase on the permeate side may be lower than that on the membrane interface. The boundary conditions can be given for that case as:

$$\nu C + \beta^{\circ} \left(C^{\circ} - C \right) = J \text{ at Y=0}$$
 (44)

$$\nu C_{\delta} + \beta_{\delta}^{o} \left(C_{\delta} - C_{\delta}^{o} \right) = J_{\delta} \text{ at Y=1}$$

$$\tag{45}$$

Boundary conditions given by eqs. (44) and (45) are only valid in two phase flows. Where C^o_δ denotes the concentration on the downstream side, β^o and β^o_δ are mass transfer coefficients in the continuous phase, β^o_m the membrane mass transfer coefficient ($\beta^o_m = D_m / \delta_m$), H_m denotes the distribution coefficient between the continuous phase and the membrane phase. The solution of the algebraic equations obtained, applying eqs. 42 to 45, can be received by means of known mathematical manipulations. Thus, the values of T and S obtained are as follows:

$$T = -\frac{\beta^o \varphi_2 C^o - \beta^o_\delta \varphi_4 C^o_\delta}{\varphi_2 \varphi_3 - \varphi_1 \varphi_4} \frac{1}{\beta^o_m}$$

$$\tag{46}$$

and

 $S = \frac{\beta^{o} \varphi_{1} C^{o} + \beta^{o}_{\delta} \varphi_{3} C^{o}_{\delta}}{\varphi_{2} \varphi_{3} - \varphi_{1} \varphi_{4}} \frac{1}{\beta^{o}_{m}}$ $\tag{47}$

where

$$\varphi_{1} = \left(\frac{Pe_{m}}{H_{m}} + \frac{\beta_{\delta}^{o}}{\beta_{m}^{o}H_{m}} - \tilde{\lambda}\right)e^{\lambda}; \quad \varphi_{2} = \left(\frac{Pe_{m}}{H_{m}} + \frac{\beta_{\delta}^{o}}{\beta_{m}^{o}H_{m}} - \lambda\right)e^{\tilde{\lambda}};$$

$$\varphi_3 = \frac{Pe_m}{H_m} - \frac{\beta^o}{\beta_m^o H_m} - \tilde{\lambda} ; \quad \varphi_4 = \frac{Pe_m}{H_m} - \frac{\beta^o}{\beta_m^o H_m} - \lambda ;$$

An important limiting case should also be mentioned, namely the case when the external diffusive mass transfer resistances on both sides of membrane can be neglected, i.e. when $\beta^o \to \infty$ and $\beta^o_\delta \to \infty$. For that case the concentration distribution and the inlet mass transfer rate can be expressed by eqs. 48 and 49, respectively.

$$C = \frac{e^{Pe_m(Y-1)/2}}{\sinh\Theta} H_m \left\{ C^o e^{Pe_m/2} \sinh\left[\Theta(1-Y)\right] + C^o_\delta \sinh\left(\Theta Y\right) \right\}$$
(48)

$$J = \beta_m \left(C^o - \frac{\Theta}{e^{Pe_m/2} \left[Pe_m \sinh \Theta / 2 + \Theta \cosh \Theta \right]} C^o_{\delta} \right)$$
 (49)

with

$$\beta_{m} = \frac{\beta_{m}^{o} H_{m} \left(Pe_{m} \tanh \Theta / 2 + \Theta \right)}{\tanh \Theta}$$

An important limiting case when the outlet concentration is zero, i.e. $C^o_\delta = 0$, accordingly the mass transfer rate is as $(\beta^o_\delta \to \infty)$:

$$J = \frac{2\beta^{o}\Theta c^{o}}{\beta_{m}^{o} \left(Pe_{m} - 1\right)\left(e^{-2\Theta} - 1\right) - \left(\tilde{\lambda}e^{-2\Theta} - \lambda\right)}$$

$$(50)$$

Model B. For the convective flow catalytic membrane reactor operating in another mode, for instance in dead-end mode as in Figure 4b, the boundary condition on the permeate side of the membrane should be changed. In this case the concentration of the permeate phase does not change during its transport from the membrane interface. If there is no sweeping phase on the downstream side then the correct boundary conditions will be as:

$$\nu C + \beta^{\circ} \left(C^{\circ} - C \right) = J \text{ at } Y = 0$$
 (51)

$$\nu C_{\delta} = J_{\delta} \text{ at Y=1}$$
 (52)

After solution one can get as:

$$T = -\frac{\beta_1^o \varphi_2 C^o}{\varphi_2 \varphi_3 - \varphi_1 \varphi_4} \frac{1}{\beta_m^o}$$
 (53)

$$S = \frac{\beta^o \varphi_1 C^o}{\varphi_2 \varphi_3 - \varphi_1 \varphi_4} \frac{1}{\beta_m^o}$$
 (54)

where

$$\varphi_1 = \left(\frac{Pe_m}{H_m} - \tilde{\lambda}\right)e^{\lambda} \quad \varphi_2 = \left(\frac{Pe_m}{H_m} - \lambda\right)e^{\tilde{\lambda}}$$

The values of φ_3 and φ_4 are the same as they are given after eq. 47.

2.2.2 Mass transport accompanied by zero-order reaction

The effect of the zero-order reaction will be discussed here for intrinsically catalytic membrane layer, only. This reaction has no important role in the case of membrane reactor.

The differential mass balance equation to be solved is as:

$$D_{m} \frac{d^{2}C}{dy^{2}} - \upsilon \frac{dC}{dy} - k_{0} = 0$$
 (55)

Similarly to eqs. 19, the differential mass balance equation for the catalytic membrane can be given as:

$$\frac{d^2C}{dY^2} - Pe_m \frac{dC}{dY} = \Phi^2 \tag{56}$$

where

$$\Phi = \sqrt{\frac{k_0 \delta_m^2}{D_m c^o}}$$

Look at first the solution with the following boundary conditions:

$$Y=0$$
 then $C=1$ (57a)

Y=1 then
$$C = C_{\delta}^{o}$$
 (57b)

The general solution of Eq. (56) is as:

$$C = T_m e^{Pe_m Y} - \frac{\Phi^2}{Pe_m} Y + Q_m \tag{58}$$

Applying the boundary conditions [Eqs. (57a) and (57b] one can get:

$$C = \frac{e^{Pe_m Y/2}}{\sinh(Pe_m/2)} \left\{ C_b \left(\sinh\left[\frac{Pe_m}{2}(1-Y)\right] + S \right) + e^{-Pe_m/2} \sinh\left(\frac{Pe_m Y}{2}\right) C_p \right\}$$
(59)

The mass transfer rate can be given as:

$$J = \beta_m c^o \left(1 - T C_\delta^o \right) \tag{60}$$

where

$$\beta_m = \beta_m^o c^o \left(1 + \frac{\Phi^2}{Pe_m^2} \right); \quad T = \frac{e^{-Pe_m}}{1 + \Phi^2 / Pe_m^2}$$

The outlet mass transfer rate should also be given:

$$J_{\delta} = \beta_m^o \alpha_{\delta} c^o \left(1 - C_{\delta}^o \frac{e^{-Pe_m}}{\alpha_{\delta}} \right)$$
 (61)

where

$$\alpha_{\delta} = 1 - \frac{\Phi^{2}}{Pe_{m}^{2}} e^{-Pe_{m}/2} \left(1 - e^{Pe_{m}} \left[1 - Pe_{m} \right] \right) = 1 - \frac{\Phi^{2}}{Pe_{m}^{2}} \left(Pe_{m} - 2e^{-Pe_{m}/2} \sinh\left(\frac{Pe_{m}}{2}\right) \right)$$
(62)

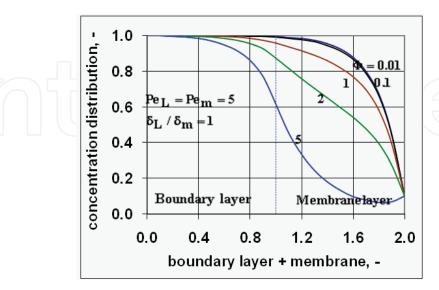


Fig. 6. The effect of first-order chemical reaction on the concentration distribution in the two layers at different values of reaction modulus ($C^o_\delta = 0.1$; $\beta^o_L = \beta^o_m = 1x10^{-4} \, m \, / \, s$)

Now look at the solution with mass transfer resistance in the feed phase, thus, the boundary condition for Y=0 will be as:

$$\nu C + \beta^{o} \left(C^{o} - C \right) = J \tag{63}$$

The physical mass transfer rate through the boundary layer can be expressed, in case of convective + diffusive flows as follows (Nagy & Kulcsar, 2009):

$$J^{o} = \beta^{o} c^{o} \left(1 - e^{-Pe_{L}} C \right) \tag{64}$$

with

$$\beta^{o} = \frac{D_{L}}{\delta_{L}} P e_{L} \frac{e^{P e_{L}}}{e^{P e_{L} - 1}} \tag{65}$$

whore

$$Pe_L = \frac{\upsilon \delta_L}{D_L}$$

Applying the well known two-layer theory the mass transfer rate can be given by means of the overall mass transfer resistance applying eqs. (60) and (64), the two mass transfer rate are equal to each other, as follows

$$J = \beta_{tot}c^{o}\left(1 - Te^{-Pe_{L}}C_{\delta}^{o} / H_{m}\right)$$

$$\tag{66}$$

with

$$\beta_{tot} = \frac{1}{\frac{1}{\beta^o} + \frac{H_m e^{-Pe_L}}{\beta_m}} \tag{67}$$

The values of β^{o} and β_{m} are defined in eqs. 65 and 60.

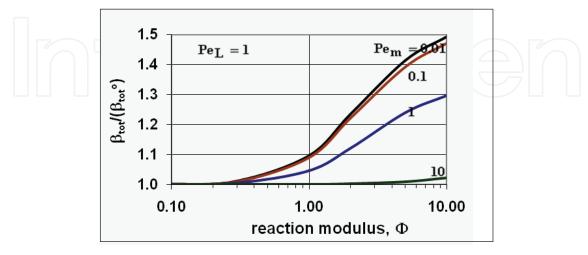


Fig. 7. The overall mass transfer coefficient as a function of first-order reaction rate constant at different values of Pe_m number $\beta_L^o = \beta_m^o = 1x10^{-4} m / s$

2.2.3 Analysis of the mass transport

The simultaneous effect of the convective flow and the reaction rate is less intensively investigated (Nagy, 2009a, 2009b, Nagy & Kulcsár, 2009; Nagy & Borbély, 2009). The importance of convective velocity can essentially improve the efficiency of the membrane reactor (Ilinitch, 2000; Nagy, 2010). Let us show the concentration distribution in case of first-order reaction (Fig. 6). The Φ reaction modulus can significantly lower the concentration in the phases. At larger modulus the curve change from concave one to convex one through an a inflexion point. The dimensionless outlet concentration was chosen to be 0.1. The concentration can decrease below this value indicating that $C_{\delta}^{o} = 0.1$ can maintain only when there is inlet flow of reactant on the permeate side. Obviously, this is not a real case. The effect of the first-order chemical reaction on the mass transfer coefficient is illustrated at different, increasing values of the Pe_m and with external resistance (Fig. 7). Obviously, the mass transfer rate will have limiting value due to the increasing effect of the external mass transfer resistance when the reaction rate increases. Description of the reduction of aqueous nitrates applying mono- and bimetallic, palladium-copper catalysts impregnated in γ-Al₂O₃ support layers is discussed by Nagy (2010) applying the mass transfer rate developed.

2.3 Mass transfer with microsized, dispersed, catalyst particles: applying the heterogeneous model

Mostly, depending also on the membrane thickness, when particles are falling into the micrometer size regime, the internal mass transport mechanism, inside of catalyst particles, has to be taken into account. A simple physical model could be applied for the description the process in this case, as it is schematically illustrated in Fig. 8. The gas (or liquid) reactant enters first the catalytic membrane layer and then diffuses to the first catalytic particle,

perpendicular to the membrane interface. The chemical reaction, namely a first-order, irreversible chemical reaction, takes place in the catalyst particles only. It is assumed that the concentration of the organic reactant should be much higher in a hydrophobic polymer membrane than that of the reactant investigated, e.g. peroxides, oxygen or hydrogen, etc. Then the unreacted reactant diffuses through the first, catalytic particle to its

other side and enters again the polymer membrane matrix and so on (the route of this mass transfer process is illustrated by an arrow denoted by J in Fig. 8). This diffusion path exists only for the heterogeneous part of the membrane interface (which is the projection of the cubic catalyst particle onto the membrane interface). There can be a portion of membrane interface, that is the so called homogeneous part of the interface, where the diffusing reactant does not cross any catalyst particle (this mass stream is denoted by Jo in Fig. 8). This also affects the resultant mass transfer rate. The assumed cubic (Yawalkar et al., 2001, Mehra, 1999, Nagy, 2002) catalyst particles are supposed to be uniformly distributed in the polymer membrane matrix.

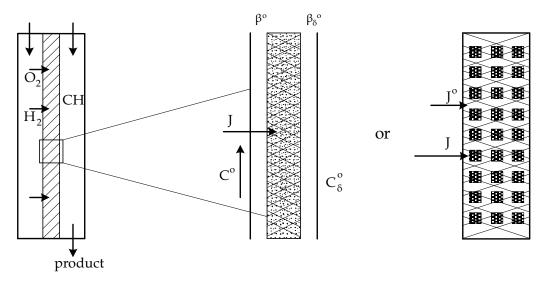


Fig. 8. Membrane reactor with dispersed catalyst particles (for heterogeneous model the spherical particles are modeled as cubic ones.

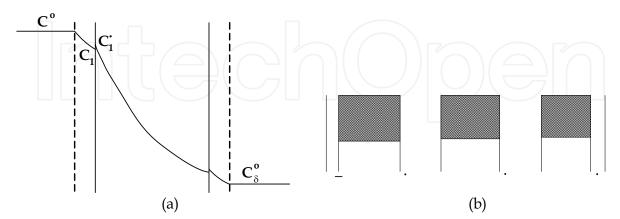


Fig. 9. Concentration distribution in the membrane reactor (a) and a particle line with notations (b)

For the description of this transport process, the catalyst membrane layer should be divided into 2N+1 sub-layers, perpendicular to the membrane interface. Namely, N sub-layers for

catalyst particles located perpendicular to the membrane interface, N+1 sub-layers for the polymer membrane matrix between particles (ΔY) and between the first particle (Y_1) and the last particle (1- Y_N^*) and the membrane interfaces (Fig. 9b). In order to get a mathematical expression for the mass transfer rates, differential mass balance equation should be given for each sub-layer. Thus, one can obtain a differential equation system containing 2N+1 second order differential equations. This equation system with suitable boundary conditions can be solved analytically which is also demonstrated in this paper. The number of particles, N, and the distance between them, ΔY , can be calculated from the particle size, d_p and the catalyst phase holdup, ϵ (Nagy, 2007). The distance of the first particle from the membrane interface, Y_1 (Y_1 = y_1/δ_m) can be regulated by the preparation method of the catalytic membrane layer. The differential mass balance equations for the sections of the polymer membrane phase and for that of the catalyst particles can be given, in dimensionless form, as follows, respectively:

$$\frac{D_m}{\delta_m^2} \frac{d^2 C}{dY^2} = 0 \qquad 0 \le Y \le Y_1 , \ Y_i + \frac{d}{\delta} \le Y \le Y_{i+1}, \ Y_i \le Y \le 1$$
 (68)

$$\frac{d^2C_p}{dY^2} - \frac{k_1\delta_m^2}{D_p}C_p = 0 \qquad Y_i \le Y \le Y_i + \frac{d}{\delta}$$

$$\tag{69}$$

The solutions of the above differential equations, for the ith sections, are well known, namely:

$$C = T_i Y + S_i \qquad 1 \le i \le N + 1 \tag{70}$$

$$C_p = E_i \exp(\Phi Y) + F_i \exp(-\Phi Y) \qquad 1 \le i \le N$$
(71)

with

$$\Phi = \sqrt{\frac{k_1 \delta_m^2}{D_n}}$$

Thus, one can obtain 2N+1 algebraic equations with twice as many parameters, T_i , S_i (i=1, 2, 3, ..., N+1) as well as E_i and F_i (i=1, 2, 3, ..., N), which are to be determined. Their values can be determined by means of suitable boundary conditions at the external interfaces of the membrane, at Y=0 and Y=1 as well as at the internal interfaces of every segment in the membrane matrix, at Y_i and Y_i^* with i=1, 2, 3, ..., N. The effect of the external mass transfer resistances should be taken into account:

At Y=0 then
$$\beta^{o}\left(C^{o}-C\right) \equiv \beta^{o}\left(C^{o}-\frac{S_{1}}{H_{m}}\right) = -\frac{D_{m}}{\delta_{m}}\frac{dC}{dY} \equiv -\beta_{m}^{o}T_{1} \tag{72}$$

At Y=1 then
$$\beta_{\delta}^{o}\left(C_{\delta} - C_{\delta}^{o}\right) \equiv \beta_{\delta}^{o}\left(\frac{T_{N+1} + S_{N+1}}{H_{m}} - C_{\delta}^{o}\right) = -\frac{D_{m}}{\delta_{m}}\frac{dC}{dY} \equiv -\beta_{m}^{o}T_{N+1}$$
(73)

The boundary conditions for the internal interfaces of the sub-layers are also well known (Fig. 9b):

at
$$Y = Y_i$$

$$(T_i Y_i + S_i) H = E_i e^{\Phi Y_i} + F_i e^{-\Phi Y_i}$$
 (74)

at
$$Y = Y_i$$

$$D_m A_i = D_p \Phi \left(E_i e^{\Phi Y_i} - F_i e^{-\Phi Y_i} \right)$$
 (75)

as well as for the other side of the catalyst particles, namely at $Y = Y_i^*$:

at
$$Y = Y_i + \frac{d}{\delta} = Y_i^*$$
 $\left(T_{i+1}Y_i^* + S_{i+1}\right)H = E_i e^{\Phi Y_i^*} + F_i e^{-\Phi Y_i^*}$ (76)

at
$$Y = Y_i + \frac{d}{\delta} = Y_i^*$$
 $D_m T_{i+1} = D_p \Phi \left(E_i e^{\Phi Y_i^*} - F_i e^{-\Phi Y_i^*} \right)$ (77)

Eqs 74 and 76 express that there is equilibrium on the sub-layer interfaces, while eqs 73 and 77 involve that there is no accumulation or source at the internal interfaces. Thus, an algebraic equation system with 2(N+1) equations can be obtained that can be solved analytically with a traditional method using the Cramer rules. The solution is briefly discussed in Nagy's paper (2007).

As a result of this solution, the mass transfer rate on the upstream side of the membrane interface, related to its heterogeneous part (which is the projection of the cubic catalyst particle onto the membrane interface), can be given as follows:

$$J = \beta_m H_m c^o \left(1 - T C_\delta^o \right) \tag{78}$$

where

$$\beta_{m} = \beta_{m}^{o} H \frac{U_{n(N+1)} + HH_{m} \frac{\beta_{m}^{o}}{\beta_{\delta}^{o}}}{U_{N+1} + HH_{m} \frac{\beta_{m}^{o}}{\beta_{\delta}^{o}}} \prod_{i=1}^{N} \left(\frac{U_{ni}}{U_{i}} \frac{\alpha_{ni}}{\alpha_{i}} \right)$$

$$(79)$$

as well as

$$T = \frac{1}{\left(U_{n(N+1)} + HH_m \frac{\beta_m^o}{\beta_\delta^o}\right) \prod_{i=1}^N \left(U_{ni}\alpha_{ni}\right)}$$
(80)

with

$$U_{n(N+1)} = \left[H \left(1 - Y_N^* \right) + \frac{\xi_{nN}}{\alpha_{nN}} \right] \tag{81}$$

$$U_{(N+1)} = \left[H \left(1 - Y_N^* \right) + \frac{\xi_N}{\alpha_N} \right] \tag{82}$$

as well as

$$HC = C_p$$
; $Y_N^* = Y_1 + (N-1)\Delta Y + N\frac{d}{\delta_m}$; $H_mC_j = C_j^*$ with $j = 1,2$

The values of U_{ni} , α_{ni} , and ξ_{ni} as well as U_i , α_i , and ξ_i should be calculated from sub-layer to sub-layer, that is from 1 to N (α_{ni} , ξ_{ni} , α_i , ξ_i) or N+1 (U_{ni} , U_i), from equations given in Table 1.

for i=2 to N
$$\alpha_{ni} = \frac{1}{U_{ni}} + \kappa \tanh(Ha_d); \qquad \xi_{ni} = \frac{\tanh(Ha_d)}{U_{ni}\kappa} + 1; U_{ni} = H\Delta Y + \frac{\xi_{ni}}{\alpha_{ni}}$$
for i=1 to N
$$\alpha_i = \frac{1}{U_i} + \kappa \tanh(Ha_d); \quad \xi_i = \frac{\tanh(Ha_d)}{U_i\kappa} + 1; \quad U_i = H\Delta Y + \frac{\xi_i}{\alpha_i}; \quad \Delta Y = Y_i - Y_{i-1}^*$$
as well as
$$U_{n1} = 1; \quad \alpha_{n1} = \kappa \tanh(Ha_d); \qquad \xi_{n1} = 1$$

$$U_1 = \left(HY_1 + HH_m \frac{\beta_m^o}{\beta^o}\right) \qquad Ha_d = \sqrt{\frac{k_1 d^2}{D_p}} \qquad \text{with } \kappa = \frac{\lambda D_d}{D}$$

Table 1. The list of variables that should be applied for the calculation of the mass transfer rates.

It may also be important to know the portion of the reactant that reacts in the catalytic membrane layer during its diffusion, or, there is an unreacted portion of the diffused reactant that passes on the downstream side of the membrane into the continuous phase. This outlet mass transfer rate, for the heterogeneous part of the membrane interface, at X=1 can be given as follows:

$$J_{\delta} = \beta_{out} H_{m} c^{o} \left(1 - \frac{C_{\delta}^{o}}{\prod_{i=1}^{N} \cosh(Ha_{d}) - \frac{\xi_{i}}{\alpha_{i}} \sinh(Ha_{d})} \right)$$
(83)

with

$$\beta_{\delta} = \beta_{m}^{o} H \frac{\prod_{i=1}^{N} \left(\cosh(Ha_{d}) - \frac{\xi_{i}}{\alpha_{i}} \sinh(Ha_{d}) \right)}{U_{N+1} + HH_{m} \frac{\beta_{m}^{o}}{\beta_{s}^{o}}}$$
(84)

The physical mass transfer rate for the heterogeneous part of the interface is as follows:

$$J^{o} = \beta_{tot}^{o} H_{m} c^{o} \left(1 - C_{\delta}^{o} \right) \tag{85}$$

The physical mass transfer coefficient, with external mass transfer resistances, for the portion of the membrane interface where there are particles in the diffusion path taking into account the effect of the catalyst particles, as well, can be given by the following equation:

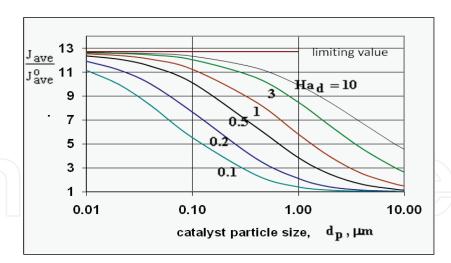


Fig. 10. Effect of the catalyst particle size on the mass transfer rate related to the total membrane interface, as a function of the reaction modulus (H_m=H=1; D_m=1 x 10⁻¹⁰ m²/s; $C^{o}_{\delta}=0$; $\beta^{o}=\beta^{o}_{\delta}\to\infty$; Y₁=1 µm; δ_{m} =30 µm; ϵ =0.1)

$$\beta_{tot}^{o} = \frac{1}{\frac{H_m}{\beta^o + \beta_{\delta}^o} + \frac{1}{\beta_m^o} + \frac{Nd}{D_p} \left(\frac{1}{H} - \frac{D_p}{D_m}\right)}$$
(86)

Depending on the value of the D_p diffusion coefficient, H solubility coefficient (C_p =CH) as well as the number of particles perpendicular to the interface, N, the value of the physical mass transfer coefficient of the membrane with catalytic particles, β_{tot}^o might be completely different from that of the membrane layer without catalyst particles, β_m^o ($\beta_m^o = D_m / \delta_m$). For example, in the case of a polymer membrane filled with zeolite particles as catalyst, the value of D_p can be lower by about 4 orders of magnitude than that in the polymer matrix (Jawalkar et al., 2001). The specific mass transfer rate related to the total catalytic membrane interface (Nagy, 2007) can be given as:

$$J_{ave} = KJ\varepsilon^{2/3} + J^{o}\left(1 - K\varepsilon^{2/3}\right) \tag{87}$$

or that for the outlet mass transfer rate:

$$J_{ave\delta} = KJ_{\delta}\varepsilon^{2/3} + J^{o}\left(1 - K\varepsilon^{2/3}\right) \tag{88}$$

The value of the mass transfer rate can be easily obtained for the homogeneous part of the interface, J^o , namely:

$$J^{o} = \beta_{m}^{o} H_{m} c^{o} \left(1 - C_{\delta}^{o} \right) \tag{89}$$

In order to calculate the enhancement during the mass transfer accompanied by chemical reaction, the physical mass transfer rate related to the total membrane interface should also be defined:

$$J_{ave}^{o} = K J_{tot}^{o} \varepsilon^{2/3} + J^{o} \left(1 - K \varepsilon^{2/3} \right)$$

$$\tag{90}$$

The value of the factor K can be obtained from the distribution of catalyst particles in the polymer membrane matrix. Its value, depending on the particles distribution in the membrane matrix, should be K=1 or K=1.8715 (for details see Nagy, 2007) The effect of the catalyst particle size is illustrated in Fig. 10 at different reaction rate applying the heterogeneous model. The mass transfer rate is very sensitive to the particle size, namely its value strongly decreases with the increase of the size. With increasing size decreases the number of particle because distance between them increases. Accordingly the particle size should be decreasing as low as possible. Detailed analysis of this model and its comparison to the homogeneous one is given in Nagy's paper (2007).

3. Mass transport through biocatalytic membrane layer

The principle of the mass transport of substrates/nutrients into the immobilized enzyme/cells, through a solid, porous layer (membrane, biofilm) or through a gel layer of enzyme/cells is the same. The structure, the thickness of this mass transport layer can be very different, thus, the mass transport parameters, namely diffusion coefficient, convective velocity, the bio-reaction rate constant, their dependency on the concentration and/or space coordinate is characteristic of the porous layer and of the nature of the biocatalysts. Several investigators modeled the mass transport through this biocatalyst layer, through enzyme membrane layer or cell culture membrane layer (Schonberg & Belfort, 1990, Kelsey et al., 1990, Piret & Cooney, 1991, Ferreira et al., 2001) Recently Nagy (2009a, 2009b) studied the mass transfer rate into a biocatalytic membrane layer with constant mass transport parameters. He defined the mass transfer rates for both side of the membrane surface. The rate equations are expressed as product of the mass transfer coefficient and driving force as it is traditionally applied for the diffusion systems, e.g. in gas-liquid systems. Applying these inlet mass transfer rate, the concentration profile of the two layers, namely that of the boundary layer and biocatalytic membrane layer can be calculated. These will be demonstrated in the case of first- and zero-order reactions as well as in the case of the general Monod kinetics. Assumptions, made for expression of the differential mass balance equation to the biocatalytic membrane layer, are:

- Reaction occurs at every position within the biocatalyst layer;
- Reaction has one rate-limiting substrate/nutrient;
- Mass transport through the biocatalyst layer occurs by diffusion and convection;
- The partitioning of the components (substrate, product) is negligible (Thus, $C^*=C^*_m$);
- The mass transport parameters (diffusion coefficient, convective velocity, bioreaction rate coefficient) are constant or varying;
- The effect of the concentration boundary layer should also be taken into account; For the sake of simplification, let us regard a steady-state reaction as well as let us use the Cartesian co-ordinate, thus, the differential mass balance equation, applying the Michaelis-Menten kinetics, can be given as follows (y is here the transverse space coordinate, perpendicular to the membrane interface):

$$\left\{ \frac{d}{dy} \left(D_m \frac{dC}{dy} \right) - \frac{d(\nu C)}{dy} \right\} - \frac{v_{\text{max}} C}{K_M + C} = 0$$
(91)

In general case, as it was mentioned, the diffusion coefficient and/or convective velocity can depend on the space coordinate, thus $D_m=D_m(y)$, υ (y), [or on the concentration, $D_m=D_m(C)$ or both of them, $D_m=D_m(C,y)$]. In the boundary conditions the external mass transfer resistance should also be taken into account. The membrane layer with biomass colony on it and the concentration boundary layer are illustrated, with important notations and concentration profiles, in Fig. 12.

The boundary conditions of eq. 91 are the same, in general case, as it given in eqs. 44 and 45. That is, the fluid phase mass transfer resistances of both sides of the membrane are taken into account. Eq. 91 can be solved numerically or by analytical approach, only. In the following sub-section we give an analytical approach, where the mass transfer rate is expressed in explicit, closed form, to its solution. The general solution of eq. 91 is discussed by Nagy (2008, 2009a). Here a solution without mass transfer resistance will only be shown.

3.1 Approaching analytical solution of the mass transport with variable parameters and/or with Monod kinetics for single membrane layer

Let us first to solve the differential mass balance eq. 91 with the boundary conditions 57a. and 57b. where the diffusive mass transfer resistances are negligible. The solution with boundary conditions given by eqs. 44 and 45 is much more complicated (not shown here). This solution was given by Nagy (2008) for diffusional mass transport through membrane reactor and by Nagy & Borbély, (2009) for diffusive+convective mass transport with variable parameters. In essentials, this solution methodology serves the mass transfer rate and the concentration distribution in closed, explicit mathematical expression. The method can be applied for Cartesian coordinate and cylindrical coordinate as will be shown. For the solution of the eq. (91), the biocatalytic membrane should be divided N sub-layer (Fig. 13), in the direction of the mass transport, that is perpendicular to the membrane interface, with thickness of $\Delta\delta$ ($\Delta\delta=\delta/N$) and with constant transport parameters in every sub-layer [for details of a general solution see (Nagy, 2009a)]. Thus, for the nth sub-layer of the membrane layer, using dimensionless quantities, it can be obtained (for better understanding m subscript for D is not written here):

$$D_n \frac{d^2 C_n}{dy^2} - v \frac{dC_n}{dy} - k_n C_n = 0 \quad y_{n-1} < y < y_n$$
 (92)

where the value of k_n can be obtained according e.g. to the Michaelis-Menten kinetics as follows:

$$k_n = \frac{v_{\text{max}}}{K_M + \overline{C}_n} \tag{93}$$

where \overline{C}_n denotes the average value of C in the nth membrane sub-layer. In dimensionless form one can get the following equation:

$$\frac{d^2C_n}{dY^2} - Pe_n \frac{dC_n}{dY} - \Phi_n^2 C_n = 0 , \qquad (94)$$

where

$$\Phi_n = \sqrt{\delta_m^2 k_n / D_n} ; \quad Pe_n = \frac{\upsilon \delta_m}{D_n}$$

The solution of eq. 92 can be easily obtained by well known mathematical methods, as it is geiven in sub-section 2.2.1, as it follows:

$$C = T_n e^{(\lambda_n Y)} + S_n e^{(\tilde{\lambda}_n Y)} \quad Y_{n-1} < Y < Y_n$$
(95)

with

$$\lambda_n = \frac{Pe_n}{2} + \Theta_n$$
 $\tilde{\lambda}_n = \frac{Pe_n}{2} - \Theta_n$ $\Theta = \sqrt{\frac{Pe_n^2}{4} + \Phi_n^2}$

 T_n and P_n parameters of Eq. (95) can be determined by means of the boundary conditions for the n^{th} sub-layer (with $1 \le n \le N$). The boundary conditions at the internal interfaces of the sub-layers ($1 \le n \le N-1$; $Y_m = n\Delta Y$; $\Delta Y = 1/N$) can be obtained from the following two equations [Eqs. (96a) and (96b)]:

$$-\frac{dC_n}{dY} + Pe_n C_n = \frac{D_{n+1}}{D_n} \left(-\frac{dC_{n+1}}{dY} + Pe_{n+1} C_{n+1} \right) \text{ at } Y = Y_n$$
 (96a)

$$C_n|_{Y=Y_n^+} = C_{n+1}|_{Y=Y_{n+1}^-} \text{ at } Y=Y_n$$
 (96b)

After solution of the algebraic equation system containing 2N equations, the mass transfer rate on the upstream side of the membrane can be given, for that case, as follows:

$$J = \frac{D_1 c^o}{\delta_m} \left(\tilde{\lambda}_1 T_1 + \lambda_1 P_1 \right) \tag{97}$$

$$T_{1} = \frac{c^{o} \varphi_{N}^{T}}{\varphi_{N}^{O}} \left(1 - \frac{C_{\delta}^{o}}{\varphi_{N}^{T} \left(\prod_{j=2}^{N} \varphi_{j-1}^{T} D_{mi} \tilde{\lambda}_{i} - \chi_{j-1}^{T} \right)} \right)$$
(98a)

and

$$S_{1} = -\frac{c^{o} \varphi_{N}^{S}}{\varphi_{N}^{O}} \left[1 - \frac{C_{\delta}^{o}}{\varphi_{N}^{S} \left(\prod_{j=2}^{N} \varphi_{j-1}^{S} D_{mi} \tilde{\lambda}_{i} - \chi_{j-1}^{S} \right)} \right]$$

$$(98b)$$

with

$$\chi_{i}^{j} = \frac{\left(\lambda_{i} e^{\tilde{\lambda}_{i} \Delta Y} - \tilde{\lambda}_{i} e^{\lambda_{i} \Delta Y}\right)}{\lambda_{i} - \tilde{\lambda}_{i}} - \frac{D_{mi} \tilde{\lambda}_{i} e^{\lambda_{i} \Delta Y}}{D_{mi} \tilde{\lambda}_{i} - \chi_{i-1}^{j} / \varphi_{i-1}^{j}} \text{ with i=2,3,...,N and j=T,S,O}$$
(99)

and

$$\varphi_i^j = \frac{e^{\tilde{\lambda}_i \Delta Y} - e^{\lambda_i \Delta Y}}{D_{mi} \left(\lambda_i - \tilde{\lambda}_i\right)} - \frac{e^{\lambda_i \Delta Y}}{D_{mi} \tilde{\lambda}_i - \chi_{i-1}^j / \varphi_{i-1}^j} \text{ with i=2,3,...,N and j=T,S,O}$$
(100)

The starting values of χ_i^T , χ_i^S , χ_i^O and φ_i^T , φ_i^S , φ_i^O are as follows:

$$\chi_1^T = D_{m1}\lambda_1 e^{\tilde{\lambda}_1 \Delta Y} \; ; \; \chi_1^S = D_{m1}\tilde{\lambda}_1 e^{\lambda_1 \Delta Y} \quad \chi_1^O = D_{m1} \left(\lambda_1 e^{\tilde{\lambda}_1 \Delta Y} - \tilde{\lambda}_1 e^{\lambda_1 \Delta Y}\right)$$

as well as

$$\varphi_1^T = e^{\tilde{\lambda}_1 \Delta Y}$$
; $\varphi_1^S = e^{\lambda_1 \Delta Y}$ $\varphi_1^O = e^{\tilde{\lambda}_1 \Delta Y} - e^{\lambda_1 \Delta Y}$

If one want to calculate the concentration distribution he should start with values of T_1 and S_1 and applying the known boundary conditions, the values of T_i and S_i (i=2,3,...,N) can easily be calculated. The internal boundary conditions could be as follows:

$$\left(\nu C - D_i \frac{\partial C}{\partial Y} \right)_{Y = Y_i^-} = \left(\nu C - D_{i+1} \frac{\partial C}{\partial Y} \right)_{Y = Y_i^+}$$
(101)

$$C\Big|_{Y=Y_{i}^{-}} = C\Big|_{Y=Y_{i}^{+}} \tag{102}$$

The outlet mass transfer rate can be similarly given (not shown here). This value should be as less as possible to avoid the loss of the substrate during the process. In the next section some typical figures will be shown to illustrate the effect of the parameters on the mass transport through the catalytic membrane layer.

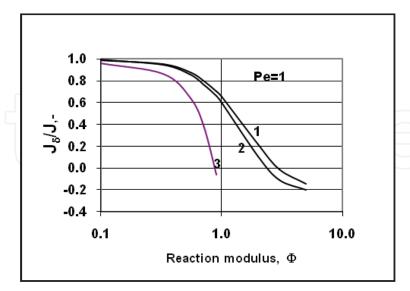


Fig. 11. The relative value of outlet mass transfer rate as a function of reaction modulus applying the Michaelis-Menten kinetics (line 2) and its limiting cases, namely first-order (line 1) and zero-order kinetics. (D_m=5.4 x 10⁻¹⁰ m²/s; $C_{\delta}^{o}=0$; $\beta^{o}=\beta_{\delta}^{o}\to\infty$; $K_{\rm M}/c^{o}=1$; $\delta_{\rm m}=100~\mu{\rm m}$)

In the case of microorganism the substrate concentration should be kept above its critical level. To hold it is important to apply convective mass flow, as well. A typical figure (Fig. 11) illustrates how the reaction rate influences the outlet mass transfer rate related to the inlet one calculated it in limiting cases and with the Michaelis-Menten kinetics (line 2). Fig. 11 illustrates clearly that the approach of the limiting case can cause essential error in prediction of the reactor efficiency.

4. Membrane reactor with biofilm on it

There is an increasing interest in the membrane-aerated biofilm reactor as a technology that can enhance the application of biofilms in wastewater treatment. In this reactor the biofilm is naturally immobilized on an substrate permeable membrane and counter- or (regarding the conventional biofilms where both the dissolved oxygen and substrates diffuse in the same direction) co-diffusion of oxygen and nutrients (organic component, ammonia, etc.) can take place. Several investigators have reported performance advantages of membraneaerated biofilm reactors for wastewater treatment (Aryal et al., 2009; Monthlagh et al., 2006) oxidation of organic components (Casey et al., 2000; Gross et al., 2007), nitrification (Rittman & Manem, 1992; Wang et al., 2009) etc. The structure of the biofilm can be homogeneous or heterogeneous depending on the substrate concentration (Piciorenau et al., 2001) because the growth rate of microorganisms depends strongly on the substrate concentration. Taking into account that the sum of the reacted amount of substrate in the biofilm, J_r and the inlet mass transfer rate into the membrane layer (layer m) is equal to that of inlet transfer rate of the biofilm layer, $(J=J_r+J_m)$, where J_m denotes the mass transfer rate that enters the membrane layer), the J value can be expressed for two reactive layers, similarly to the two-film theory as (Fig. 12):

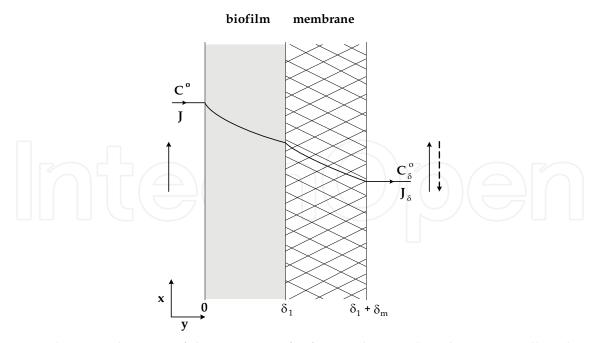


Fig. 12. Schematic diagram of the mass transfer for two bio-catalytic layers as well as the important notations

$$J = \beta_{tot} c^{o} \left(1 - L C_{\delta}^{o} \right) \tag{103}$$

where

$$\beta_{tot} = \frac{1/A + \beta_r / (\beta_M - \beta E)}{1/(\beta_m - \beta E) + 1/(\beta A)}$$
(104)

$$\beta = \frac{D_m}{\delta} \frac{(Pe_m / 2) \tanh \Theta + \Theta}{\tanh \Theta}$$
 (105)

$$\beta = \frac{D_m}{\delta_m} \frac{(Pe_m / 2) \tanh \Theta + \Theta}{\tanh \Theta}$$

$$A = \frac{\Theta e^{-Pe/2}}{\cosh \Theta \langle (Pe / 2) \tanh \Theta + \Theta \rangle}$$
(105)

$$L = \frac{\beta_m A}{\left(1 / A + \beta_r / \langle \beta_m - \beta_r E \rangle\right) (\beta_m - \beta_r E)}$$
(107)

where

$$\beta_r = \frac{D\Phi^2}{\delta \tanh\Theta} \frac{(Pe/2)\tanh\Theta + \Theta(e^{-Pe/2}/\cosh\Theta - 1)}{(Pe/2)^2 - \Theta^2}$$
(108)

and

$$E = \frac{Pe_m \sinh\Theta + \Theta\left(e^{-Pe_m/2} - \cosh\Theta\right)}{\Theta\left(e^{Pe_m/2} - Pe_m e^{-Pe_m/2} / 2\right)}$$
(109)

This equation does not involve the effect of the external mass transfer resistance.

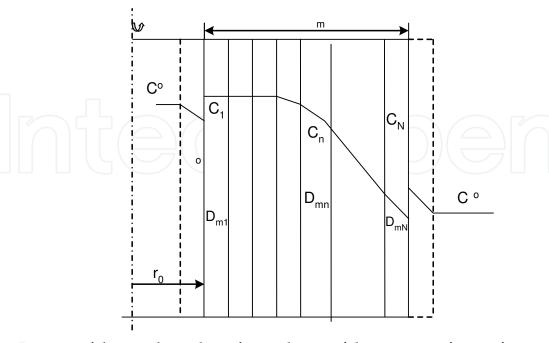


Fig. 13. Division of the membrane layer for prediction of the mass transfer rate for cylindrical and/or variable parameters.

5. Approaching solution in case of cylindrical coordinate

Applying the Michaelis-Menten equation, the mass balance equation with cylindrical coordinate is as:

$$D_m \left(\frac{dc^2}{dr^2} + \frac{1}{r} \frac{dc}{dr} \right) - \upsilon \frac{dc}{dr} - \upsilon_{\text{max}} \frac{c}{K_M + c} = 0$$
 (110)

The differential mass balance equation for state-state conditions, for cylindrical coordinate and for the nth sub-layer (Fig. 13) is as:

$$D_{mn} \left(\frac{dc^2}{dr^2} + \frac{1}{r_n} \frac{dc}{dr} \right) - \upsilon \frac{dc}{dr} - \upsilon_{\text{max}} \frac{c}{K_M + \overline{c}_n} = 0$$
 (111)

Rearranging eq. 111, the following dimensionless equation is obtained to be solved:

$$\frac{dC^{2}}{dR^{2}} - (Pe_{n} - 1)\frac{dC}{dR} - k_{n}C = 0$$
(112)

where

$$k_n = \frac{v_{\text{max}}}{K_M + \overline{C}_n} \qquad Pe_n = \frac{vR_o}{D_{mn}}$$

The solution of eq. 112 is the same as eq. 92:

$$C = T_n e^{(\lambda_n R)} + P_n e^{(\tilde{\lambda}_n R)} \quad R_{n-1} < R < R_n$$
(113)

with

$$\lambda_{n} = \frac{\left(Pe_{n} - 1\right) + \sqrt{\left(1 - Pe_{n}\right)^{2} + 4\Phi_{n}^{2}}}{2} \qquad \tilde{\lambda}_{n} = \frac{\left(Pe_{n} - 1\right) - \sqrt{\left(1 - Pe_{n}\right)^{2} + 4\Phi_{n}^{2}}}{2}$$

as well as

$$\Phi = \sqrt{\frac{k_n R_o^2}{D_n}}$$

where R_o is the radius of the cylindrical membrane, $R=1+n(R_o+\delta_m)/R_o$, $R=r/R_o$. The determination of T_n , S_n parameters is the same as in the case of Cartesian coordinate.

6. Full-scale description of the process in hollow fiber membrane

Mostly used membrane configuration is capillary module, though the industry often applies tubular, spiral-wound and plate-and frame modules (Baker, 2004). Here the mass balance or continuity equations are given in cylindrical coordinate. These can easily be rewritten to Cartesian one, as well (Cebeci, 2005). For cross-flow processes mass balance equation should be given for both the lumen and the shell sides of the membrane. The feed stream in the

tube or outside of the membrane, is modeled by the Navier-Stokes equations. The simplified mass and momentum equations, i.e., Navier-Stokes equations expressed in cylindrical coordinates with axisymmetry assumption, are as (Damak et al., 2004):

$$\frac{1}{r}\frac{\partial}{\partial r}(r\upsilon) + \frac{\partial u}{\partial x} = 0 \tag{114}$$

$$\rho\left(\upsilon\frac{\partial u}{\partial r} + u\frac{\partial u}{\partial x}\right) = -\frac{\partial P}{\partial x} + \mu\left\{\frac{\partial}{\partial r}\left[\frac{1}{r}\frac{\partial(ru)}{\partial r}\right] + \frac{\partial^{2}u}{\partial x^{2}}\right\}$$
(115)

$$\rho \left(\upsilon \frac{\partial \upsilon}{\partial r} + \upsilon \frac{\partial \upsilon}{\partial x} \right) = -\frac{\partial P}{\partial x} + \mu \left\{ \frac{\partial}{\partial r} \left[\frac{1}{r} \frac{\partial (r\upsilon)}{\partial r} \right] + \frac{\partial^2 \upsilon}{\partial x^2} \right\}$$
(116)

as well as for solute concentration in case of constant density

$$u\frac{\partial C}{\partial z} + \upsilon \frac{\partial C}{\partial r} = \frac{\partial}{\partial r} \left(D\frac{\partial C}{\partial r} \right) + \frac{1}{r} \frac{\partial (DC)}{\partial r} + \frac{\partial}{\partial x} \left(D\frac{\partial C}{\partial x} \right)$$
(117)

Tranfer equations in the porous wall; The momentum equation, i.e., Darcy's law, is as follows:

$$\upsilon = -\frac{\kappa}{\mu} \frac{\partial P}{\partial r} \tag{118}$$

$$u = -\frac{\kappa}{\mu} \frac{\partial P}{\partial x} \tag{119}$$

where κ is the dimensionless Darcy's law permeability coefficient.

Boundary conditions; At the inlet, a fully developed laminar profile can be considered, i.e., Poiseuille flow which leads to:

$$u = 2u_o \left[1 - \left(\frac{r}{R} \right)^2 \right]$$
 and $v = 0$ at $x = 0$; $0 \le r \le R$ (120)

 u_o represents the axial velocity at the inlet. At the exit, a fully developed profile is assumed as follows:

$$\frac{\partial u}{\partial x} = 0 \text{ and } v = 0 \text{ at } x = L; 0 \le r \le R$$
 (121)

At the axis of symmetry there are no momentum fluxes crossing the boundary:

$$\frac{\partial u}{\partial x} = 0$$
 and $v=0$ at $r=0$; $0 \le x \le L$ (122)

At the permeable wall:

$$v = \frac{\kappa}{\mu} \frac{\Delta P}{\delta}$$
 and u=0 at r=R; $0 \le z \le L$ (123)

The most hollow fiber configurations, the ratio R/L are very small, less than 1 x 10^{-3} , thus, the inertial terms can be neglected (Mondor & Moresoli, 1999). Finally, because the velocity gradients are smaller in the axial direction than in radial direction, the axial stress terms can be neglected in the momentum equation. Thus, the simplified form the momentum and the continuity equations are, respectively, given as:

and
$$\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial u}{\partial r}\right) = \frac{dP}{dx}$$

$$\frac{\partial u}{\partial x} + \frac{1}{r}\frac{\partial(rv)}{\partial r} = 0$$
(124)

The solute balance equation with constant diffusion coefficient:

$$u\frac{\partial C}{\partial x} + \upsilon \frac{\partial C}{\partial r} = D \left(\frac{\partial^2 C}{\partial r^2} + \frac{1}{r} \frac{\partial C}{\partial r} + \frac{\partial^2 C}{\partial x^2} \right)$$
(126)

The J mass transfer rate presented should be inserted into the boundary condition given by eq. 2c of mass balance eq. (1a) or eq. (126), thus this differential equation can be solved. In the case of membrane reactor or bioreactor, the axial pressure gradient within the membrane is often negligible compared to the radial pressure gradient, thus the first term in eq. 125 can often be neglected. When there is no change of volume of the fluid phase because the low convective permeation rate or the case of dilute fluid phase, the mass balance equation given by eq. 126 should be taken into account during the mass transport calculation (Piret & Cooney, 1991).

7. Conclusion

Mass transfer rate and, in some cases, the concentration distribution inside a membrane reactor were defined. Exact solutions of the mass transfer rate were given, taking into account the external mass transfer resistance on the both sides of the catalytic membrane layer. The membrane is either intrinsically catalytic or catalytic particles are dispersed in the membrane matrix. For this letter case, both pseudo-homogeneous model (for nanometer sized particles) and heterogeneous one (for microsized catalyst particles) have been presented. An analytical approaching solution was developed for cylindrical coordinate and/or variable mass transport parameters, as e.g. diffusion coefficient, chemical reaction rate constant. The mass transfer rates obtained then should be inserted as a boundary condition into differential mass balance equations in order to describe the full-scale mass balance equation given for capillary or plate-and-frame modules.

8. Appendix

The differential mass balance equations for the reactants in the membrane layer assuming that $Q=k_2c_Ac_{B_i}$ for component A and B, respectively:

$$D_A \frac{d^2 C_A}{dy^2} - k_2 c_A c_B = 0 (A1)$$

$$D_B \frac{d^2 C_B}{dy^2} - k_2 c_A c_B = 0 (A2)$$

Let us apply the following boundary conditions:

y=0 then
$$c_A = c_A^o \quad c_B = c_{B\delta}$$
 (A3)

and

$$y=\delta_{\rm m}$$
 then $c_A=c_{A\delta}$ $c_B=c_B^o$ (A4)

Dividing the membrane layer into N very thin, sub-layers, the following approach can be applied regarding the concentrations: the mass balance equation is given one of the reactants while its average value, e.g. $(c_{Ai-1}+c_{Ai})/2$ is considered for the other component in this equation. Thus, one can write for e.g. components A the following differential equation, in dimensionless form, for the ith sub-layer:

$$\frac{d^2C_A}{dY^2} - \Phi_{Ai}C_A = 0 \qquad Y_{i-1} \le Y \le Y_i \tag{A5}$$

with

$$\Phi_{Ai} = \sqrt{\frac{k_2 \delta_m^2 c_A^o c_B^o \overline{C}_{Bi}}{D_{mA}}}$$
 (A6)

The mass balance equation can similarly be given for component with the following Φ_{Bi} value:

$$\Phi_{Bi} = \sqrt{\frac{k_2 \delta_m^2 c_A^o c_B^o \overline{C}_{Ai}}{D_{mB}}}$$
 (A7)

The general solution of eq. A5 for the ith sub-section is as follows:

$$C_A = T_i e^{\Phi_{Ai} Y} + S_i e^{-\Phi_{Ai} Y} \qquad Y_{i-1} \le Y \le Y_i$$
 (A8)

This equation should be given for every sub-layer, thus, one can get N mass balance equation for component A with two parameters, namely T_i and S_i in them. The values of T_i and S_i with i=1,2,...,N can be determined by the following boundary conditions:

at
$$Y=0$$
 C=1 (A9)

at
$$Y_{i-1} \le Y \le Y_i$$
 $D_{mi-1} \frac{dC_A}{dY} = D_{mi} \frac{dC_A}{dY}$ with i=1,2,...,N (A10)

at
$$Y_{i-1} \le Y \le Y_i$$
 $C_{Ai-1} = C_{Ai}$ with i=1,2,...,N (A11)

at Y=1
$$C_A = C_{A\delta}$$
 (A12)

It is worth to mention that the method presented makes possible to calculate the mass transport when the diffusion coefficient of the reactant is variable. They can depend on the

space coordinate and/or on the concentration. In this case a constant diffusion coefficient had to be given for every sub-layer. This is taken into account in eq. A10, where D_{mi} should not be equal to D_{mi-1} . Then the variable diffusion coefficient should be involved in the values of Φ_{Ai} and Φ_{Bi} .

According to eqs. A9 to A12, one can obtain 2N algebraic equations. This equation system can analytically be solved. Thus, the parameters can be given by means of the mass transport parameters, namely diffusion coefficient, reaction rate constant, etc. details on this method can be found in Nagy's papers (Nagy, 2008, 2010).

After solution of the N differential equation with 2N parameters to be determined the T_1 and S_1 parameters for the first sub-layer can be obtained as (ΔY is the thickness of sub-layers):

$$T_{1} = -\frac{1}{2\xi_{N}^{O}\cosh(\Phi_{A1}\Delta Y)} \left(\xi_{N}^{T} - \frac{C_{A\delta}^{o}}{\prod_{i=2}^{N}\cosh(\Phi_{Ai}\Delta Y)} \right)$$
(A13)

and

$$S_{1} = \frac{1}{2\xi_{N}^{O}\cosh(\Phi_{A1}\Delta Y)} \left(\xi_{N}^{S} - \frac{C_{A\delta}^{o}}{\prod_{i=2}^{N}\cosh(\Phi_{Ai}\Delta Y)}\right)$$
(A14)

Knowing the T_1 and S_1 the other parameters, namely T_i and S_i (i=2,3,...,N) can be easily be calculated by means of the internal boundary conditions given by eqs. A10 and A11, from starting from T_2 and S_2 up to T_N and S_N . Thus, one can get the following equations for prediction of the T_i and S_i from T_{i-1} and S_{i-1} :

$$T_i e^{\Phi_i Y_i} + S_i e^{-\Phi_i Y_i} = \Gamma_{i-1} \tag{A15}$$

$$D_{mi}\Phi_i \left(T_i e^{\Phi_i Y_i} - S_i e^{-\Phi_i Y_i} \right) = \Xi_{i-1} \tag{A16}$$

with

$$\Gamma_{i-1} = T_{i-1}e^{\Phi_{i-1}Y_i} + S_{i-1}e^{-\Phi_{i-1}Y_i}$$
(A17)

$$D_{mi-1}\Phi_{i-1}\left(T_{i-1}e^{\Phi_{i-1}Y_i} - S_{i-1}e^{-\Phi_{i-1}Y_i}\right) = \Xi_{i-1}$$
(A18)

Now knowing the T_i and S_i (with i=1,2,...,N) parameters, the concentration distribution can be calculated easily through the membrane, i.e. its value for every sub-layer.

Notations

c = concentration in the membrane, $[=w\rho/(Mc^o)]$, mol/m³

C = dimensionless concentration in the membrane, $(=c/c^{\circ})$,-

```
CO
          = bulk phase concentration, mol/m<sup>3</sup>
          = concentration at the membrane interface, mol/m<sup>3</sup>
C
d_{p}
          = particle size, m
          = d_v \sqrt[3]{6/\pi/\delta}
d
D
          = diffusion coefficient, m<sup>2</sup>/s
          = distance between cubic particles (Nagy, 2007), m
h
          = solubility coefficient of reactant between polymer matrix and catalyst particle, -
Н
          = solubility constant of reactant between the continuous phase and the polymer
H_{m}
          membrane matrix,-
          = Hatta-number of the cubic particles in the heterogeneous model, \left(=\sqrt{k_1R_p^2}/D_p\right)
Had
          = Hatta-number of catalyst particles (Ha_d = 2.324 Ha_p), \left(= \sqrt{k_1 R^2} / D_p\right)
Hap
          = mass transfer rate to catalyst particle, mol/(m<sup>2</sup>s)
Jo
          = physical mass transfer rate, mol/(m^2s)
          = mass transfer rate in presence of chemical reaction, mol/(m<sup>2</sup>s)
J
J_m^o
          = physical mass transfer rate related to the homogeneous membrane interface,
          mol/(m^2s)
          = outlet mass transfer rate, mol/(m<sup>2</sup>s)
Jδ
k
          = reaction rate constant, 1/s
          = length of capillary, m
L
          = molecular weight of reactant, g/mol
M
          = number of particle perpendicular to the membrane interface
N
P
          = pressure, Pa
          = radius of the spherical catalyst particles, m
r
R
          = dimensiomles radius, (r/R_o)
R_{o}
          = capillary radius, m
          = time, s
t
          = convective velocity in axial direction, m/s
u
uo
          = inlet velocity, m/s
          = axial space coordinate, m
X
Χ
          = dimensionless space coordinate (=x/L)
          = space coordinate through the membrane, m
y
Y
          = dimensionless space coordinate (=y/\delta_m)
          = distance of first particles from the interface (Y_1=y_1/\delta_m, Y_1=\overline{y}_1/\delta_m), m
y_1, \overline{Y}_1
ΔΥ
          = distance between particles in the membrane (\Delta Y = \Delta y / \delta_m), m
          = distance of the ith particle from the interface, -
X_{i}
Y_i
          = Y_i + d
          = concentration of reactant in the membrane, kg/kg
Greek letters
βo
          = physical mass transfer coefficient of fluid phase, m/s
          = mass transfer coefficient of the polymer membrane layer (=D_m/\delta_m), m/s
\beta_m^o
          = mass transfer coefficient with chemical reaction, m/s
```

= physical mass transfer coefficient with overall resistance, m/s

= external mass transfer coefficient around particles (=2D/d_p+ D/ δ _p), m/s

 β_{δ} = mass transfer coefficient in the outlet rates, m/s

 $\delta_{\rm m}$ = thickness of the membrane layer, m

 δ_p = diffusion boundary layer around particles, (=[h-d_p]/2), m

 ρ = average density of the membrane, kg/m³

 ε = catalyst phase holdup

 ω = specific interface of catalyst particles, m²/m³

 ω = specific interface of catalyst particles in the membrane, $(=6\varepsilon/d_p)$, m²/m³

Subscripts

A = reactant A

ave = average

B = reactant B

i = integer parameter

m = polymer membrane

L = fluid phase

p = catalyst particle

 δ = permeate side of membrane

1 first-order

0 zero-order

9. References

Aryal R., Lebegue, J., Vigneswaran S., Kandasamy, J., Grasmick, A. (2009) Identification and characterization of biofilm formed on membrane bio-reactor, *Sep. Purif. Technol.*, 67 86-94

Baker, R.W. (2004) Membrane technology and application, Wiley & Sons, England

Belfort, G. (1989), Membranes and Bioreactors: A Technical Challenge in Biotechnology, *Biotechnology and Bioengineering*, 33, 1047-1066.

Breure, B.; Peters, E.A.J.F. & Kerkhof, P.J.A.M. (2008). Separation of azeotropic mixtures of alcohols and water with FricDiff, *Sep. Purif. Technology*, 62, 350-363.

Brotherton, J.D. & Chau, P.C. (1990). Modeling analysis of an intercalated-spiral alternate-dead-ended hollow fiber bioreactor for mammalian cell cultures, *Biotechnology and Bioengineering*, 35, 375-394.

Brotherton, J.D. & Chau, P.C. (1996) Biotechnology Progress, 112, 575-590

Cabral, J.M.S. & Tramper, J. (1994) Bioreactor Design, In *Applied Biocatalysis*. (Ed. By J.M.S. Cabral, D. Best, L. Boross, J. Tramper, Harwood Academic Publishers, Switzerland, pp. 330-370.

Calabro, V., Curcio, S., & Iorio, G. (2002), A theoretical analysis of transport phenomena in a hollow fiber membrane bioreactor with immobilized biocatalyst, *J. Membrane Sci.*, 206, 217-241.

Carvalho CML, Aires-Barros M.R. & Cabral JMS. (2000) A continuous membrane bioreactor for ester synthesis in organic media: II Modeling of MBR continuous operation, *Biotechnology and Bioengineering*, 72, 136-143

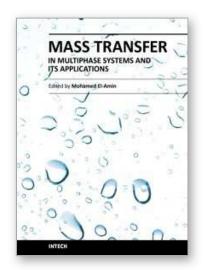
Casey, E., Glennon, B., Hamer, G. (2000) Biofilm development in a membrane-areated biofilm reactor: Effect of flow velocity on performance, *Biotechnol. Bioeng.*, 67 (4) 476-486.

Cebeci, T., Shao, J.P., Kafyeke, F. & Laurendeau, L. (2005) *Computational fluid dynamics for engineers*, Horizons Publishing Inc. and Springer, Heidelberg.

- Charcosset, C. (2006), Membrane processes in biotechnology: An overview, *Biotechnology Advances*, 24, 482-492.
- Champagnie, A.M., Tsotsis. T.T., Minet, R.G. & Webster, I.A. (1990) *Chem. Engng. Sci.*, 45, 2423-2429.
- Damak, K.; Ayadi, A.; Zeghmati, B. & Schmitz, P. (2004). A new Navier-Stokes and Darcy's law combined model for fluid flow in crossflow filtration tubular membranes, *Desalination*, 161, 67-77.
- Ferreira, B.S., Fernandes, P. & Cabral J.M.S. (2001) Design and modeling of immobilized biocatalytic reactors, in *Multiphase bioreactor design* (Ed. by J.M.S. Cabral, M. Mota, J. Tramper), Taylor and Francis, London, UK., pp. 85-180.
- Frazeres, D.M.F. & Cabral, J.M.S. (2001) Enzymatic membrane reactors, In: J.M.S. Cabral, M. Mota, J. Tramper (Eds.), *Multiphase bioreactor design*, Taylor Francis., pp. 135-184.
- Giorno L. & Drioli E. (2000) Biocatalytic membrane reactors: applications and perspectives, *Trends in biotechnology* 18, 339-349.
- Godongwana, B.; Sheldon, M.S. & Solomons, D.M. (2007). Momentum transfer inside a vertically orientated capillary membrane bioreactor, *J. Membr. Sci.*, 303, 86-99.
- Gross, R., Hauer, B., Otto, K., Schmid, A. (2007) Microbial biofilms: New catalysis for maximizing productivity of long-term biotransformation, *Biotechnol. Bioeng.*, 98 (6) 1123-1134..
- Habulin, M. & Knez, Z. (1991), Enzymatic synthesis of n-butyl oleate in a hollow fiber membrane reactor, *J. Membrane Sci.*, 61, 315-324.
- Hossain MM. & Do DD. (1989) General Theory of Determining Intraparticle Active Immobilized Enzyme Distribution and Rate Parameters, *Biotechnology and Bioengineering*, 33, 963-975.
- Ilinitch O.M., Cuperus, F.P., Nosova, L.V. & Gribov, E.N., (2000) Catalytic membrane in reduction of aqueous nitrates: operational principles and catalytic performance, *Catalysis Today*, 56, 137-145.
- Yawalkar, A.A., Pangarkar, V.G. & Baron, G.V. (2001) Alkene epoxidation with peroxide in a catalytic membrane reactor: a theoretical study, *J. Membrane Science*, 182, 129-137.
- Julbe, A.; Farusseng, D. & Guizard, C. (2001) Porous ceramic membranes for catalytic reactors-overview and new ideas, *J. Membrane Sci.*, 181, 3-20.
- Kelsey, L.J., Pillarella, M.R. & Zydney, A.L. (1990), Theoretical analysis of convective flow profiles in a hollow-fiber membrane bioreactor, *Chem. Eng. Sci.*, 45 (11), 3211-3220.
- Lu SG, Imai T, Ukita M, Sekine M, Higuchi T. & Fukagawa M. (2001) A model for membrane bioreactor process based on the concept of formation and degradation of soluble microbial products, *Wat. Res.*, 35, 2038-2048.
- Long, W.S.; Bhatia S. & Kamaruddin, A. (2003). Modeling and simulation of enzymatic membrane reactor for kinetic resolution of ibuprofen ester, *J. Membr. Sci.*, 219, 69-88.
- Marcano, J.G.S. & Tsotsis, T.T. (2002). Catalytic membranes and membrane reactiones, Wiley-VCH, Veinheim.
- Maira, A.J., Lau, W.N.; Lee, C.Y.; Chan, C.K. & Yeung, K.L. (2003) Performance of a membrane-catalyst for phocatalyc oxidation of volatile organic compounds, *Chem. Eng. Sci.*, 58, 959-962.
- Marror,B., Barnos-Martinez,A., Moulin,P. & Roche,N. (2004), Industrial Wastewater Treatment in a Membrane Bioreactor: A Review, *Environmental Prograss*, 23, 59-68.

- Mehra, A. (1999) Heterogeneous modeling of gas absorption in emulsion, *Ind. Eng. Chem. Res.*, 38, 2460-2468.
- Melo, L.F. & Oliveira, R. (2001). Biofilm reactors, In: J.M.S. Cabral, M. Mota, J. Tramper (Eds.), *Multiphase bioreactor design*, Taylor Francis., pp. 271-308.
- Mondor, M. & Moresoli C. (1999). Theoretical analysis of the influence of the axial variation of the transmembrane pressure in cross-flow filtration of rigid spheres, *J. Membr. Sci.*, 152, 71-87.
- Mothlagh, A.R., Voller, V.R., Semmens, M.J. (2006) Advective flow through membrane-areated biofilms, Modeling results, *J. Membr. Sci.*, 273 143-151.
- Mulder, M.H.V. Polarization phenomena and membrane fouling. In Noble and Stern (Ed.) *Membrane separation technology, principles and applications,* Elsevier, Oxford, 1995
- Nagy E. (1995) Three-phase mass transfer: One-dimensional heterogeneous model, *Chem. Engng. Sci.*, 50, 827-836.
- Nagy, E. (2002) Three-phase oxygen absorption and its effect on fermentation, *Adv. Biochem. Eng./Biotechnol.*, 75, 51-81.
- Nagy E. (2009a) Mathematical Modeling of Biochemical Membrane Reactors, in *Membrane Operations, Innovative Separations and Transformations*, Ed. by E. Drioli and L. Giorno, WILEY-VCH Velag, Weinheim, pp. 309-334
- Nagy, E. (2009b) Basic equations of mass transfer through biocatalytic membrane layer, *Asia-Pacific J. of Chem. Eng.*, 4, 270-278.
- Nagy E., Kulcsár E. (2009) Mass transport through biocatalytic membrane reactors, *Desalination* 245, 422-436.
- Nagy E., Borbély G. (2009) Mass transport through anisotropic membrane layer, *Desalination*, 240, 54-63.
- Nagy E. (2007) Mass transfer through a dense, polymeric, catalytic membrane layer with dispersed catalyst, *Ind. Eng. Chem. Res.*, 46, 2295-2306.
- Nagy, E. (2006) Binary, coupled mass transfer with variable diffusivity through cylindrical membrane, *J. Membrane Science*, 274, 159-168
- Nagy E. & Moser, A. (1995) Three-phase mass transfer: Improved pseudo-homogeneous model, *AIChE J.*, 41, 23-34.
- Nagy, E. (2008) Mass transport with varying diffusion- and solubility coefficient through a catalytic membrane layer, *Chem. Eng. Res. Design*, 86, 723-730
- Nagy, E., Blickle, T. & Ujhidy, A. (1989) Spherical effect on mass transfer between fine solid particles and liquid accompanied by chemical reaction, *Chem. Engng. Sci.*, 44, 198-201.
- Nagy E. (2010) Mass transport through a convection flow catalytic membrane layer with dispersed nanometer-sized catalyst, *Ind. Eng. Chem Res.*, 49, 1057-1062.
- Nakajima, M. & Cardoso, J.P. (1989) Forced-flow bioreactor for sucrose inversion using ceramic membrane activated by silanization, *Biotechnol. Bioeng.*, 33, 856-861.
- Picioreanu, C., van Loosdrecht, M.C.M., Heijnen, J.J. (2001) Two-dimensional model of biofilm detachment caused by internal stress from liquid flow, *Biotechnol. Bioeng.*, 72 (2) 205-218.
- Piret, J.M. & Cooney, C.L. (1991). Model of oxygen transport limitations in hollow fiber bioreactors, *Biotechnology and Bioengineering*, 37, 80-92.
- Rios, G.M., Belleville, M-P. & Paolucci-Jeanjean, D. (2007), Membrane engineering in biotechnology: quo vamus? *Trends in Biotechnology*, 25, 242-246.

- Rittman, B.E., Manem J.A. (1992) Development and experimental evaluation of a steady-state, multispecies biofil model, *Biotechnol. Bioeng.*, 39 914-922.
- Rosenberger, S., Krüger, U., Witzig, R., Manz, W., Szewzyk, U. & Kraume, M. (2002), Performance of a bioreactor with submerged membranes for aerobic treatment of municipal waste water, *Water Research*, 36, 413-420.
- Salzman, G., Tadmor, R., Guzy, S., Sideman, S., & Lotan, N. (1999), Hollow fiber enzymic reactors for a two substrate process: analytical modeling and numerical simulations, Chem. Eng. Proc., 38, 289-299.
- Saracco, G.& Specchia, V.; (1995) Catalytic ceramic filters for flue gas cleaning. 2: catalytic performance and modeling thereof. *Ind. Eng. Chem. Res.*, 34, 1480-1487.
- Sardonini, C.A. & Dibiasio, D. (1992), An Investigation of the Diffusion-Limited Growth of Animal Cells Around Single Hollow Fibers, *Biotechnology and Bioengineering*, 40, 1233-1242.
- Schonberg JA. & Belfort G. (1987) Enhanced nutrient transport in hollow fiber perfusion bioreactors: a theoretical analysis, *Biotechnology Progress*, 3, 81-89.
- Schmidt, A.; Haidar, R. & Schomacker, R. (2005) Selectivity of partial hydrogenation reactions performed in a pore-through-flow catalyst membrane reactor, *Catal. Today*, 104, 305-312.
- Seidel-Morgenstern, A. (2010), Membrane reactors, Wiley-WCH, Weinheim
- Sheldon, M.S. & Small, H.J. (2005), Immobilisation and biofilm development of *Phanerochaete chrysosporium* on polysulphone and ceramic membranes, *J. Membrane Sci.*, 263, 30-37.
- Strathmann H, Giorno L, & Drioli E. (2006) *An introduction to membrane science and technology,* Institute on Membrane Technology, Italy
- Yamada, M.; Fugii, K.; Haru, H. & Itabashi, K.; (1988) Preparation and catalytic properties of special alumina membrane formed by anodic oxidation of aluminum. *Tech. Rep. The Light Metal Educational Foundation*, Inc. Research Group for Functionalizing of Aluminum and its Surface Films
- Yang, W., Cicek, N.& Ilg, J. (2006) State-of-the-art of membrane bioreactors: Worldwide research and commercial applications in North America, J. Membrane Sci., 270, 201-211
- Vancelecom, I.F.J. & Jacobs, P.A. (2000). Dense organic catalytic membrane for fine chemical synthesis, *Catalysis Today*, 56, 147-157.
- Vincent, M.J.& Gonzales, R.D. (2002) Selective hydrogenation of acetylene through a short contact time reactor, *AIChE J.*, 48, 1257-1263.
- Vital, J., Ramos, A.M., Silva, I.F., Valenete, H. & Castanheiro, J.E. (2001) Hydration of α-pinene over zeolites and activated carbons dispersed in polymeric membranes, *Cataysis Today*, 67, 217-223.
- Wang, R., Terada, A., Lackner, S., Smets, B.F., Henze, M., Xia, S., Zhao, J. (2009) Nitritation performance and biofilm development of co- and counter-diffusion biofilm reactors: Modeling and experimental comparison, *Water Research*, 43, 2699-2709.
- Waterland, L.R., Robertson, C.R. & Michaelis, A.S. (1975) Enzymatic catalysis using asymmetric hollow fiber membranes, *Chem. Eng. Commun.*, 2: 37-47.
- Waterland, L.R., Robertson, C.R. & Michaelis, A.S. (1974). A theoretical model for enzymatic catalysis using asymmetric hollow fiber membranes, *AIChE Journal*, 20, 50-59
- Westermann T., Melin T. (2009) Flow-through catalytic membrane reactors- Principles and applications, *Chem. Eng. and Processing*, 48, 17-28.



Mass Transfer in Multiphase Systems and its Applications

Edited by Prof. Mohamed El-Amin

ISBN 978-953-307-215-9 Hard cover, 780 pages **Publisher** InTech

Published online 11, February, 2011

Published in print edition February, 2011

This book covers a number of developing topics in mass transfer processes in multiphase systems for a variety of applications. The book effectively blends theoretical, numerical, modeling and experimental aspects of mass transfer in multiphase systems that are usually encountered in many research areas such as chemical, reactor, environmental and petroleum engineering. From biological and chemical reactors to paper and wood industry and all the way to thin film, the 31 chapters of this book serve as an important reference for any researcher or engineer working in the field of mass transfer and related topics.

How to reference

In order to correctly reference this scholarly work, feel free to copy and paste the following:

Nagy Endre (2011). Mass Transfer through Catalytic Membrane Reactor, Mass Transfer in Multiphase Systems and its Applications, Prof. Mohamed El-Amin (Ed.), ISBN: 978-953-307-215-9, InTech, Available from: http://www.intechopen.com/books/mass-transfer-in-multiphase-systems-and-its-applications/mass-transfer-through-catalytic-membrane-reactor



InTech Europe

University Campus STeP Ri Slavka Krautzeka 83/A 51000 Rijeka, Croatia Phone: +385 (51) 770 447

Fax: +385 (51) 686 166 www.intechopen.com

InTech China

Unit 405, Office Block, Hotel Equatorial Shanghai No.65, Yan An Road (West), Shanghai, 200040, China 中国上海市延安西路65号上海国际贵都大饭店办公楼405单元

Phone: +86-21-62489820 Fax: +86-21-62489821 © 2011 The Author(s). Licensee IntechOpen. This chapter is distributed under the terms of the <u>Creative Commons Attribution-NonCommercial-ShareAlike-3.0 License</u>, which permits use, distribution and reproduction for non-commercial purposes, provided the original is properly cited and derivative works building on this content are distributed under the same license.



