

Rayleigh-Benard Convective Instability: Concentration Rayleigh Number for Isothermal Passive Transmembrane Transport Processes

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Summary

We derive a formula of the concentration Rayleigh number for isothermal membrane transport processes. The formula include diffusive permeability coefficient (ω_{ks}), concentration polarization coefficient (ϑ_k^i) and solution parameters: density (ρ), kinematic viscosity (ν), diffusion coefficient (D_{ks}). Coefficients ω_{ks} and ϑ_k^i were determined for configurations A and B of the single-membrane cell system, which consisted a flat polymeric membrane and glucose in $0.2 \text{ mol}\cdot\text{l}^{-1}$ aqueous ethanol solutions. In configuration A water was placed in compartment above the membrane and the solution below. In configuration B the arrangement of water and solution was reversed. The results of calculation showed that critical value of concentration Rayleigh number (R_s^i) in configuration A is $(R_1^A)_{crit.} = 277.1$ and in the configuration B – $(R_1^B)_{crit.} = 225.2$.

Konwekcyjna niestabilność Rayleigha-Benarda: stężeniowa liczba Rayleigha dla procesów izotermicznego transportu membranowego

Streszczenie

Wyprowadzono formułę dla stężeniowej liczby Rayleigha dla procesów izotermicznego transportu membranowego. Formuła zawiera współczynnik przepuszczalności dyfuzyjnej, ω_{ks} , współczynnik polaryzacji stężeniowej (ϑ_k^i) oraz parametry roztworu: gęstość (ρ), lepkość

kinematyczną (ν), współczynnik dyfuzji (D_{ks}). Współczynniki ω_{ks} oraz ϑ_k^i określono dla konfiguracji A i B komórki jednomembranowej, która zawierała płaską membranę polimerową i roztwór glukozy w $0,2 \text{ mol}\cdot\text{l}^{-1}$ wodnym roztworze etanolu. W konfiguracji A wodę umieszczono w przedziale nad membraną a roztwór – pod membraną. W konfiguracji B kolejność ustawienia wody i roztworu była odwrotna. Otrzymane rezultaty pokazują, że krytyczna wartość stężeniowej (R_s^i) wynosi $(R_1^A)_{crit.} = 277,1$ a w konfiguracji A – $(R_1^B)_{crit.} = 225,2$.

Key words: membrane transport, concentration boundary layer, natural convection, concentration Rayleigh number.

Słowa kluczowe: transport membranowy; stężeniowa warstwa graniczna; konwekcja swobodna; stężeniowa liczba Rayleigha

1. INTRODUCTION

Passive membrane transport in membrane systems (biological and artificial) generated by concentration gradient leads to the concentration boundary layers (CBL) formation on both sides of horizontally or vertically mounted membrane [1-7]. These layers are regions in the external solutions, adjacent to the membrane interfaces and act as pseudo-membranes in series with the physical membrane. Consequently, they can cause substantial reductions in the transmembrane flows [8-13].

The study of the CBLs formation may be performed in the artificial systems in terrestrial conditions, using the single- or double-membrane osmotic-diffusive cell. The directly observed effects of CBL creation in different configurations of a single-membrane system are: curves of interference stripes in the area near the membrane received on Mach-Zehnder interferograms [5,17], changes in the volume and solute fluxes [11,12] and in the diffusive membrane potential [15].

Gravitational effects on transmembrane volume and solute flows of the nonelectrolyte and electrolyte solutions in single-membrane system were reported in previous papers [11,12,14]. From these papers, results that the influence of the gravitational field on transmembrane transport manifests itself in Rayleigh-Taylor instability [6,11], rectifying and amplifying properties of osmotic and solute fluxes [11,12] and gravielectric effect [15]. These

findings were interpreted in terms of a gravitational instability, which leads to reduction boundary layer dimensions and increases the permeability coefficient (Ω_{ks}^i) of the complex system: boundary layer/membrane/boundary layer. Model equations of the transport through a horizontally mounted membrane were presented in [11,12]. In these equations a concentration-gradient Rayleigh number was used. They found transition from stable to unstable behaviour when experimental parameters exceed a critical Rayleigh number.

The mechanism of the natural convection generation in a horizontal fluid layer heated from below has been one of the primary problems of fluid dynamics since Bénard made first quantitative experiments to examine the conditions for the pattern appearance [18]. Rayleigh [19] made a theoretical analysis by modelling Benard's experiments and proved that the thermal convection is induced by gravitational instability of the motionless thermal conduction state when a dimensionless parameter R_T (temperature Rayleigh number) exceeds a critical value R_T^c . Rayleigh [19] and Taylor's [20] analysis and most subsequent papers [21-23] are framed in terms of $(\partial\rho/\partial T)$, the density change with temperature. The linear stability theory proposed by Rayleigh explained the mechanism of the occurrence of the thermal convection and was proved to be valid and very useful. Later, Pearson [20] noted that gravitational instabilities could be driven by $(\partial\rho/\partial C)$, the density change with concentration. The concentration Rayleigh number is defined as [25-27]

$$R_C = \frac{g\alpha_c\beta_c d^4}{D\nu} \quad (1)$$

where g is the gravitational acceleration, d the fluid depth along the gravitational (z) direction, $\alpha_c = (\partial\rho/\partial C)/\rho$ the variation of density with concentration, $\beta_c = \partial C/\partial z$ the concentration gradient, D the solute diffusion coefficient and ν kinematic viscosity.

The paper is organized as follows. In sections 2 and 3 we obtain the formula of the concentration Rayleigh number for isothermal membrane transport processes. The formula include diffusive permeability coefficient, ω_{ks} , concentration polarization coefficient, ϑ_k^i and solution parameters (density, ρ ; kinematic viscosity, ν ; diffusion coefficient, D_{ks}). In Sections 4, 5 and 6 we present the experimental procedure with the results on the concentration characteristic of solute fluxes, the concentration characteristic concentration polarization coefficients of CBL/M/CBL complex. Coefficients ω_{ks} and ϑ_k^i were determined in single-membrane cell system which consisted a flat polymeric membrane and glucose in $0.2 \text{ mol}\cdot\text{l}^{-1}$ aqueous ethanol solutions. Configurations A and B of single-membrane cell system in

gravitational field were studied. In configuration A water was placed in compartment above the membrane and the solution below. In configuration B the arrangement of water and solution was reversed. The predictions of our theoretical model are compared with the experimental data. Our study we summarize in Section 7.

2. MEMBRANE SYSTEM

Let us consider the configurations A and B of the single-membrane system represented in Figure 1. We assume that this membrane system in both configurations contains ternary, non-ionic and heterogeneous (not mechanically stirred) solutions, separated by an isotropic and symmetric membrane (M) of thickness d . In both configurations the membrane has horizontally oriented planes and the respective concentration gradients are antiparallel and parallel to the gravitational force. The transport processes are isothermal and stationary and no chemical reactions occur in the solutions. In this single-membrane system water and the dissolved substance diffusing across the membrane will lead to the formation of concentration boundary layers (CBL). In this steady state system, the layers l_l^i and l_h^i of thickness δ_l^i and δ_h^i (superscript $i=A, B$ pertain to configurations A and B) are formed on both sides of the membrane. These layers can be treated as pseudo-membranes. The implication from the above is that the concentrations of solutions at the membrane-layers interfaces are different from those in the outside layers (in the bulk solutions). We denote respectively by C_{sl}^A , C_{sh}^A (in configuration A) and by C_{sl}^B and C_{sh}^B (in configuration B), the concentrations of solutions at interfaces: membrane/concentration boundary layers, while the concentrations of solutions outside the layers are denoted by C_{sl} and C_{sh} ($C_{sh} > C_{sl}$), ($s=1, 2$). For configurations A and B and for any solutions, the following relations hold

$$C_{sh} > C_{sh}^B > C_{sl}^B > C_{sl}, C_{sh} > C_{sh}^A > C_{sl}^A > C_{sl} \quad (2)$$

Besides, for solutions whose densities are proportional or inversely proportional to their concentration, the following relations are fulfilled, respectively

$$C_{sl}^B > C_{sl}^A > C_{sl}, C_{sh} > C_{sh}^B > C_{sh}^A \quad (3)$$

$$C_{sl}^A > C_{sl}^B > C_{sl}, C_{sh} > C_{sh}^A > C_{sh}^B \quad (4)$$

The membrane is characterised by the hydraulic permeability (L_p), reflection (σ_s) and diffusive permeability (ω_{ks}) coefficients. The solute fluxes across membrane are denoted by J_{sm}^A and J_{sm}^B . The reflection and diffusive permeability coefficients of the complexes: concentration boundary layer/membrane/concentration boundary layer ($l_l^A/M/l_h^A$ and $l_l^B/M/l_h^B$) are denoted by σ_s^A ,

σ_s^B , Ω_{ks}^A and Ω_{ks}^B , respectively. The solute fluxes across the complexes $l_l^A/M/l_h^A$ and $l_l^B/M/l_h^B$ are denoted by J_s^A and J_s^B , respectively. The layers l_l^A , l_l^B , l_h^A and l_h^B are characterised by diffusive permeability coefficients: $(\omega_{ks}^A)_l$, $(\omega_{ks}^B)_l$, $(\omega_{ks}^A)_h$ and $(\omega_{ks}^B)_h$ respectively. The solute fluxes across layers l_l^A , l_l^B , l_h^A and l_h^B are denoted by J_{sl}^A , J_{sl}^B , J_{sh}^A and J_{sh}^B , respectively.

The basic manner of description of membrane transport processes of non-homogeneous non-electrolyte solutions, originated from non-equilibrium thermodynamics of irreversible processes, is the modified Kedem-Katchalsky model equation [12]. In the case of osmotic-diffusive membrane transport of ternary non-electrolyte solutions this model consists of equations

$$J_s^i = RT \sum_{k=1}^2 \zeta_k^i \omega_{ks} (C_{kh} - C_{kl}) + J_v^i (1 - \zeta_s^i \sigma_s) \bar{C}_s \quad (5)$$

where J_s^i and J_v^i are the solute and volume fluxes, respectively; RT is the product of the constant and thermodynamic temperature; C_{sh} and C_{sl} (with $C_{sh} > C_{sl}$) are higher (superscript h) and lower (superscript l) the bulk solution concentrations, ω_{ks} is the diffusion permeability coefficient of the membrane for s -th substance, which passes across membrane, under the influence of gradient of the k -th substance osmotic pressure difference ($\omega_{ks} = \omega_{sk} \bar{C}_s \cdot \bar{C}_k^{-1}$) and $s \neq k=1, 2, \dots, n$; ζ_k^i ($0 \leq \zeta_k^i \leq 1$) is the concentration polarisation coefficient, $\Omega_{ks}^i = \zeta_k^i \omega_{ks}$ is the diffusion permeability coefficient of the complex $l_l^i/M/l_h^i$ for s -th substance, which passes across complex $(l_l^i/M/l_h^i)$ respectively, under the influence of gradient of the k -th substance osmotic pressure difference, L_p is the hydraulic permeability coefficient of the membrane, σ_s is the reflection coefficient of the membrane, $\sigma_s^i = \zeta_s^i \sigma_s$ is reflection coefficient of the of the complex $l_l^i/M/l_h^i$ for s -th substance, $\bar{C}_s = (C_s^h - C_s^l) \{ \ln(C_s^h / C_s^l) \} \approx (C_s^h + C_s^l) / 2$ is the average concentration of solution for the s -th substance, $(n+1)$ -th component is a solvent, for which $\sigma_{n+1} = 0$.

For situation in which membrane separate the diluted solutions the following relation is fulfilled

$$RT \sum_{k=1}^2 \zeta_k^i \omega_{ks} (C_{kh} - C_{kl}) \gg J_v^i (1 - \zeta_s^i \sigma_s) \bar{C}_s \quad (6)$$

On the base of equations (5) and (6) for membrane system presented in Figure 1 we can write

$$J_{sl}^i = RT \sum_{k=1}^2 (\omega_{ks}^i)_l (C_{kh}^i - C_{kl}^i) \quad (7)$$

$$J_{sh}^i = RT \sum_{k=1}^2 (\omega_{ks}^i)_h (C_{kh}^i - C_{kl}^i) \quad (8)$$

$$J_s^i = RT \sum_{k=1}^2 \zeta_s^i \omega_{ks} (C_{kh} - C_{kl}) \quad (9)$$

where J_{sl}^i , J_{sh}^i and J_s^i are the solute fluxes across the layer l_l^i , the layer l_h^i and the complex $l_l^i/M/l_h^i$, respectively; $(\omega_{ks}^i)_l$ and $(\omega_{ks}^i)_h$ are the diffusive permeability coefficients of the layer l_l^i and the layer l_h^i , respectively; C_{sl}^i , C_{sh}^i are the concentrations of solutions at interfaces: membrane/concentration boundary layers; C_{sl} and C_{sh} are the concentrations of solutions outside the CBLs.

The diffusive permeability coefficients $(\omega_{ks}^i)_l$, ω_{ks} and $(\omega_{ks}^i)_h$, can be expressed as follows [28]

$$(\omega_{ks}^i)_l = \frac{(D_{ks}^i)_l}{RT \delta_l^i} \quad (10)$$

$$\omega_{ks} = \frac{D_{ks}}{RT \delta_m} \quad (11)$$

$$(\omega_{ks}^i)_h = \frac{(D_{ks}^i)_h}{RT \delta_h^i} \quad (12)$$

where $(D_{ks}^i)_l$, D_{ks} and $(D_{ks}^i)_h$ are the coefficients of solute diffusion in the layer l_l^i , in the membrane M and in the layer l_h^i , respectively; δ_l^i , δ_m and δ_h^i are the thickness of the layer l_l^i , the membrane M and the layer l_h^i , respectively.

For coefficient ζ_s^i the next relation is valid

$$\zeta_s^i = \left\{ 1 + RT \omega_{ss} \left[\frac{\delta_l^i}{(D_{ss}^i)_l} + \frac{\delta_h^i}{(D_{ss}^i)_h} \right] \right\}^{-1} \quad (13)$$

where $s=1$ or 2 .

The non-diagonal coefficients are smaller by two orders of magnitude than the diagonal coefficients [12]. Thus, here is the basis of reduction of equations (7) – (9) accepting $(\omega_{21}^i)_l \ll (\omega_{11}^i)_l$, $(\omega_{12}^i)_l \ll (\omega_{22}^i)_l$, $(\omega_{21}^i)_h \ll (\omega_{11}^i)_h$, $(\omega_{12}^i)_h \ll (\omega_{22}^i)_h$, $\omega_{21} \ll \omega_{11}$ and $\omega_{12} \ll \omega_{22}$.

3. EQUATION FOR RAYLEIGH NUMBER

For the concentration boundary layers l_l^i and l_h^i equations (1) can be written in the following forms

$$(R_s^i)_l = \frac{g(\alpha_s^i)_l(\beta_s^i)_l(\delta_l^i)^4}{(D_{ss}^i)_l \nu_l} \quad (14)$$

$$(R_s^i)_h = \frac{g(\alpha_s^i)_h(\beta_s^i)_h(\delta_h^i)^4}{(D_{ss}^i)_h \nu_h} \quad (15)$$

where g is the gravitational acceleration, $(\delta_s^i)_l$ and $(\delta_s^i)_h$ are the concentration boundary layers (l_l^i and l_h^i) thickness, $(D_{ks}^i)_l$ and $(D_{ks}^i)_h$ are the solute diffusion coefficients in layers l_l^i and l_h^i , ν_l and ν_h are the kinematic viscosity coefficients, $s=1, 2$. $(\alpha_s^i)_l = (\partial \rho_l / \partial C_{sl})^i / \rho_l$ and $(\alpha_{sh}^i) = (\partial \rho_h / \partial C_{sh})^i / \rho_h$ represent the variation of solution density with configuration and concentration, while $(\beta_s^i)_l = (\partial C_{sl} / \partial z)^i$ and $(\beta_s^i)_h = (\partial C_{sh} / \partial z)$ represent the concentration gradient along the vertical axis, respectively. When $(\alpha_s^i)_l(\beta_s^i)_l$ and $(\alpha_s^i)_h(\beta_s^i)_h$ are negative, i.e., the near membrane layer is denser than the liquid beneath it, convective instability occurs when $(R_s^i)_l$ and $(R_s^i)_h$ exceeds a critical value. This convection reduces near membrane layers thickness, thereby increasing transmembrane volume and solute fluxes.

For $(\alpha_s^i)_l(\beta_s^i)_l$ and $(\alpha_s^i)_h(\beta_s^i)_h$ the following relations hold [11]

$$(\alpha_s^i)_l(\beta_s^i)_l = \frac{\rho_l^i - \rho_l}{\rho_l \delta_l^i} \quad (16)$$

$$(\alpha_s^i)_h(\beta_s^i)_h = \frac{\rho_h - \rho_h^i}{\rho_h \delta_h^i} \quad (17)$$

where ρ_l^i and ρ_h^i are the densities of solutions at interfaces: membrane/concentration boundary layers.

In order to estimate $\rho_l^i - \rho_l$ and $\rho_h - \rho_h^i$ in steady-state, in which the condition is fulfilled

$$J_{sl}^i = J_{sh}^i = J_s^i \quad (18)$$

we will make following consideration.

If we include equations (7), (8) and (9) in equation (18) for $s=1, 2$ we will obtain relations

$$C_{sl}^i - C_{sl} = \gamma_1 (C_{sh} - C_{sl}) \quad (19)$$

$$C_{sh} - C_{sh}^i = \gamma_2 (C_{sh} - C_{sl}) \quad (20)$$

where $\gamma_1 = \frac{RT \zeta_s^i \omega_{ss} \delta_l^i}{(D_{ss}^i)_l}$, $\gamma_2 = \frac{RT \zeta_s^i \omega_{ss} \delta_h^i}{(D_{ss}^i)_h}$, $(D_{ss}^i)_l$ and $(D_{ss}^i)_h$ are the diagonal coefficients of s -th substance diffusion in the layers l_l^i and l_h^i , respectively. Combining equations (14) – (17) we obtain

$$(R_s^i)_l = \frac{\rho_l^i - \rho_l}{\rho_l} \cdot \frac{g(\delta_l^i)^3}{(D_{ss}^i)_l v_l} \quad (21)$$

$$(R_s^i)_h = \frac{\rho_h - \rho_h^i}{\rho_h} \cdot \frac{g(\delta_h^i)^3}{(D_{ss}^i)_h v_h} \quad (22)$$

Let us assume that, in a steady state, the diffusion coefficient in the solution does not depend on both gravitational direction and solute concentration

$$(D_{ss}^i)_l = (D_{ss}^i)_h = D_{ss} \quad (23)$$

and for complex of $l_l^i/M/l_h^i$, in steady-state, the following conditions are fulfilled

$$(R_s^i)_l = (R_s^i)_h = R_s^i \quad (24)$$

Assuming that the thickness of both boundary layers are the same and using equation (13) we can write

$$\delta_l^i = \delta_h^i = \frac{D_{ss}}{2RT\omega_{ss}} \left(\frac{1}{\zeta_s^i} - 1 \right) \quad (25)$$

For small concentration the dependence of density on concentration is linear, so

$$\rho = \rho_o + \sum_{s=1}^n \frac{\partial \rho}{\partial C_s} \cdot C_s \quad (26)$$

where ρ and ρ_o are densities of solution and solvent respectively, and $(\partial \rho / \partial C_s) = \text{const.}$

For ternary solutions equation 26 we can write in following form

$$\rho_l^i - \rho = RT \delta_l^i \sum_{s=1}^2 \frac{\zeta_s^i \omega_{ss}}{D_{ss}^i} \left(\frac{\partial \rho}{\partial C_s} \right) (C_{sh} - C_{sl}) \quad (27)$$

$$\rho - \rho_h^i = RT \delta_h^i \sum_{s=1}^2 \frac{\zeta_s^i \omega_{ss}}{D_{ss}^i} \left(\frac{\partial \rho}{\partial C_s} \right) (C_{sh} - C_{sl}) \quad (28)$$

Introducing the equations (19), (20), (26) and boundary layer thickness in the form (25) into equations (21) and (22), we obtain the relation

$$R_s^i = \mathfrak{S}_s \left[\sum_{k=1}^n \left(\frac{\partial \rho}{\partial C_k} \right) (1 - \zeta_k^i) (C_{kh} - C_{kl}) \right] \cdot \frac{(1 - \zeta_s^i)^3}{(\zeta_s^i)^3} \quad (29)$$

where $\mathfrak{S}_s = \frac{g(D_{ss})^2}{16\nu\rho(RT)^3(\omega_{ss})^3}$.

We have obtained equation describing the concentration Rayleigh number for isothermal passive transmembrane transport processes. The equations can be used to calculate the critical value of Rayleigh number in any gravitational configuration of a single-membrane system.

4. MATERIALS AND METHODS

Studies on diffusive flows across a horizontally-mounted membrane were carried out by means of the measuring apparatus whose detailed description has been given in previous papers [11]. Experiments were performed with flat sheet *Nephrophane* (cellophane) dialyzer membrane. The membrane was housed between two Plexiglas vessels, each with a volume of 200 ml, with $3.36 \pm 0.2 \text{ cm}^2$ of available membrane area. An additional support, consisting of a large mesh screen on each side of the membrane, was used to prevent buckling or tearing of thin dialysis membranes. The stirring speed in each chamber was maintained at approximately 500 rpm using independently controlled stirrer motors. One of vessels of the membrane system contained aqueous glucose and/or ethanol solution at varied concentration. This vessel was attached to a calibrated pipette, which allowed to measure the volume with accuracy $\pm 0.5 \text{ mm}^3$. The second vessel in all experiments contained pure water. This vessel was contacted to an external reservoir at the same height as the pipette. Values of the transport parameters of this membrane for glucose (subscript 1) and ethanol (subscript 2) are following: $L_p = (5.0 \pm 0.2) \cdot 10^{12} \text{ m}^3 \cdot \text{N}^{-1} \cdot \text{s}^{-1}$, $\sigma_1 = (6.8 \pm 0.2) \cdot 10^2$, $\sigma_2 = (2.5 \pm 0.1) \cdot 10^2$, $\omega_{11} = (8.0 \pm 0.3) \cdot 10^{10} \text{ mol} \cdot \text{N}^{-1} \cdot \text{s}^{-1}$, $\omega_{22} = (2.00 \pm 0.08) \cdot 10^{10} \text{ mol} \cdot \text{N}^{-1} \cdot \text{s}^{-1}$, $\omega_{12} = (8.1 \pm 3.5) \cdot 10^{13} \text{ mol} \cdot \text{N}^{-1} \cdot \text{s}^{-1}$ and $\omega_{21} = (1.63 \pm 0.60) \cdot 10^{12} \text{ mol} \cdot \text{N}^{-1} \cdot \text{s}^{-1}$. Suitable values of density (ρ) and kinematic viscosity (ν) of solutions are listed in Table 1. From this table results that both ρ and ν increase linearly with increase of ΔC_1 . Each experiment was performed for two gravitational configurations of the membrane system: first, with water in the vessel above the membrane and the solution below (configuration A); second, with these

positions reversed (configuration B). All experiments were performed at constant temperature $T = (295 \pm 0.1)$ K.

Evaluation of the global concentration changes in the solution was performed by standard isotope method. The measured value of the global concentration exchanges (dC/dt) was used to calculate the solute flux (J_s) on the basis of following equation

$$J_s^i = \frac{V_v}{S} \left(\frac{dC_s}{dt} \right)_{J_v^i=0}^i \quad (30)$$

where V_v is the volume of the measuring vessel and S is the membrane surface area (superscript $i=A, B$ pertain to configurations A and B). Measurements of J_s^i for both configurations were performed according to the following procedure. The first step involved the measurement of the solute flux in the membrane system by means of mechanical stirring of the solution at 500 rpm. After achieving the steady state during which J_s^o was constant, stirring was stopped, and subsequently the evolution of solute flux was measured up to the steady state, while the J_s^A was constant. The same procedure was followed for configuration B.

5. RESULTS

5.1. Time dependence of solute flux

A plots of the time dependence for J_1^i in configurations A and B of the membrane system in the case of a $0.05 \text{ mol}\cdot\text{l}^{-1}$ glucose in 0.2 aqueous ethanol solutions and *Nephrophane* membrane are presented in Figure 2. Line 1, common for both configurations, contains the results obtained under conditions of thorough mechanical stirring, and shows that J_1^o is independent of gravitational configuration of the single-membrane system. Curves 1A and 1B, obtained for configurations A and B respectively, demonstrate that the J_1^i values for both configurations differ ($J_1^A \neq J_1^B$). The evolution of the J_1^o to the J_1^A or J_1^B is a reflection of the process of formation of concentration boundary layers on both sides of the membrane.

5.2. Concentration dependencies of solute flux

The results of measurements of glucose flux (J_1^i) are shown in Figure 3. In this figure the dependencies of $J_1^i = f(\Delta C_1)$ for solution of glucose in 0.2 mol·l⁻¹ aqueous ethanol solution are presented. The curves 1A and 1B in Figure 3 were obtained when the solutions were mechanically unstirred, whereas curve 1 in this figure represented the dependence $J_1^o = f(\Delta C_1)$ obtained with the solutions being left mechanically stirred at 500 rpm. These curves show a non-linear dependencies of $J_1^A = f(\Delta C_1)$ and $J_1^B = f(\Delta C_1)$ in configurations A and B, respectively. In contrast, when the solutions were mechanically stirred, J_1^o was directly proportional to ΔC_1 and independent of system configuration. Similar results for ethanol flux were obtained.

The concentration polarization coefficient of single-membrane system can be defined by the following expressions

$$\zeta_s^i = \left(\frac{J_s^i}{J_s^o} \right)_{J_v^i=0, \Delta\pi_k=const.} \quad (31)$$

where J_s^o and J_s^i are the solute fluxes through a membrane and complex $l_l^i/M/l_h^i$, respectively ($s=1, 2$; $i=A, B$). The coefficients ζ_1^i and ζ_2^i were calculated on the basis of $J_1^o, J_1^i, J_2^o, J_2^i$ and are presented in Figure 4. Our investigations indicate that the values of the coefficients ζ_1^i and ζ_2^i for binary solutions are independent on the solute concentrations and dependent on the configuration of the single-membrane system. Then, the values of these coefficients for ternary solution are dependent on both the solute concentration and configuration of the single-membrane system.

5.3. Concentration dependencies of coefficients ζ_1^i and ζ_2^i for ternary solutions

In Figure 4 the dependencies of coefficients ζ_1^i and ζ_2^i ($i=A, B$) on concentration differences of glucose (ΔC_1) were presented with constant concentration difference of ethanol (ΔC_2) for configuration A (curves 1A and 2A) and configuration B (curves 1B and 2B). From curves 1B and 2B presented in this figure results, that values of coefficients ζ_1^B and ζ_2^B are initially constant and amounts $\zeta_1^B = \zeta_2^B = 0.031$ and next from $\Delta C_1 = 0.025$ mol·l⁻¹ increase linearly, to

value $\zeta_1^B=0.5$ and $\zeta_2^B=0.4$. For $\Delta C_1>0.05 \text{ mol}\cdot\text{l}^{-1}$, ζ_1^B and ζ_2^B are constant. From the curves 1A and 2A presented in this figure results, that values of coefficients ζ_1^A and ζ_2^A decreases non-monotonically from value 0.5 to 0.03 and from 0.39 to 0.028. Moreover, from Fig. 3 comes out that curves 1A and 1B criss-cross in the point: $\zeta_1^B=\zeta_1^A=0.203$ and $\Delta C_1=0.0338 \text{ mol}\cdot\text{l}^{-1}$ and curves 2A and 2B in the point: $\zeta_2^B=\zeta_2^A=0.117$ and $\Delta C_1=0.0314 \text{ mol}\cdot\text{l}^{-1}$. From Table 1 follows that the density of a $0.0338 \text{ mol}\cdot\text{l}^{-1}$ glucose in 0.2 aqueous ethanol solutions is almost the same as density of water.

5.4. Calculation results of the Rayleigh Number (R_1^i)

For ternary solution ($s=1, 2$) the equation (29) can be written in the form

$$R_1^i = \mathfrak{S}_1 \cdot \left[\left(\frac{\partial \rho}{\partial C_1} \right) (1 - \zeta_1^i)(C_{1h} - C_{1l}) + \left(\frac{\partial \rho}{\partial C_2} \right) (1 - \zeta_2^i)(C_{2h} - C_{2l}) \right] \left(\frac{1 - \zeta_1^i}{\zeta_1^i} \right)^3 \quad (32)$$

where $\mathfrak{S}_1 = \frac{g(D_{11})^2}{16\rho\nu(RT)^3(\omega_{11})^3}$.

Taking into consideration of above equation, the values of parameters ζ_1^A and ζ_1^B presented in Figure 4, values of ρ and ν listed in Table 1 and constant values of $g = 9.81 \text{ m}\cdot\text{s}^{-2}$, $D_{11}=0.69\times 10^{-9} \text{ m}^2\cdot\text{s}^{-1}$, $D_{22}=7.6\times 10^{-9} \text{ m}^2\cdot\text{s}^{-1}$, $R = 8.31 \text{ J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1}$, $T=295 \text{ K}$, $\omega_{11}=8\times 10^{-10} \text{ mol}\cdot\text{N}^{-1}\cdot\text{s}^{-1}$, $\omega_{22}=2\times 10^{-10} \text{ mol}\cdot\text{N}^{-1}\cdot\text{s}^{-1}$, $\rho_o=998 \text{ kg}\cdot\text{s}^{-3}$, $\nu_o=1.012\times 10^{-6} \text{ m}^2\cdot\text{s}^{-1}$, $\Delta C_2=0.2 \text{ mol}\cdot\text{l}^{-1}$, $\delta\rho/\delta C_1=0.06 \text{ kg}\cdot\text{mol}^{-1}$, $\delta\rho/\delta C_2=-0.0095 \text{ kg}\cdot\text{mol}^{-1}$, the concentration Rayleigh number R_1^i ($i = A, B$) for A and B configurations of single-membrane cell system were calculated by means of Mathcad 2000 professional. The selected results of calculations of Rayleigh Number (R_1^i) were presented in Figures 5 and 6.

In Figure 5 the dependencies of R_1^i ($i = A, B$) on the concentration difference of glucose (ΔC_1) were presented, with a constant concentration difference of ethanol ($\Delta C_2=0.2 \text{ mol}\cdot\text{l}^{-1}$) for configuration A (curve 1A) and configuration B (curve 1B). From curve A we note that R_1^A , for $\Delta C_1<0.02 \text{ mol}\cdot\text{l}^{-1}$ is independent of ΔC_1 . For $\Delta C_1>0.02 \text{ mol}\cdot\text{l}^{-1}$, R_1^i increases and for $\Delta C_1=0.0334 \text{ mol}\cdot\text{l}^{-1}$ it reaches a maximal value: $(R_1^A)_{\max}=277.1$. Moreover, for $\Delta C_1>0.0334 \text{ mol}\cdot\text{l}^{-1}$, R_1^A decreases non-monotonically and for $\Delta C_1=0.034 \text{ mol}\cdot\text{l}^{-1}$, $R_1^A = 0$. If $\Delta C_1>0.034 \text{ mol}\cdot\text{l}^{-1}$, then $R_1^A < 0$ (e.g. for $\Delta C_1=0.06 \text{ mol}\cdot\text{l}^{-1}$, $R_1^A=-1.8 \times 10^6$). From curve B it results that the

value of coefficient R_1^B increases non-monotonically from negative values to maximal positive value $(R_1^B)_{\max}=225.2$ for $\Delta C_1=0.0356 \text{ mol}\cdot\text{l}^{-1}$. For $\Delta C_1>0.0356 \text{ mol}\cdot\text{l}^{-1}$, R_1^B decreases non-monotonically to value, which is independent of ΔC_1 for $\Delta C_1>0.045 \text{ mol}\cdot\text{l}^{-1}$. The curves A and B criss-cross in the point: $R_1^A=R_1^B=135.3$ and $\Delta C_1=0.0338 \text{ mol}\cdot\text{l}^{-1}$. For $\Delta C_1<0.036 \text{ mol}\cdot\text{l}^{-1}$, $R_1^B<0$ (e.g. for $\Delta C_1=0.06 \text{ mol}\cdot\text{l}^{-1}$, $R_1^B=-1.9\times 10^6$) and if $\Delta C_1=0.036 \text{ mol}\cdot\text{l}^{-1}$, then $R_1^B=0$.

In the Figure 6 the dependencies R_1^i on $(1-\zeta_1^i)^3/(\zeta_1^i)^3$ are shown. The results illustrated by curves 1 and 2 were obtained for configurations A and B of single-membrane osmotic-diffusive cell, respectively. The curves 1A and 1B show that dependence $R_1^i = f[(1-\zeta_1^i)^3/(\zeta_1^i)^3]$ attain maximum $(R_1^A)_{\max.}=277.1$ for $(1-\zeta_1^A)^3/(\zeta_1^A)^3=65.2$ (in the case of curve 1A) and $(R_1^B)_{\max.}=225.2$ for $(1-\zeta_1^B)^3/(\zeta_1^B)^3=35.1$ (in the case of curve 1B). For $(1-\zeta_{11}^A)^3/(\zeta_{11}^A)^3=181.4$ and $(1-\zeta_1^B)^3/(\zeta_1^B)^3=106.2$, $R_1^A=R_1^B=0$. For $(1-\zeta_1^A)^3/(\zeta_1^A)^3>181.4$ and $(1-\zeta_1^B)^4/(\zeta_1^B)^3>106.2$, $R_1^A<0$ and $R_1^B<0$, respectively.

6. DISCUSSION

Form the experimental data presented in this paper results that in the case of mechanically stirred ternary non-electrolyte solutions solute flux is directly proportional to solution concentrations and does not depend on orientation of membrane and measurement chamber with solution, relative to vector of gravitational force. In the case of mechanically unstirred ternary non-electrolyte solutions solute flux is a non-linear function of one solute concentration in solvent when the other solute concentration is constant and strongly depends on the sequence of solutions relative to the horizontal membrane orientation and also on solutions' composition. Similar results for electrolyte solutions were obtained [29].

The results of studies on diffusive flows illustrated in Figure 3 and studies of osmotic flow presented in previous paper [11,12] indicates that for some ΔC_1 and ΔC_2 , both diffusive and osmotic flows of ternary solutions are not depended on configuration of single-membrane osmotic-diffusive cell. It means that for diffusive flows, the relation $J_1^A=J_1^B$ should be fulfilled. From Figure 3 results that the curves 1A and 1B criss-cross in a point which has co-ordinates $\Delta C_1\approx 0.0345 \text{ mol}\cdot\text{l}^{-1}$, $\Delta C_2=0.2 \text{ mol}\cdot\text{l}^{-1}$ and $J_1^A=J_1^B\approx 1.37\times 10^{-5} \text{ mol}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$. From Figure 4 results that curves 1A and 1B criss-cross in a point which has co-ordinates $\zeta_1^A=\zeta_1^B=0.203$

$(R_1^A=267.2, R_1^B=187.4)$ and $\Delta C_1=0.0344 \text{ mol}\cdot\text{l}^{-1}$. But for $(R_1^A)_{\max.}=277.1, \zeta_1^A=0.199$ and for $(R_1^B)_{\max.}=225.2, \zeta_1^B=0.234$. From Table 2 follows that the density of a $0.0345 \text{ mol}\cdot\text{l}^{-1}$ glucose in 0.2 aqueous ethanol solutions is almost the same as density of water. In the situation when $(R_1^A)_{\max.}=277.1$ and $\Delta C_1\approx 0.0334 \text{ mol}\cdot\text{l}^{-1}, J_1^A\approx 1.52\times 10^{-5} \text{ mol}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$. In this case, a point with these co-ordinates may determine the moment of transition from convectional to unconvective state and $(R_1^A)_{\max.}=277.1$ may have sense of critical value of concentration Rayleigh number in configuration A of single-membrane osmotic-diffusive cell. In the situation when $(R_1^B)_{\max.}=225.2$ and $\Delta C_1\approx 0.0356 \text{ mol}\cdot\text{l}^{-1}, J_1^B\approx 1.66\times 10^{-5} \text{ mol}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$. In this connection, a point with this co-ordinates may estimate the moment of transition from non-convectional to convectional state and $(R_1^B)_{\max.}=225.2$ may have sense of critical value of concentration Rayleigh number on configuration B of single-membrane osmotic-diffusive cell. But both $(R_1^A)_{\text{crit.}}=277.1$ and $(R_1^B)_{\text{crit.}}=225.2$ are far smaller than R_T for the thermally driven instability, which lies in the $1000\text{-}2000$ range. From paper [30] appears that for membraneous electrodiffusion in a range of concentration $0.01\div 0.1 \text{ mol}\cdot\text{l}^{-1}$, $(R_C^i)_{\text{crit.}}$ lies in a $11.6\div 116$ range. For convection induced by concentration gradients in a horizontal layer of a multicomponent fluid $(R_C)_{\text{crit.}}=19.35$ [31].

Moreover, on the basis of Figure 3 and 4 and Table 1 it can be estimated that for configuration A $(J_1^A)_{\text{crit.}}\approx (1.52\pm 0.08)\times 10^{-5} \text{ mol}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$, $(\Delta C_1^A)_{\text{crit.}}\approx (0.0334\pm 0.005) \text{ mol}\cdot\text{l}^{-1}$, $(\zeta_{11}^A)_{\text{crit.}}\approx (0.199\pm 0.06)$ and $(\Delta\rho_1^A)_{\text{crit.}}\approx (0.006\pm 0.001) \text{ kg}\cdot\text{m}^{-3}$. Then, for configuration B $(J_1^B)_{\text{crit.}}\approx (1.66\pm 0.08)\times 10^{-5} \text{ mol}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$, $(\Delta C_1^B)_{\text{crit.}}\approx (0.0356\pm 0.005) \text{ mol}\cdot\text{l}^{-1}$, $(\zeta_{11}^B)_{\text{crit.}}\approx (0.234\pm 0.06)$ and $(\Delta\rho)_{\text{crit.}}^B\approx (0.137\pm 0.001) \text{ kg}\cdot\text{m}^{-3}$. These observations can be examined in terms of gravitational stability or instability of CBLs. In configuration A natural convection appears when $R_1^A\leq (R_1^A)_{\text{crit.}}=277.1$. This condition we obtained for: $\Delta C_1\leq (\Delta C_1^A)_{\text{crit.}}=0.0334 \text{ mol}\cdot\text{l}^{-1}$, $\zeta_{11}^A\geq (\zeta_{11}^A)_{\text{crit.}}=0.199$, $J_1^A\leq (J_1^A)_{\text{crit.}}=1.52\times 10^{-5} \text{ mol}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ and $\Delta\rho\leq (\Delta\rho)_{\text{crit.}}=0.006 \text{ kg}\cdot\text{m}^{-3}$. Next, in configuration B natural convection appears for $R_1^B\leq (R_1^B)_{\text{crit.}}=225.2$. This condition we obtained for: $\Delta C_1^B\geq (\Delta C_1^B)_{\text{crit.}}=0.0356 \text{ mol}\cdot\text{l}^{-1}$, $\zeta_{11}^B\geq (\zeta_{11}^B)_{\text{crit.}}=0.234$, $J_1^B\leq (J_1^B)_{\text{crit.}}=1.66\times 10^{-5} \text{ mol}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ and $\Delta\rho\geq (\Delta\rho)_{\text{crit.}}^B=0.137 \text{ kg}\cdot\text{m}^{-3}$.

The results of studies, presented in our paper, may be important to understand the influence of gravitation force on the cell physiology. The gravitation force may influence on

biological cells in a direct and indirect way. Directly, this force influence on cell's organelles and macromolecules, which play a function as gravireceptors [32]. Whereas, indirectly – by provoking physico-chemical changes inside a cell, namely, hydrodynamic instabilities and pattern formation in a cytoplasmic medium in terrestrial conditions [33]. The change of conditions for membrane transport across biological membranes is a reason of organism reaction for microgravitation state. Lack of sedimentation and convection in microgravity reduce the gradient of substrates like nutrients and oxygen in a cell. Therefore, in microgravity on a border cell-solution may occur changes of potential and concentration favour creation boundary layers [34]. Then, in cells, where speed of using up oxygen and nutrients is greater than speed of diffusion, metabolism may be substantially distorted. The effects of spaceflight observed in various types of microorganisms and single plant and animal cells have been reviewed extensively by Cogoli and Gmünder [35]. No satisfactory explanation for these effects has been offered so far. Based on theoretical considerations, Mesland [36] suggest that mechanism of direct action of gravity could be: focusing via cell networks, amplification via non-linear dynamic system and suppression of bifurcation in non-linear dynamic systems. In particular, the biological state transitions may have windows of gravity dependence. In these windows, a minimal time of uni-directional gravity would be required for an effect.

7. CONCLUSIONS

1. The experimental data presented in this paper indicate that gravitational force has essential influence on solute flow non-electrolyte solutions through horizontally mounted membrane in a single-membrane osmotic-diffusive cell. Solution/membrane/water system was examined both with solutions less dense, and with solutions more dense, than water. J_s^i was substantially larger with the denser liquid (solution or water) above the membrane. This observation was explained in terms of the gravitational stability or instability of CBLs at the membrane-liquid interfaces. Through an extension of Rayleigh-Taylor stability analysis, density gradients were linked to a concentration Rayleigh number (R_s^i).
2. The presented method of evaluation the concentration Rayleigh number (R_s^i) for the isothermal membrane transport, requires determination of diffusive permeability (ω_s) and

concentration polarization (ζ_s^i) coefficients, diffusion coefficient for solute in solution (D_{ss}), density (ρ) and kinematic viscosity (ν) for solutions. The data of the concentration Rayleigh number make it possible to observe the kinetic of the solute transport through the membrane and the concentration boundary layers. It is known, that this number defines transition conditions from pure to convective diffusion.

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FIGURE CAPTIONS

Fig. 1. Graphic illustration of the configurations A and B of single-membrane cell system with concentration profile. M, membrane; δ_m – thickness of membrane; l_l^A , l_h^A , l_l^B and l_h^B – concentration boundary layers in configurations A and B; δ_l^A , δ_h^A , δ_l^B and δ_h^B – thicknesses of CBLs; C_{sl} and C_{sh} – global solution concentrations; C_{sl}^A , C_{sh}^A , C_{sl}^B and C_{sh}^B – local solution concentration in configurations A and B; J_{vs}^A , J_{vs}^B , J_s^A and J_s^B – volume and solute fluxes across the complex $l_l^i/M/l_h^i$ (i=A, B) in configurations A and B, respectively; J_{sm}^A , J_{sl}^A , J_{sh}^A , J_{sm}^B , J_{sl}^B and J_{sh}^B – solute fluxes across the membrane (M) and layers l_l^A , l_h^A , l_l^B and l_h^B in configurations A and B, respectively; ω_{ks} , Ω_{ks}^A and Ω_{ks}^B – diffusive permeability coefficients of membrane and complex: boundary layer/membrane/ boundary layer; $(\omega_{ks}^A)_l$, $(\omega_{ks}^A)_h$, $(\omega_{ks}^B)_l$ and $(\omega_{ks}^B)_h$ – diffusive permeability coefficients of layers l_l^A , l_h^A , l_l^B and l_h^B in configurations A and B, respectively.

Rys. 1. Graficzna ilustracja konfiguracji A i B komórki jedno-membranowej z profilem stężeniowym. M – membrana; δ_m – grubość membrany; l_l^A , l_h^A , l_l^B oraz l_h^B – stężeniowe warstwy graniczne w konfiguracji A i B; δ_l^A , δ_h^A , δ_l^B i δ_h^B – grubości CBL; C_{sl} oraz C_{sh} – globalne stężenia roztworów; C_{sl}^A , C_{sh}^A , C_{sl}^B oraz C_{sh}^B – lokalne stężenia roztworów odpowiednio w konfiguracjach A i B; J_{vs}^A , J_{vs}^B , J_s^A oraz J_s^B – strumień objętościowy i substancji rozpuszczonych przez kompleks $l_l^i/M/l_h^i$ (i=A, B) odpowiednio w konfiguracjach A oraz B; J_{sm}^A , J_{sl}^A , J_{sh}^A , J_{sm}^B , J_{sl}^B oraz J_{sh}^B – strumień substancji rozpuszczonych przez membranę (M) i warstwy l_l^A , l_h^A , l_l^B oraz l_h^B odpowiednio w konfiguracjach A oraz B; ω_{ks} , Ω_{ks}^A and Ω_{ks}^B – współczynniki przepuszczalności dyfuzyjnej membrany i kompleksu warstwa graniczna/membrana/warstwa graniczna; $(\omega_{ks}^A)_l$, $(\omega_{ks}^A)_h$, $(\omega_{ks}^B)_l$ i $(\omega_{ks}^B)_h$ – współczynniki przepuszczalności dyfuzyjnej warstw l_l^A , l_h^A , l_l^B and l_h^B odpowiednio w konfiguracjach A i konfiguracjach.

Fig. 2. The dependencies of $J_1^i = f(t)$ for glucose solution with concentration $0.05 \text{ mol}\cdot\text{l}^{-1}$ in $0.2 \text{ mol}\cdot\text{l}^{-1}$ aqueous ethanol solution and for the configurations A (curve 1A) and B (curve 1B). The line 1 was received for the system with mechanical mixing of solutions; the curves 1A and 1B – for the membrane system without mechanical mixing.

Rys. 2. Zależności $J_1^i = f(t)$ dla roztworu glukozy o stężeniu $0,05 \text{ mol}\cdot\text{l}^{-1}$ w $0.2 \text{ mol}\cdot\text{l}^{-1}$ wodnym roztworze etanolu dla konfiguracji A (krzywa A) i B (krzywa B). Prostą 1 otrzymano dla układu z mechanicznym mieszaniem roztworów; krzywe 1A i 1B – dla układu membranowego bez mieszania mechanicznego.

Fig. 3. The dependencies $J_1^i = f(\Delta C_1)$ for glucose solutions in $0.2 \text{ mol}\cdot\text{l}^{-1}$ aqueous ethanol solution and for the configurations A (curve 1A) and B (curve 1B). The line 1 was received for the membrane system with mechanical mixing.

Rys. 3. Zależności $J_1^i = f(\Delta C_1)$ dla roztworów glukozy w $0,2 \text{ mol}\cdot\text{l}^{-1}$ wodnym roztworze etanolu dla konfiguracji A (krzywa 1A) i B (krzywa 1B). Linie 1 otrzymano dla układu membranowego z mieszaniem mechanicznym roztworów.

Fig. 4. Glucose concentration dependencies of ζ_1^i (curve 1A and 1B) and ζ_2^i (curve 2A and 2B) ($i = A, B$) in $0.2 \text{ mol}\cdot\text{l}^{-1}$ aqueous ethanol solution in configuration A (curves 1A and 2A) and in configuration B (curves 1B and 2B).

Rys. 4. Zależności ζ_1^i od stężenia glukozy (krzywe 1A i 1B) oraz ζ_2^i (krzywe 2A i 2B) ($i = A, B$) w $0,2 \text{ mol}\cdot\text{l}^{-1}$ wodnym roztworze etanolu dla konfiguracji A (krzywe 1A i 2A) i dla konfiguracji B (krzywe 1B i 2B).

Fig. 5. The dependencies $R_1^i = f(\Delta C_1)$ of a glucose solution in $0.2 \text{ mol}\cdot\text{l}^{-1}$ aqueous ethanol solution for configurations A (curve 1A) and B (curve 1B) calculated on the grounds of equation (32).

Rys. 5. Zależności $R_1^i = f(\Delta C_1)$ od stężenia glukozy w $0,2 \text{ mol}\cdot\text{l}^{-1}$ wodnym roztworze etanolu dla konfiguracji A (krzywa 1A) i B (krzywa 1B) obliczone na podstawie równania (32).

Fig. 6. The dependencies $R_c^i = f[(1 - \zeta_1^i)^3 / (\zeta_1^i)^3]$ of a glucose solution in $0.2 \text{ mol}\cdot\text{l}^{-1}$ aqueous ethanol solution for configurations A (curve 1A) and B (curve 1B) calculated on the grounds of equation (27).

Rys. 6. Zależności $R_c^i = f[(1 - \zeta_1^i)^3 / (\zeta_1^i)^3]$ dla roztworu glukozy w $0,2 \text{ mol}\cdot\text{l}^{-1}$ wodnym roztworze etanolu dla konfiguracji A (krzywa 1A) i B (krzywa 1B) obliczona na podstawie równania (27).

Table 1. The values of density (ρ), kinematic viscosity (ν), for different values of glucose concentration difference (ΔC_1) in 0.2 mol·l⁻¹ aqueous ethanol solution (ΔC_2)

Tabela 1. Wartości gęstości (ρ), lepkości kinematycznej (ν), dla różnych wartości stężenia glukozy (ΔC_1) w 0.2 mol·l⁻¹ roztworze wodnego etanolu (ΔC_2)

ΔC_1 (mol·l ⁻¹)	ρ (kg/m ³)	ν (10 ⁶ m ² /s)
	$\Delta C_2=0.2$ (mol·l ⁻¹)	
0.000	996.0	1.049
0.005	996.3	1.051
0.010	996.6	1.053
0.015	996.9	1.055
0.020	997.2	1.057
0.025	997.5	1.060
0.030	997.8	1.062
0.035	998.1	1.064
0.040	998.4	1.066
0.045	998.7	1.068
0.050	999.0	1.071
0.055	999.3	1.073
0.060	999.6	1.075