The Investigation of Time-dependent Solute Transport through Horizontally Situated Membrane: The Effect of Configuration Membrane System

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Abstract. This paper presents an experimental and theoretical investigation of solute transport through a horizontally situated membrane. The experimental investigation was carried out by the laser interferometric method in association with a computer system of interference image analysis. On the basis of this analysis thickness of near-membrane layers, solution concentration drops on these layers as well as diffusion fluxes of diluted substance are determined. Different fluxes of the soluble substance are observed depending on the configuration of the system. The results of the experimental and theoretical investigation of diffusion fluxes are conformable in respect of measurement error, with one adjustment parameter, i.e. the solute partition coefficient.

Key words: Diffusion, Concentration polarization, Gravitational convection, Diffusion flux, Membrane, Concentration boundary layers

1. Introduction

Transport of soluble substance and water through biological membranes [1–3] as well as through polymer membranes [4, 5] leads to the formation of concentration boundary layers in their neighbourhood. Occurrence of these layers in membrane systems constitutes an important factor which modifies (diffusion and osmotic) transport of substance through membrane [6, 7].

Of particular significance for the process of many membrane phenomena are boundary layers in systems whose solutions are not mixed or their mixing is not effective. In such systems the physico-chemical state of concentration boundary layers depends on the geometrical form of membrane and its location in respect of the direction of gravitation force [8–11]. The state of those layers changes as a result of the impact of gravitation force on them. These changes concern mainly the thickness of layers, concentration drops and their hydrodynamic state. They also depend on whether solution of higher concentration is either over or under the membrane.

Therefore, it was purposeful in our investigation to distinguish two configurations α and β of the system which contains a horizontally situated membrane; α when solution of higher concentration is over the membrane, and when solution of lower concentration is under the membrane; and β with reverse arrangement

of solutions. In the mentioned configurations of the membrane system, boundary layers have different physico-chemical states which determine concentration polarization [2, 4, 7] of this system. The result of this polarization are respective diffusion flows of soluble substance, characteristic of a given configuration, through this system's membrane. They indicate that gravitation force is an important factor which has an impact on transport processes occurring in membrane systems. This effect is manifested through the influence of gravitation force on the concentration boundary layers, which leads to their hydrodynamic stability or instability [8, 10, 12–14]. In the conditions of hydrodynamic instability of those layers in the system, there occurs gravitation convection which causes solutions to be mixed on both sides of the membrane. On the other hand, occurrence of hydrodynamically stable boundary layers in the system causes the solutions not to be mixed, and substance transport in the system has a diffusive character. Such a transport process in the membrane system is also interesting from the biological point of view because it may reveal the participation of the gravitation factor in the process of certain membrane phenomena (e.g., gravielectric effect [15, 16], graviosmosis [17, 18], asymmetry of osmotic and diffusive transport [19]).

The present paper presents results of the experimental investigation of the flows of the soluble substance through a horizontally situated membrane which separates solutions of different concentrations, for configurations (α and β) of the membrane system.

The investigation was carried out by the laser interferometric method in association with a computer analysis of interference images which are related to the areas membrane-solution. The employed methodology of the investigation of concentration boundary layers in dynamic conditions does not disturb processes occurring in a given system.

The results of the investigation permitted to determine the thickness of given layers, solution concentration drops on those layers, as well as the amount of flowing soluble substance, and next the diffusion flux of this substance.

Various ethanol aqueous solutions of different concentrations were used in the investigation in order to fill the compartments of the membrane system. Furthermore, we present a theoretical description of diffusion fluxes through the membrane of a given system, for two of its configurations.

2. Method

The investigation of the transport of soluble substance through a horizontally situated membrane was carried out using the measurement set described in the paper [20]. At present we only recall that it consisted of a Mach-Zehnder interferometer, membrane system, TV CCD camera, and a computer together with a system for processing and analysis of interference images. Recording of these images was carried out for different times and different concentrations of aqueous solutions of ethanol. The basic function of the computer program applied for the acquisition and

analysis of given images was to find the quantities characteristic of near-membrane areas, i.e., thickness of concentration boundary layers, solution concentration drops on these layers, and concentration profiles. In the realization of these functions of the computer program of the analysis of interference images it was taken into consideration that deviation x of the interference line at the determined point of the near-membrane layer with respect to the nearest point of the area of undisturbed lines reflects changes in the refraction coefficient $\Delta n(x, t)$ (between these points) in the direction perpendicular to the membrane surface. These changes of the refraction coefficient are accompanied by concentration changes $\Delta C(x, t)$ which are determined with respect to their linear dependence in accordance with equation [21]:

$$\Delta C(x, t) = a \cdot \Delta n(x, t) = \frac{\lambda \cdot a \cdot x}{h \cdot f},$$

where: λ = the wavelength of He-Ne laser, h = distance between interference lines in the area where they run parallelly, f = thickness of solution layer in the measurement cuvette; a = proportionality factor refractive index and concentration which is $0.319 \cdot 10^6 \text{ mol/m}^3$ for aqueous solutions of ethanol.

In the performed investigations we applied a cellulose membrane of transport coefficients: filtration $L_p = (1.17 \pm 0.05) \cdot 10^{-12} \text{ N}^{-1} \text{ s}^{-1} \text{ m}^3$ and permeability $P = (1.55 \pm 0.25)/10^6 \text{ m/s}$. The active area of the membrane was equal to $7/10^5 \text{ m}^2$. The experimental investigation of the diffusion flux of soluble substance was carried out for two configurations (α, β) of the membrane system.

In configuration α the compartment (B) under the membrane was filled with water, and over the membrane (A) – with aqueous solution of ethanol. In configuration β is reverse filling, i.e., the compartment under the membrane contained solution, and over the membrane – water.

Diffusion flux ($J_{s\alpha}$) of solute which flows through the membrane of the system in configuration α (solutions are not mixed) was determined in accordance with equation:

$$J_{s\alpha} = \frac{\Delta N(t)}{S \cdot \Delta t} = \frac{1}{\Delta t} \left(\sum_i C_{2i}(x, t + \Delta t) \Delta x - \sum_i C_{2i}(x, t) \Delta x \right),$$

where: $\Delta N(t) = S (\sum_i C_{2i}(x, t + \Delta t) \Delta x - \sum_i C_{2i}(x, t) \Delta x)$ is the amount of solute which diffused from the solution of compartment (A) through membrane of area S to solution in compartment (B) in time Δt . $C_2(x, t)$ and $C_2(x, t + \Delta t)$ are solution concentrations at a point situated in distance x_i from the membrane surface, after times t and $t + \Delta t$, respectively, determined on the basis of scanning of interference images. Δx is a scanning step which corresponds to displacement by one pixel equal to $2.3/10^5 \text{ m}$. On the other hand, the solute flux ($J_{s\beta}$) through the membrane of a system in configuration β (solutions are mixed convectively) are determined in accordance with the equation:

Figure 1. Interferograms obtained for configuration (α), using ethanol solution of concentration 250 mol/m³.

$$J_{s\beta} = \frac{V \Delta C_1(t)}{S \cdot \Delta t},$$

where: $\Delta C_1(t)$ is a global change concentration in compartment (A) of the membrane system, V – is solution volume in this compartment.

3. Results of the Investigation and Discussion

We carried out the investigation of the areas membrane – solution in the membrane system for the two of its configurations (α , β) using the above-mentioned measurement set. Aqueous solutions of ethanol were used in the performed investigation. The results of the computer analysis of interference images referring to the areas membrane – solution of the investigated membrane system enabled us to determine concentration shifts on boundary layers and on the membrane, concentration profiles, and thickness of given layers. On the basis of these results we determined the diffusion flux in the system in configuration (α), and the other in configuration (β).

3.1. INTERFEROGRAMS

In Figure 1 are represented some selected interferograms which refer to the membrane system in configuration (α) – compartment (A) of this system contained aqueous solution of ethanol of concentration 250 mol/m³, and compartment (B) – contained water. The successive interferograms, a, b, and c were recorded after times equal to 4, 10, and 20 minutes, respectively. A wide set of interferograms obtained for different concentrations of ethanol (125 mol/m³, 500 mol/m³, 750 mol/m³) in the system of a given configuration was presented in the paper [20]. The courses of interference lines on the represented interferograms indicate distinct changes in the solution concentrations in boundary layers.

Figure 2. Interferograms obtained for configuration (β), using ethanol solution of concentration 250 mol/m^3 .

Figure 3. Interferograms obtained for configuration (β), using ethanol solution of concentration 500 mol/m^3 .

Figure 2 represents some selected interferograms referring to the membrane system in configuration β in which ethanol solution of concentration 250 mol/m^3 was in compartment (B). Also, the next set of interferograms contained in Figure 3 refers to the membrane system of configuration β obtained by use of ethanol solution of concentration 500 mol/m^3 . The interferograms a, b, and c contained in Figures 2 and 3 were recorded after the time intervals of 4, 10, and 20 minutes, respectively. The courses of the interference lines on these interferograms indicates that on both sides of the membrane are formed boundary layers whose thickness is significantly smaller (10 times) than that of the layers observed on the interferograms represented in Figure 1. The observed interference in the courses of interference lines indicates that in the system occurs convective mixing of solution caused by the hydrodynamic instability of boundary layers [8, 10, 11, 13, 14].

Figure 4. Concentration shift in boundary layers as a function of distance from membrane surface: A – over membrane, B – under membrane, in configuration (α), obtained for different time intervals: ($\circ$) – 4 minutes, (Δ) – 10 minutes, ($\square$) – 20 minutes, and ($*$) – 40 minutes, using ethanol solution of concentration 250 mol/m^3 .

3.2. CONCENTRATION SHIFTS IN NEAR-MEMBRANE BOUNDARY LAYERS

The results of the computer analysis of interference images which refer to near-membrane areas enabled us to obtain data concerning concentration shifts $\Delta C_1(x, t_0)$ and $\Delta C_2(x, t_0)$ in near-membrane layers for different times of experiment t_0 . In Figure 4A are shown the results of the investigation of concentration shifts $\Delta C_1(x, t_0)$ in the boundary layer situated over the membrane of configuration α which contains ethanol solution of concentration 250 mol/m^3 , and in Figure 4B are shown the results of the investigation of concentration shifts $\Delta C_2(x, t_0)$ which occur in a layer situated under the membrane of a given system. The results of the investigation of $\Delta C_1(x, t_0)$ and $\Delta C_2(x, t_0)$ indicate that the shift of solution

Figure 5. Concentration shift in boundary layers as a function of distance from membrane surface: A – over membrane, B – under membrane, in configuration (β), obtained for different time intervals: (○) – 4 minutes, (△) – 10 minutes, (□) – 20 minutes, and (*) – 30 minutes, using ethanol solution of concentration 250 mol/m^3 .

concentration occurs mainly on near-membrane boundary layers and not on the membrane. This point to the occurrence of high concentration polarization of the membrane system.

The subsequent results of concentration shifts of $\Delta C_1(x, t_0)$ and $\Delta C_2(x, t_0)$ in the near-membrane layers of the system in configuration β are contained in Figure 5. Figures 5A and 5B represent the results of the investigation of $\Delta C_1(x, t_0)$ and $\Delta C_2(x, t_0)$, respectively, which refer to near-membrane layers in the system containing ethanol solution of concentration 250 mol/m^3 . The results of this investigation indicate that boundary layers which occur in the surrounding of the membrane of a given system have a relatively small thickness and do not exceed the value of $(0.4 \pm 0.1) \text{ mm}$. However, drops of solution concentrations on these

Figure 6. Dependences of diffusion flux on time, for configuration (α). Investigation results: experimental, obtained for ethanol solutions of concentrations (o) – 250 mol/m³, (Δ) – 500 mol/m³, ($\square$) – 750 mol/m³; theoretical – continuous lines 1, 2, and 3 – for respective concentrations.

layers are much smaller than that which occurs on the membrane of a given system. The observed fluctuations of the values $\Delta C_1(x, t_0)$ and $\Delta C_2(x, t_0)$ point to the occurrence of gravitation convection [11, 13, 14] in near-membrane areas of the system, which causes mixing of solutions on both sides of the membrane.

3.3. DIFFUSION FLUXES OF SOLUTE

The results of the computer analysis of interferograms referring to the membrane system of configuration α enabled us to determine the courses of changes in diffusion flux through this system's membrane in the time function. The results of this investigation are contained in the point form in Figure 6. The investigation was carried out with use of ethanol, solution of concentrations 250 mol/m³, 500 mol/m³ and 750 mol/m³. In this way it was shown that diffusion flux decreases rapidly for initial times, and for later times – it decreases more slowly, approaching after some time the steady state. Such a course of changes in diffusion flux in a given configuration of the system is predetermined by significant concentration changes on the boundary membrane – solution.

As solutions are not mixed, then on one side of the membrane occurs depletion of soluble substance, and on the other side occurs accumulation. These processes lead to a significant reduction in the difference of concentrations on the membrane, the effect of which being the observed course of changes in diffusion flux.

The results of the analogous dependence of solute flux but referring to the membrane system in configuration β are presented in the form of discrete points

Figure 7. Dependences of diffusion flux on time, for configuration (β). Investigation results: experimental, obtained for ethanol solutions of concentrations ($\circ$) – 250 mol/m³, (Δ) – 500 mol/m³, ($\square$) – 750 mol/m³; theoretical – continuous lines 1, 2, and 3 – for respective concentrations.

in Figure 7. The results of this investigation were obtained for the same solution as for those in configuration α . The presented results of this investigation indicate small changes in diffusion flux in time.

In the given configuration of the membrane system, the boundary layers which are formed in the surrounding of the membrane are hydrodynamically unstable. Therefore, in a given configuration there occurs gravitation convection which causes solutions on both sides of the membrane to be mixed. Concentration shifts on boundary layers of a given system are relatively small and they hardly depend on time. The observed diffusion flux has, within the boundaries of measurement error, has a quasi-stationary character. The measured values of diffusion flux in a given configuration are about 2.5 times greater than the flux in configuration (α) for these same initial solution concentrations.

4. Results of the Theoretical Investigation

DESCRIPTION OF ANALYTICAL DIFFUSION FLUXES

In order to describe mathematical diffusion fluxes in both configurations of the membrane system, we shall make use of theoretical considerations concerning time-dependent processes of transport of substance through membranes presented in papers [22–24]. This description will be presented separately for each configuration (α and β) of the membrane system.

Description for configuration α . As the solutions in the considered configuration of the membrane system are not mixed, we can distinguish three separate regions which include membrane and two near-membrane regions. Solute transport in the mentioned regions occurs in accordance with II Fick's law. Thus we may write:

$$\frac{\partial C_i}{\partial t} = D_i \frac{\partial^2 C_i}{\partial x^2} \quad \begin{cases} i = 1 & -\infty < x < 0 \\ i = m & 0 < x < 1 \\ i = 2 & 1 < x < +\infty \end{cases} \quad (1)$$

where: $i = 1$ concerns region over the membrane, and $i = 2$ – under the membrane, and $i = m$ – membranes.

In our considerations we assume that coefficients of solute diffusion in near-membrane regions are identical $D_1 = D_2 \equiv D$ and constant. Also, the coefficient of solute diffusion (D_m) is constant in the membrane.

The boundary conditions for a give configuration of the membrane system are as follows:

$$D \frac{\partial C_1}{\partial x} \Big|_{x=0} = D_m \frac{\partial C_m}{\partial x} \Big|_{x=0} \quad (2)$$

$$D \frac{\partial C_2}{\partial x} \Big|_{x=1} = D_m \frac{\partial C_m}{\partial x} \Big|_{x=1} \quad (3)$$

$$k_\alpha C_1(0, t) = C_m(0, t) \quad t > 0 \quad (4)$$

$$k_\alpha C_2(1, t) = C_m(1, t) \quad t > 0 \quad (5)$$

where k_α is the solute partition coefficient defined by the equations (4) and (5). The initial conditions for a given configuration of the investigated system are given by the expressions:

$$C_1(x, 0) = C_{01} \quad -\infty < x < 0 \quad (6)$$

$$C_m(x, 0) = -k_\alpha C_{01}(x/1) + k_\alpha C_{01} \quad 0 < x < 1 \quad (7)$$

$$C_2(x, 0) = C_{02} = 0 \quad 1 < x < +\infty \quad (8)$$

where C_{01} is concentration of solution which filled compartment A of the membrane system.

In the considered membrane system volume flux (J_v) is equal to zero. This condition can also be presented in the following form:

$$J_v = \sum_{i=1}^2 J_i \bar{V}_i = 0, \quad i = 1, 2 \quad (9)$$

where: $\bar{V}_i$ is partial volume of i th substance, J_i – diffusion flux of i th substance.

In accordance with I Fick's equation this flux is equal to:

$$J_i(t) = -D_{mi} \nabla C_i, \quad i = 1, 2. \quad (10)$$

Solution of the equation set (1–8) carried out by the method of Laplace's transform [23, 24] enabled us to obtain expressions $C_1(x, t)$, $C_2(x, t)$ and $C_m(x, t)$ [22] which describe concentration profiles in the mentioned regions. The experimental and theoretical investigation of profiles $C_1(x, t)$ and $C_2(x, t)$ were also presented in the paper [20].

As a result of algebraic transformations of expressions $C_1(x, t)$, $C_2(x, t)$ and applying Equation (10) we obtained an expression which describes time dependences of solute diffusion flux through the system's membrane in configuration α . It has the form:

$$J_{s\alpha}(t) = \frac{D_m k_\alpha}{1} C_{01} \left[1 - \frac{8\sqrt{D_m t}}{1} \cdot \frac{r^2}{r^2 - 1} \sum_{n=1}^{\infty} \left(\frac{1-r}{1+r} \right)^n \cdot \left\{ \operatorname{ierfc} \left(\frac{n \cdot 1}{2\sqrt{D_m t}} \right) - \frac{1}{\sqrt{\pi}} \right\} \right] \quad (11)$$

where:

$$r = k_\alpha \sqrt{\frac{D_m}{D}}, \quad \operatorname{ierfc}(y) = \int_y^{\infty} \operatorname{erfc}(\xi) d\xi = \frac{1}{\sqrt{\pi}} \exp(-y^2) - y \operatorname{erfc}(y).$$

On the basis of Equation (11) computer numerical calculation of diffusion flux in time function was performed for fixed concentrations of ethanol solutions. The results of these calculations are represented in Figure 6 (continuous lines). Curves 1, 2, and 3 in this figure were obtained for ethanol concentrations which were 250 mol/m³, 500 mol/m³, and 750 mol/m³, respectively. The calculations included values of the diffusion coefficients of solute: in water $D = 1.28 \cdot 10^{-9}$ m²/s [25], in membrane $3.35 \cdot 10^{-10}$ m²/s [20], and membrane thickness $l = (160 \pm 5) \cdot 10^{-6}$ m. The partition coefficient of solute (k_α) was treated as a parameter. Its value $k_\alpha = 0.6$ secures a good conformity of the theoretical results with experimental ones for a given configuration of the membrane system.

Description for configuration β . The results of the experimental investigation presented in this paper for the areas membrane – solute, in configuration β indicate that boundary layers occur in the membrane surrounding. Furthermore, we can distinguish in this configuration of the membrane system three regions, i.e. a membrane of thickness l and two boundary layers of thickness δ each.

Substance transport through membrane and the surrounding layers are described by diffusion equation (1).

Whereas boundary conditions for a given configuration are following:

$$\frac{V}{k_\beta S} \cdot \frac{\partial C_m(0, t)}{\partial t} = -D_m \left. \frac{\partial C_m}{\partial x} \right|_{x=0} \quad (12)$$

$$\frac{V}{k_\beta S} \cdot \frac{\partial C_m(1, t)}{\partial t} = D_m \frac{\partial C_m}{\partial x} \Big|_{x=1} \quad (13)$$

Initial conditions are given by the expressions:

$$C_1(x, 0) = C_{01} = 0 \quad -\infty < x < 0 \quad (14)$$

$$C_m(x, 0) = -k_\beta C_{02}(x/1) + k_\beta C_{02} \quad 0 < x < 1 \quad (15)$$

$$C_2(x, 0) = C_{02} \quad 1 < x < +\infty \quad (16)$$

where k_β is the solute partition coefficient for configuration β , C_{02} – solution concentration which filled compartment B of the membrane system.

Solution of the equation set (1) and (11–15), presented by Hoogervorst et al. [26], allowed to obtain the expression which describes the dependence of the solute concentration difference on membrane on time. It has the form:

$$\Delta C(t) = C_{02} \sum_{n=1}^{\infty} A_n \exp - \frac{Q_n^2 D_m t}{l^2} \quad (17)$$

where: A_n , Q_n are quantities connected with experimental conditions and the parameters of the membrane system. A detailed description of these quantities is presented by Hoogervorst et al. [26].

Taking into consideration a weak dependence on the time of concentration shifts on boundary layers as well as changes in time of diffusion flux in a given configuration of the membrane system, it is possible to assume that substance transport has a quasi stationary character. Such a character of substance in a given configuration permits to simplify Equation (17) as a result of which it assumes the following form [27]:

$$\Delta C(t) = C_{02} \exp - b P_{ms} t \quad (18)$$

where $b = 2S/V$, and P_{ms} is the apparent coefficient of permeability of membrane and diffusion layers which fulfil the equation [2, 4, 19]:

$$\frac{1}{P_{ms}} = \frac{1}{k_\beta D_m} + \frac{2\delta}{D}. \quad (19)$$

Therefore, solute flux in the conditions of quasi stationary transport can be expressed [24, 27] by the equation:

$$J_{s\beta}(t) = P_{ms} \Delta C(t) \quad (20)$$

where $\Delta C(t)$ is a sum of concentration drops on membrane and on near-membrane layers.

Taking into consideration in Equation (20) expressions (18) and (19), we get:

$$J_{s\beta}(t) = \frac{k_{\beta} D_m D}{lD + 2\delta k_{\beta} D_m} C_{02} \exp - \frac{b \cdot k_{\beta} D_m D \cdot t}{1D + 2\delta k_{\beta} D_m}. \quad (21)$$

On the basis of Equation (21) computer numerical calculations of diffusion flux in time function were performed for fixed concentrations of ethanol solution.

The results of these calculations are represented in Figure 7 (continuous lines). Curves 1, 2, and 3 in this figure were obtained for concentrations of solutions which were 250 mol/m³, 500 mol/m³, and 750 mol/m³. The values of coefficients D and D_m as well as membrane thickness were the same as those in the calculation for configuration α . The partition coefficient of solute k_{β} is a parameter. Its value $k_{\beta} = 0.9$ secures a good conformity of experimental results with theoretical ones.

5. Results

1. The presented method of the investigation permits precise measurements of the physico-chemical state of concentration boundary layers in the membrane system. The results of this investigation enable one to trace the kinetics of substance transport through membrane and the adjoining layers.
2. The course of changes of diffusion flux in time function for configuration (α) is determined mainly by concentrations on the interface membrane – solution. A rapid drop of the flux value for initial time intervals indicates that the diffusion flow of solute through membrane dominates over free diffusion in solution. In a given configuration on one side of the membrane there occurs solute accumulation and on the other side – its depletion, which is evidence of high concentration polarization of a given system.
3. The course of changes in time of diffusion flux in configuration (β) indicates that convective mixing of solutions causes increase in the intensity of solute transport through membrane ($J_{s\beta} > J_{s\alpha}$). Small concentration drops on diffusion layers also indicate that concentration polarization in configuration (β) is much smaller than in configuration (α).
4. A good conformity of the experimental results of dependences $J_{s\alpha}(t)$ and $J_{s\beta}(t)$ with the theoretical results, with one parameter of their adjustment, i.e. the partition coefficient of solute equal to $k_{\alpha} = 0.6$ and $k_{\beta} = 0.9$, respectively, confirms correctness of the presented description of these fluxes.

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