

On the Derivation of the Kargol's Mechanistic Transport Equations from the Kedem–Katchalsky Phenomenological Equations

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Abstract. In the present article, it was demonstrated that – by starting from the so-called adjusted Kedem–Katchalsky (KK) phenomenological equations (Suchanek et al. 2004), i.e. the equations:

$$\begin{aligned}J_v &= L_p \Delta P - L_{pD} \Delta \Pi \\J_D &= -L_{Dp} \Delta P + L_D \Delta \Pi\end{aligned}$$

it is possible to derive practical transport equations (for the volume flow and the solute flow) in the form of the Kargol's mechanistic transport equations (Kargol and Kargol 2000, 2001, 2003a,b,c, 2004; Kargol 2002).

On this basis, it has been found that the KK thermodynamic formalism for membrane transport (practical equations) is in general identical with the mechanistic equations for membrane transport.

Key words: Membranes — Substance transport — KK equations — Mechanistic equations

Introduction

In order to describe substance transport across membranes, since the late 1950's and early 1960's, Kedem–Katchalsky (KK) practical thermodynamic equations (Kedem and Katchalsky 1958; Katchalsky and Curran 1965) have been widely used. These equations have the following forms:

$$J_v = L_p \Delta P - L_p \sigma \Delta \Pi \quad (1a)$$

$$j_s = (1 - \sigma) \bar{c}_s J_v + \omega \Delta \Pi \quad (1b)$$

in which J_v , j_s are flows; and L_p , σ , ω are coefficients (of filtration, reflection and diffusive permeability of the solute); ΔP and $\Delta \Pi$ are mechanical and osmotic pressure differences; and \bar{c}_s is the mean concentration of the solution.

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The assumption is that they apply to homogeneous membranes in terms of transport properties because in the KK formalism one does not go deep into the membrane microscopic structure. Yet, as far as research is concerned, we deal with porous membranes most frequently. These membranes have specific pores (channels) which are permeable to the solvent (water) and certain solutes.

In recent years, several works (Kargol and Kargol 2000, 2001, 2003a,b,c, 2004; Kargol 2002) have been published, in which these membranes have been divided into homogeneous and heterogeneous. It has been assumed that a membrane is homogeneous if its pores do not vary in terms of their cross-section radiuses. Membranes with varying pore cross-section radiuses are to be treated as heterogeneous in terms of transport properties.

It is not difficult to see that the above KK equations apply to homogeneous porous membranes. In order to demonstrate this, let us assume that the condition $\Delta P = |-\sigma\Delta\Pi|$ is satisfied. Then $J_v = 0$. In this situation, there are also no volume flows in individual pores of the membrane. This means that the term $\omega\Delta\Pi$ of Eq. (1b) formulates the diffusive solute flow. If $\Delta P \neq |-\sigma\Delta\Pi|$, then – apart from the diffusive flow $\omega\Delta\Pi$ – there occurs also the convective flow $(1 - \sigma)\bar{c}_s J_v$. The matter becomes clearly more complicated when Eq. (1b) is applied to heterogeneous porous membranes. It is here that we arrive at a state in which, at $\Delta P = |-\sigma\Delta\Pi|$, the volume flow $J_v = 0$. Yet now, in the individual pores of the membrane, there will exist volume flows varying in terms of their value and direction. This means that the term $\omega\Delta\Pi$ is now not the term to formulate the diffusive solute flow only. Moreover, at $\Delta P \neq |-\sigma\Delta\Pi|$, also the term $(1 - \sigma)\bar{c}_s J_v$ does not formulate the solute convective transport only. A difficult situation has arisen, in which it is possible to assume that the KK equation for the flow j_s does not apply to heterogeneous porous membranes, and it was to these membranes that it was mostly applied.

Under the circumstances, Kargol and Kargol (2000, 2001, 2003a,b,c, 2004) have recently derived, on the basis of mechanistic considerations, some new equations for membrane transport. These have been called mechanistic equations, and treated by their authors as alternative to the KK transport equations.

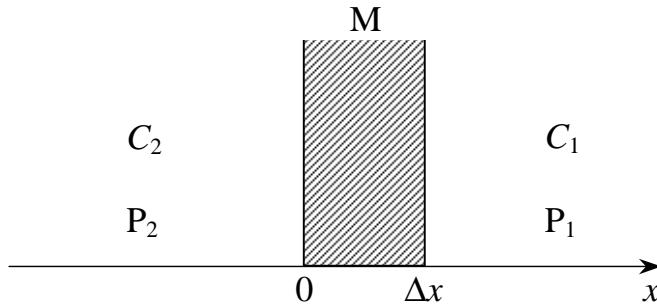


Figure 1. Membrane system. M, membrane; C_1, C_2 , concentrations; P_1, P_2 , mechanical pressures; Δx , membrane thickness; $C_2 > C_1$, $P_2 > P_1$.

The alternative nature of mechanistic equations resulted, primarily, from the fact that these equations apply both to porous homogeneous and heterogeneous membranes.

In the present article, we first present the idea of the derivation of the so-called adjusted phenomenological KK equations and outline mechanistic model for membrane transport. Next, we demonstrate that – when starting from these adjusted phenomenological equations (Suchanek et al. 2004) – it is possible to arrive at transport equations for the volume flow and the solute flow in the form of the Kargol's mechanistic transport equations. On this basis, in the present article, we shall also refer to selected aspects of mutual equivalence of the KK formalism and the Kargol's mechanistic description.

On the KK derivation procedure for phenomenological transport equations

The starting point in the KK derivation procedure for phenomenological equations is the membrane system shown in Fig. 1 and the dissipation function given by the formula (Kedem and Katchalsky 1958; Katchalsky and Curran 1965):

$$\Phi = \sum_{i=1}^n \bar{J}_i \cdot \text{grad}(-\mu_i)$$

in which \bar{J}_i is the volume flow, and μ_i is the chemical potential.

Under stationary conditions, at every point of the membrane, we have $\bar{J}_i = \text{const.}$

In order to determine the dissipation function $\Phi_{\Delta x}$ for the membrane of thickness Δx , the above expression must be integrated. Then we obtain:

$$\Phi_{\Delta x} = \int_0^{\Delta x} \Phi dx = \Phi = \int_0^{\Delta x} \sum_{i=1}^n \bar{J}_i \cdot \text{grad}(-\mu_i) dx = \sum_{i=1}^n J_i \Delta \mu_i.$$

At this juncture, Kedem and Katchalsky (1958) decided to abandon vector notation because all gradients and flows in the system are directed along the x axis.

Consequently, they obtained the following phenomenological equations:

$$J_v = L_p \Delta P + L_{pD} \Delta \Pi \quad (2a)$$

$$J_D = L_{Dp} \Delta P + L_D \Delta \Pi \quad (2b)$$

where L_p, L_{pD}, L_{Dp}, L_D are coefficients; ΔP and $\Delta \Pi$ are mechanical and osmotic pressure differences.

On the basis of these equations, they arrived at the following practical transport equations (Eqs. (1a) and (1b)), i.e.:

$$\begin{aligned} J_v &= L_p \Delta P - L_p \sigma \Delta \Pi \\ j_s &= \omega \Delta \Pi + (1 - \sigma) \bar{c}_s J_v \end{aligned}$$

In order for the equation for the flow J_v to have the above-presented notation, i.e. a notation adequate to the system as presented in Fig. 1, they had no alternative but to adopt the definition of the reflection coefficient as proposed by Staverman:

$$\sigma = -\frac{L_p D}{L_p} \quad (3)$$

in which L_{pD} is a negative value ($L_{pD} < 0$).

However, if vector notation is not abandoned for the flows and forces, then the following adjusted phenomenological equations (Suchanek et al. 2004) are obtained:

$$J_v = L_p \Delta P - L_{pD} \Delta \Pi \quad (4a)$$

$$J_D = -L_{Dp} \Delta P + L_D \Delta \Pi \quad (4b)$$

in which $L_{pD} = L_{Dp} \geq 0$.

Now, in order to arrive at Eqs. (1a) and (1b) as equations written in accordance with the system presented in Fig. 1, it suffices to adopt the following adjusted definition of the reflection coefficient (Suchanek et al. 2004):

$$\sigma = \frac{L_{pD}}{L_p} \quad (5)$$

in which the coefficient L_{pD} is now greater than zero.

In the following part of this article, it was demonstrated that on the basis of these adjusted phenomenological equations it is also possible to arrive at the Kargol's mechanistic equations for membrane transport.

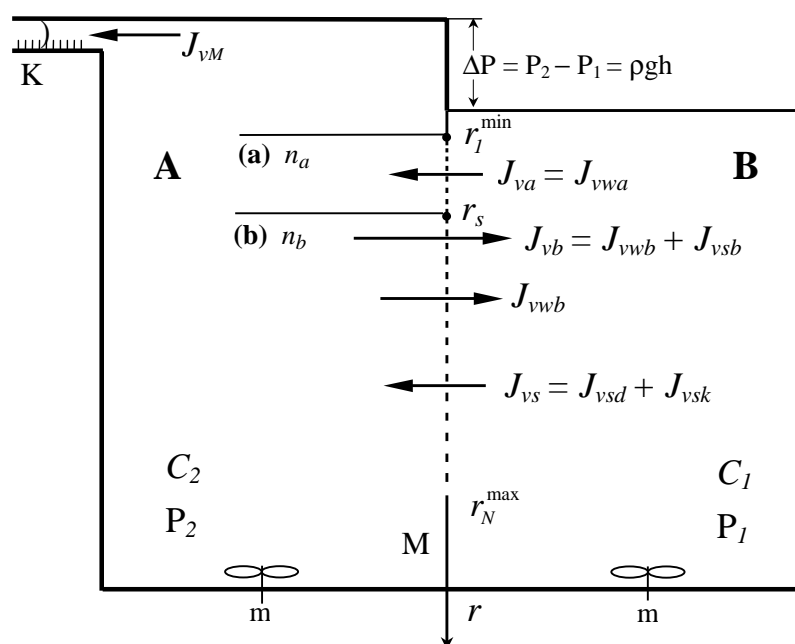
Mechanistic model for membrane transport and its description

In order to present the foundation of the Kargol's mechanistic model for membrane transport (Kargol and Kargol 2000, 2001, 2003a,b,c, 2004; Kargol 2002), we shall begin our considerations from the membrane system, presented schematically in Fig. 2.

In this system, the heterogeneous porous membrane M (whose pores vary in their cross-section radiuses (r) separates two compartments (A and B) which contain non-electrolytic solutions of the same solute of concentrations C_1 and C_2 ($C_1 < C_2$). These solutions are under the hydrostatic pressures P_1 and P_2 which satisfy the relation: $P_1 < P_2$.

Let us assume, as a model, that the heterogeneous porous membrane has a statistical number N of cylindrical pores permeable to the solvent (water) and perpendicular to the surface.

To facilitate our considerations, let us assume that the pores in the membrane M have been vertically arranged in a line which starts (at the top) from the pores with the smallest radiuses r_1^{\min} , and ends with the largest pores r_N^{\max} (at the bottom). It must be stressed here that this arrangement serves only to illustrate



the problem and is not reflected in the model membrane structure. In a typical membrane of this type, individual pores are randomly distributed.

$$r_1^{\min} = r_w < r_2 \dots < r_s < \dots < r_N^{\max}$$

In this situation, the membrane M may be divided into Part (a) containing a certain number n_a of small pores, contained within the interval from $r_1 = r_w$ to r_s , and Part (b) containing $n_b = N - n_a$ of large pores, which are found in the interval from r_s to r_N^{\max} . It is easy to see that the pores n_a will constitute a total barrier for the molecules of the solute s . These will then be semi-permeable pores. Therefore, they may be assigned the reflection coefficient amounting to $\sigma_a = 1$.

The pores n_b , in turn, which are permeable to both water and the solute s are to be assigned the reflection coefficient $\sigma_b = 0$. Following this line of reasoning, Kargol and Kargol (2000, 2001, 2003a,b,c, 2004) and Kargol (2002) have put forward a

proposal concerning the local formulation of the reflection coefficient. It postulates that an individual membrane pore may adopt values of the reflection coefficient σ_p amounting to either $\sigma_p = 1$ or $\sigma_p = 0$. The values of this parameter may not be contained in the interval of $0 < \sigma_p < 1$.

In the light of the above considerations, the membrane M (as a set of N pores permeable to water) will have the reflection coefficient $\sigma = 1$ when all pores are impermeable to the solute s . This will then be a semi-permeable membrane. On the other hand, when all the pores N are permeable to the solute s , its reflection coefficient will amount to $\sigma = 0$. This membrane ought to be treated as permeable (non-selective).

If a certain number n_a of the membrane pores are semi-permeable and the remaining pores $n_b = N - n_a$ are permeable to the solute s , the membrane will be selective. Its reflection coefficient will be contained in the interval of $0 < \sigma < 1$. This type of selective membrane has been fitted into the system presented in Fig. 2.

In view of the above, it will be seen clearly that (given the existence, on the membrane M, of the osmotic-pressure difference $\Delta\Pi = RT(C_2 - C_1)$, where R is the gas constant, T is the temperature, and of mechanical-pressure difference $\Delta P = P_2 - P_1$) the volume flow will be generated within the pores n_a :

$$J_{va} = J_{vwa} = L_{pa}\Delta P - L_{pa}\Delta\Pi \quad (6)$$

where $J_{va} = J_{vwa}$ is in fact the osmotic solvent flow (if $|\Delta P| < |\Delta\Pi|$), and L_{pa} represents the filtration coefficient of the pores n_a (i.e. Part (a) of the membrane).

In the pores n_b of the membrane (i.e. within its Part (b) with the filtration coefficient L_{pb}), the volume flow J_{vb} given by the following formula will permeate:

$$J_{vb} = L_{pb}\Delta P \quad (7)$$

since $\sigma_b = 0$.

The above statements have provided the basis for the derivation of the following transport equations by Kargol and Kargol (2000, 2001, 2003a,b,c, 2004) and Kargol (2002):

$$J_v = L_p\Delta P - L_p\sigma\Delta\Pi \quad (8)$$

and

$$j_s = \omega_d\Delta\Pi + (1 - \sigma)\bar{c}_s L_p\Delta P \quad (9)$$

These equations (in a manner analogous to the KK equations) formulate the volume flow J_s and the solute flow j_s as a function of the pressure differences ΔP and $\Delta\Pi$.

The parameters L_p (filtration coefficient), σ (reflection coefficient) and ω_d (which is the coefficient of diffusive solute permeability) occurring in these equations are thus formulated:

$$L_p = L_{pa} + L_{pb} \quad (10)$$

$$\sigma = \frac{L_{pa}}{L_p} \quad (11)$$

and

$$\omega_d = (1 - \sigma)L_p\bar{c}_s \quad (12)$$

where L_{pa} and L_{pb} are filtration coefficients of Part (a) and Part (b) of the membrane M, and \bar{c}_s is mean concentration.

The derivation procedure for mechanistic transport equations on the basis of the adjusted KK phenomenological equations

Considering the derivation of the membrane transport equations in the form of mechanistic equations (i.e. Eqs. (8) and (9)) on the basis of the adjusted KK phenomenological equations (i.e. Eqs. (4a) and (4b)), let us consider the membrane system presented in Fig. 2.

As has been stated above, for the membrane M of this system, it is possible to find such a solute s with the molecule radius r_s which will satisfy the relation: $r_1 < r_s < r_N$. In view of that, this membrane may be divided into Part (a) and Part (b). Part (a), which contains n_a pores of radiuses $r \leq r_s$ (cf. Fig. 2), may necessarily be permeated by the solvent only. On the other hand, across Part (b) of the membrane which contains n_b pores of radiuses $r > r_s$, both the solvent and the solute s permeate.

The above membrane may in fact be treated formally as two joined parallel membranes (a) and (b) with varying filtration coefficients L_{pa} and L_{pb} (respectively).

Because only the solvent permeates across Part (a) of the membrane, it is a semi-permeable membrane. It is obvious that for this membrane $\Delta P = \Delta \Pi$, if $J_{va} = 0$. In view of the above, in accordance with Eq. (4a), the following equality must be satisfied: $L_{pa} = L_{pDa}$, where L_{pDa} is the osmotic permeability coefficient for Part (a) of the membrane.

However, because the substance does not diffuse across the pores $r < r_s$, the diffusive flow of the substance in Part (a) of the membrane is equal to zero.

Eqs. (4a) and (4b) for Part (a) of the membrane are then reduced to the following:

$$J_{va} = L_{pa}(\Delta P - \Delta \Pi) \quad (13a)$$

and

$$J_{Da} = 0 \quad (13b)$$

On the other hand, Part (b) is permeable both to the solute and the solvent. In this case, cross coefficients (related to membrane selectivity with respect to the solvent and the solute) satisfy the relation: $L_{pDb} = L_{Dpb} = 0$ (which shall be demonstrated below).

In view of this, for Part (b) of the membrane, in accordance with Eqs. (4a) and (4b), we obtain:

$$J_{vb} = L_{pb}\Delta P \quad (14a)$$

$$J_{vD} = L_{Db}\Delta \Pi \quad (14b)$$

Hence, due to the equality:

$$J_v = J_{va} + J_{vb} \quad (15)$$

because of the equations of expressions (13a) and (14a), we obtain:

$$J_v = (L_{pa} + L_{pb})\Delta P - L_{pa}\Delta\Pi \quad (16)$$

With the introduction of the following:

$$L_p = L_{pa} + L_{pb} \quad (17a)$$

$$\sigma = \frac{L_{pa}}{L_p} \quad (17b)$$

Eq. (16) may be written as

$$J_v = L_p\Delta P - L_p\sigma\Delta\Pi \quad (18)$$

This equation has the same form as Eq. (7a). Moreover, the expression (17b) defines the reflection coefficient σ in a manner identical to that described in articles (Kargol and Kargol 2000, 2001, 2003a,b,c, 2004; Kargol 2002).

The mechanistic equation for the pure solute flow j_s

The total diffusive flow J_D of the solute s across the membrane M (Fig. 2) may be formulated in the following obvious expression:

$$J_D = J_{Da} + J_{Db} \quad (19)$$

in which J_{Da} and J_{Db} are diffusive flows which permeate across Part (a) and Part (b) of the membrane.

Yet, in view of formulas (13b) and (14b), the above equation will take the following form:

$$J_D = L_{Db}\Delta\Pi \quad (20)$$

If starting from the adjusted phenomenological equations KK (4a) and (4b), this flow may be formulated in the relation (Kedem and Katchalsky 1958; Katchalsky and Curran 1965):

$$J_v + J_D \approx J_w\bar{V}_w + J_s\bar{V}_s + \frac{j_s}{\bar{c}_s} - J_w\bar{V}_w \approx \frac{j_s}{\bar{c}_s} \quad (21)$$

In the case of the heterogeneous porous membrane, which is the subject of our considerations here, the flow J_D is identical with the flow J_{Db} , which in turn is related to the volume flow J_{vb} , which permeates across Part (b).

Consequently, the expression (21) is to be rewritten as

$$J_{vb} + J_{Db} \approx J_{wb}\bar{V}_w + J_{sb}\bar{V}_w + \frac{j_{sb}}{\bar{c}_s} - J_{wb}\bar{V}_w \approx \frac{j_{sb}}{\bar{c}_s} = \frac{j_s}{\bar{c}_s} \quad (22)$$

Next, by putting into the formula (22) the expressions (14a) and (14b), we obtain:

$$\frac{j_s}{\bar{c}_s} = L_{pb}\Delta P + L_{Db}\Delta\Pi = L_p \left(1 - \frac{L_{pa}}{L_p}\right) \Delta P + L_{Db}\Delta\Pi = L_p(1 - \sigma)\Delta P + L_{Db}\Delta\Pi \quad (23)$$

So, the flow of the substance which permeates across the pores in the Part (b) of the membrane (equal, in this case, to the total flow j_s across the membrane) is given by the expression:

$$j_s = L_p(1 - \sigma)\bar{c}_s\Delta P + \bar{c}_s L_{Db}\Delta\Pi \quad (24)$$

The above equation is identical with the mechanistic equation as derived by Kargol and Kargol (2000, 2001, 2003a,b,c, 2004) and Kargol (2002), which becomes obvious when the following notation is introduced:

$$L_D = L_{Da} + L_{Db} = L_{Db} \quad (\text{since } L_{Da} = 0) \quad (25)$$

and

$$\omega_d = \bar{c}_s L_D = \bar{c}_s L_{Db} \quad (26)$$

where (this is but a reminder) ω_d is the coefficient of diffusive solute permeability.

Consequently, we obtain the following equation:

$$j_s = L_p(1 - \sigma)\bar{c}_s\Delta P + \omega_d\Delta\Pi \quad (27)$$

which is an equation identical in form with Eq. (9).

Eqs. (18) and (27) have a form which is equivalent to Eqs. (4a) and (4b), if the following conditions are satisfied:

1. The total filtration coefficient for the membrane is equal to the sum of filtration coefficients for Part (a) and Part (b) of the membrane, namely $L_p = L_{pa} + L_{pb}$.

The reflection coefficient for the membrane, in accordance with the adjusted Staverman's definition (5) equal to $\sigma = \frac{L_{pD}}{L_p}$ (where L_{pD} is the osmotic permeability

coefficient), is identical to the mechanistic coefficient $\sigma = \frac{L_{pa}}{L_p}$. It follows therefrom that for heterogeneous membranes (in the sense of the Kargol's definitions, see Kargol and Kargol 2000, 2001, 2003a,b,c, 2004; Kargol 2002), the coefficient L_{pD} may be interpreted as the filtration coefficient of Part (a) of the membrane, that is the semi-permeable part, for which $\sigma_a = 1$.

2. In the case at issue, the coefficient ω_d ought to be treated as a coefficient related to substance diffusion across the pores of Part (b) of the membrane, that is the pores which are permeable to the solute s .

Even in the light of the above considerations, it may in general be stated that the KK transport formalism is equivalent to the Kargol's mechanistic formalism for membrane transport.

On the interpretation of permeability coefficient ω

The Kargol's mechanistic equations (18) and (27), derived above (on the basis of the adjusted KK phenomenological equations) are transparent in terms of interpretation with regard to the membrane system shown in Fig. 2 (especially concerning the diffusion coefficient ω_d). Yet, as the subject of the present article is in fact a survey of the analogies between mechanistic equations and the KK equations, we shall try to formally transform the mechanistic equations into the conventional form of the practical KK equations. Our objective here is to express the permeability coefficient ω of the equation for the flow j_s , through mechanistic parameters.

With this in mind, let us transform Eq. (16) to obtain the following:

$$\Delta P = \frac{1}{L_p} J_v + \frac{L_{pa}}{L_p} \Delta \Pi$$

If the above expression is put into the formula (23), we obtain:

$$\begin{aligned} \frac{j_s}{\bar{c}_s} &= L_{pb} \Delta P + L_{Db} \Delta \Pi = \frac{L_{pb}}{L_p} J_v + \frac{L_{pb} L_{pa}}{L_p} \Delta \Pi + \frac{L_{Db}}{L_p} L_p \Delta \Pi = \\ &= \frac{L_{pb}}{L_p} J_v + \frac{L_p L_D + L_{pa} L_{pb}}{L_p} \Delta \Pi \end{aligned}$$

Then, having applied the definition of the reflection coefficient as given in the relation (17b), we can write the above formula in the following form:

$$j_s = (1 - \sigma) \bar{c}_s J_v + \frac{L_p L_{Db} + L_{pb} L_{pa}}{L_p} \bar{c}_s \Delta \Pi$$

or as

$$j_s = (1 - \sigma) \bar{c}_s J_v + \omega \Delta \Pi \quad (28)$$

where

$$\omega = \frac{L_p L_{Db} + L_{pa} L_{pb}}{L_p} \bar{c}_s = \bar{c}_s L_D + \frac{L_{pa} L_{pb}}{L_p} \bar{c}_s \quad (29)$$

Eq. (28) has a form which is exactly equivalent to the practical KK equation, i.e. Eq. (1b), which defines permeability coefficient ω (by means of the measurement of the solute flow j_s , at $J_v = 0$).

From the above considerations, it follows that, with regard to the heterogeneous porous membranes, the coefficient ω is not a coefficient which formulates the diffusive solute transport only. This conclusion follows from the analysis of the expression (29). The term $(1 - \sigma) \bar{c}_s J_v$ of this equation cannot be a convective term only. In the light of the above considerations, it may be claimed that the applicability of the conventional KK equation for the flow j_s with regard to heterogeneous porous membranes may raise some doubt.

Conclusions

It has been demonstrated that the Kargol's mechanistic transport equations for the flows J_v and j_s , may be derived from the adjusted phenomenological equations. On the basis of the present article, it may generally be confirmed that the KK thermodynamic formalism is equivalent to the mechanistic equations. These (KK and mechanistic) equations yield the same research results pertaining to the flows J_v and j_s . It is not difficult to note that their parameters L_p and σ are also identical.

In the present article, the mechanistic equation for the solute flow j_s in a form analogous to the second practical KK equation has been derived. Simultaneously, we have confirmed the doubts of the authors of the mechanistic model concerning the issue of the physical interpretation of the coefficient ω (defined by Eq. (28)), if it is applied to porous heterogeneous membranes. This KK equation appears to be ambiguous in such a situation, which results from the fact that the term $\omega\Delta\Pi$ of this equation does not formulate only the diffusive substance transport, and the term $(1 - \sigma)\bar{c}_s J_v$ – convective transport only. In this context, we have demonstrated that the coefficient ω , may be expressed through the phenomenological coefficient of diffusion L_D and the filtration coefficients of both parts of the porous membrane, i.e. its permeable and semi-permeable parts. So, with regard to heterogeneous porous membranes, the equation for the flow j_s in the form (27) or (8) (used by Kargol's) is rather clear in terms of interpretation. The term $\omega_d\Delta\Pi$ of this equation formulates only the diffusive solute transport, and the term $(1 - \sigma)L_p\bar{c}_s\Delta P$ – only the convective transport.

The present article comes closer to achieving a complete elucidation of the interrelations between the KK equations and the mechanistic equations. The need to elucidate these relations has been justified by the fact that, during the investigations conducted on the basis of the KK equations, an enormous body of experimental material has been collected, pertaining especially to biological membranes, cell membranes included. Thanks to the consistency of mechanistic transport equations, and KK equations, these research results may be interpreted and considered more thoroughly.

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