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Kinetic Model for Solute Diffusion in Liquid Membrane Systems

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Abstract:

In this study, a mathematical model for the kinetics of solute transport in liquid membrane systems (LMSs) has been formulated. This model merged the mechanisms of consecutive and reversible processes with a "semi-derived" diffusion expression, resulting in equations that describe solute concentrations in the three sections (donor, acceptor and membrane). These equations have been refined into linear forms, which are satisfying in the special conditions for simplification obtaining the important kinetic constants of the process experimentally.

Key words: kinetic model, diffusion, liquid membranes, mathematical model, solute diffusion in a LMS.

Introduction:

A liquid membrane system LMSs can be defined as a system containing a definite liquid split by another immiscible liquid; in general, it is a phase split by another phase. Despite of the simplicity of this definition, it does not reduce the wide spread cases that are represented by it and their importance. The cell membranes of living organisms can be considered as one of these systems [1-3]. Additionally, these systems are widely used in research and industry for selective separation and purification of many important materials [4-9], and the capability of using membrane selective electrodes in qualitative and quantitative identification carries a latent possibility for their use for sensing and controlling many processes [10]. In environmental fields of application, LMSs are currently attracting great interest, and this interest will increase in the future [11-14]. For the reasons mentioned above, the knowledge of the material transport kinetics in LMSs is a critical point in

understanding and developing applications of these systems. In LMSs, the kinetic theoretical treatment is not as simple as most of its practical procedures because it includes many complex stages based on reversible, consecutive and diffusion phenomena. Many researchers describe the process as an ordinary consecutive mechanism for the material transport from the donor phase to the receptor phase through the membrane [15-25]. Others have merged Fick's second law of diffusion with the kinetics of reversible reactions [26-28], and some researchers have also described the process kinetics as first order mechanisms [29-34]. These schemes have sometimes merged the diffusion process in flux terms [35-40]. These schemes are still deficient because they address certain aspects of the state and neglect others. The driving aim of this study is to drive a mathematical expression that includes consecutive, reversible and diffusion mechanisms describing material transport in LMSs.

Model formulation

Note: The model below was conjugated with practical work about atropine transport using LMSs, but for the generality of the model, that work is published separately [41].

The kinetic model for the transition of atropine (solute) dissolved in benzene (donor light phase) in the first arm of a U-shaped glass tube across a section of water (heavy phase) containing the copper ion Cu^{+2} as a carrier to the second arm containing benzene (accepter light phase) has been derived according to the following equation:

$$(A)_{B1} \underset{k_2}{\overset{k_1}{\rightleftharpoons}} (A)_H \underset{k_1}{\overset{k_2}{\rightleftharpoons}} (A)_{B2}$$

Where $(A)_{B1}$,$(A)_{B2}$ *and* $(A)_H$ are the atropine concentrations dissolved in benzene in the first arm, second arm and aqueous phase, respectively, k_1 is the rate constant of atropine transmission from benzene to the water and k_2 is the rate constant of the reverse process. Panaggio in reference [42] described a similar equation with four rate constants, which we reduced to two rate constants due to the state of symmetry between the first and second arm. In addition, the final solution in the above reference is limited and includes many problems in the determination of the constants of the process. The general equations that describe the changes in the atropine concentration in the three segments with time are as following [43]:

$$\frac{d(A)_{B1}}{dt} = -k_1(A)_{B1} + k_2(A)_H \dots . (1)$$

$$\frac{d(A)_H}{dt} = k_1(A)_{B1} - k_2(A)_H - k_2(A)_H + k_1(A)_{B2} \dots \dots . (2)$$

$$\frac{d(A)_{B2}}{dt} = k_2(A)_H - k_1(A)_{B2} \dots \dots . (3)$$

The atropine concentration in the aqueous phase will increase until it reaches a maximum value. At that point, a steady-state will be established in the aqueous segment, and it will proceed to equilibrium. This phenomenon can be written by setting equation (2) equal to zero as following [44]:

$$\frac{d(A)_H}{dt} = k_1(A)_{B1} - k_2{(A)_H}^{max} - k_2{(A)_H}^{max} + k_1(A)_{B2} = 0 \dots . (4)$$

$$k_1(A)_{B1} + k_1(A)_{B2} = 2k_2{(A)_H}^{max} \dots \dots . (5)$$

Where ${(A)_H}^{max}$ is the concentration of atropine in the aqueous phase when the steady-state is established and also when the full equilibrium of the system is achieved. The substitution of equation (5) into equation (2) produces the following equation:

$$\frac{d(A)_H}{dt} = 2k_2\{{(A)_H}^{max} - (A)_H\} \dots \dots . . (6)$$

This equation is similar to elementary first order equations and can be solved at the boundary conditions, which state that the concentration of atropine in the aqueous phase is zero at

time zero, leading to the following equation:

$$ln\left\{\frac{(A)_H{}^{max}-(A)_H}{(A)_H{}^{max}}\right\} = -2k_2t \dots\dots(7)$$

Equation (7) can be written in the following form:

$$(A)_H = (A)_H{}^{max}\{1 - e^{-2k_2t}\} \dots\dots\dots\dots(8)$$

At t = 0, the limit exponential will be one, and therefore, $(A)_H$ will be zero at the beginning of the process. When the system reaches equilibrium, a steady-state or t = ∞, then the limit exponential will be zero, and thus, the concentration of atropine in the aqueous phase will adhere to the following:$(A)_H = (A)_H{}^{max}$.This is a logical description of what should be going on in the system.

Returning to equations (1) and (3), the substitution of equation (8) in these equations will produce two linear first order differential equations with just two variables. These equations can be solved using the method of integration coefficient (integrating factor) [45]. For example we can rewrite equation (1) after substitution in the following form:

$$\frac{d(A)_{B1}}{dt} + k_1(A)_{B1} = k_2(A)_H{}^{max}\{1 - e^{-2k_2t}\} \dots\dots(9)$$

Where the integration factor (IF) of this equation is $e^{k_1t}dt$. By multiplying this parameter with the above equation, we obtain the following equation:

$$d(A)_{B1}.e^{k_1t} + e^{k_1t}.k_1dt(A)_{B1} = k_2(A)_H{}^{max}\left\{e^{k_1t}dt - e^{(k_1-2k_2)t}dt\right\} \dots(10)$$

Carrying out the integration process of this equation produces the following equation:

$$(A)_{B1}.e^{k_1t} = k_2(A)_H{}^{max}\left\{\frac{e^{k_1t}}{k_1} - \frac{e^{(k_1-2k_2)t}}{(k_1-2k_2)}\right\} + I \dots(11)$$

Where I is the constant of integration, which can be obtained by the substitution of the boundary condition $(A)_{B1} = (A)_{B1}{}^0$ when t = 0, and the concentration of atropine is equal to the initial concentration at time zero leading to equation (12).

$$I = (A)_{B1}{}^0 - k_2(A)_H{}^{max}\left\{\frac{1}{k_1} - \frac{1}{(k_1-2k_2)}\right\} \dots\dots(12)$$

By substituting the constant of integration into equation (11) we obtain the following:

$$(A)_{B1}.e^{k_1t} = (A)_{B1}{}^0 + k_2(A)_H{}^{max}\left\{\frac{[e^{k_1t}-1]}{k_1} - \frac{[e^{(k_1-2k_2)t}-1]}{(k_1-2k_2)}\right\} \dots\dots(13)$$

By rearranging the above equation, equation (14) is obtained.

$$(A)_{B1} = (A)_{B1}{}^0e^{-k_1t} + k_2(A)_H{}^{max}\left\{\frac{[1-e^{-k_1t}]}{k_1} - \frac{[e^{-2k_2t}-e^{-k_1t}]}{(k_1-2k_2)}\right\} \dots\dots(14)$$

When you reach a state of equilibrium (t = ∞), the exponential terms in the previous equation will become zero, and thus, the concentration of atropine remaining in the first arm in the equilibrium state will be as following:

$$(A)_{B1}{}^{\infty} = \frac{k_2(A)_H{}^{max}}{k_1} \leftrightarrow (A)_H{}^{max} = \frac{k_1}{k_2}(A)_{B1}{}^{\infty} \dots\dots..(15)$$

$$(A)_H{}^{max} = K_e(A)_{B1}{}^{\infty} \dots\dots\dots.(16)$$

Where K_e is the thermodynamic equilibrium constant or distribution coefficient of atropine between the aqueous phase and benzene. Similarly we can obtain an equation that expresses the concentration of atropine in the second arm $(A)_{B2}$ by the integration and application of appropriate boundary conditions, resulting in equation (17).

$$(A)_{B2} = k_2(A)_H{}^{max}\left\{\frac{[1-e^{-k_1t}]}{k_1} - \frac{[e^{-2k_2t}-e^{-k_1t}]}{(k_1-2k_2)}\right\} \dots\dots\dots\dots(17)$$

At time zero, it is clear that the concentration of atropine dissolved in benzene in the second arm is zero. At equilibrium (t = ∞), the exponential terms in equation (17) become zero, resulting in equation (18).

$$(A)_{B2}{}^{\infty} = \frac{k_2}{k_1}(A)_H{}^{max} \dots\dots\dots(18)$$

By the comparison of the previous equation with equation (15), it can stated that total equilibrium is established when the equilibrium concentration of atropine dissolved in benzene in the first arm is equal to the concentration of atropine dissolved in benzene in the second arm. In other words, the operation will continue until the concentration of atropine dissolved in benzene is equal in both arms of the U-tube.

The above discussion describes the kinetic model through equations (8), (14) and (17) or the changes in the concentrations of atropine in the three sections with time. when variation in the concentration of atropine with location in one section (this situation exists when there is no efficient stirring of the solutions in these sections) is imposed, the transmission of atropine occurs from the site of a high concentration to the low concentration through the diffusion process.

Kinetic model based on the diffusion process

The solution of the second order differential equation for the diffusion process depends on the specific boundary conditions, which include the time and the size and shape of the system being studied in addition to the initial and final concentrations of the solute [46]. Diffusion in one dimension within a limited volume fits the following general empirical formula [47]:

$$C(x,t) = a + b.e^{-\alpha t}\{c.\sin(\beta x) + d.\cos(\beta x)\} \dots\dots (19)$$

where (a, b) are a constants with the units of concentration, and (c, d) are constants that are determined through the normalization of a concentration distribution spread function over the space of diffusion. Constants (α, β) are identified by making the general above formula obey the second order Fick's law. In this equation, it is clear that at a certain point the solute concentration decreases or increases exponentially with time depending on the sign of constant (β). For the distribution of the solute concentration along the variable (x), it is clear that it is determined by the section containing the trigonometric functions in the diffusion equation. It is not necessary for these trigonometric functions to give periodic behavior with a change of location, but rather they describe the behavior of decline or increase in the function depending on the period of change specified with constant (β).

The constants (a, b) can be determined by determining the initial and final concentrations of the solution at the times (0) and (∞) in the source point (x = 0). When a equilibrium (t = ∞), the second term of equation (19) becomes zero so($C_e = a$). On the other hand, when (t = 0) and at the source point (x = 0), the exponential term in equation (19) is one as well as the distribution of the trigonometric function because all of the solute will be at this point in time (t = 0). Thus,($C(x,t) = C_0$), and so we obtain ($b = C_0 - C_e$).

Constant (c) takes a zero value because the behavior of the diffusion function did not agree with the $sin(\beta x)$ function. This comes from the fact that 100% of the solute will be at the point ($x = 0$) at time zero, while the $sin(\beta x)$ function makes the amount of solute at this point equal to zero, and this contrasts with reality. Thus, equation (19) can be written as following:

$$C(x,t) = C_e + (C_0 - C_e)e^{-\alpha t}\, d\cos(\beta x) \dots (20)$$

By applying the second law of one-dimensional diffusion on this relationship, we obtain the following:

$$\frac{\partial C(x,t)}{\partial t} = D\ \frac{\partial^2 C(x,t)}{\partial x^2} \dots\dots\dots (21)$$

$$\alpha = D.\beta^2 \dots\dots\dots (22)$$

Where (D) is the diffusion coefficient in the units (m^2/sec). The constant (β) can be determined by defining the behavior of $cos(\beta x)$, which fits with the experiment throughout the following states: the concentration of the substance is at its maximum at the starting point and then decreases to a value close to zero with increasing distance far from the source point. The concentration resembles the probabilistic distribution function along the distance, and the probability distribution functions do not take a negative value. Thus, this behavior is similar to the behavior of the cos function in the first quarter of its period$(0 \rightarrow \pi / 2)$. This behavior is happening along the field of diffusion(l); therefore, the function $cos(\beta x)$ will take the form $cos(\frac{\pi}{2l} x)$, and we obtain the following:

$$\beta = \frac{\pi}{2l} \quad and \quad \alpha = D.\frac{\pi^2}{4l^2} \dots\dots.(23)$$

In other words, the form of equation (20) becomes as follows:

$$C(x,t) = C_e + (C_0 - C_e)e^{\frac{-\pi^2}{4l^2}Dt} d \cos\left(\frac{\pi}{2l}x\right) \dots (24)$$

This format is compatible with the second order diffusion equation and consistent with the general formula for diffusion within a limited volume [48]. Returning to the practical way in which the concentration of dissolved atropine change over time was recorded by taking samples from both sides of the tube during the definite time periods, and then they were homogenized to measure the concentration of atropine in the sample with UV-vis. Apparatus. This means that the measurement process was beyond the problem of a concentration gradient along the distance through shaking the sample, which gives a unified concentration along length of the sample during the measurement process. It is clear here that the concentration that was being measured is the mean concentration along the length of the sample. Thus, from the mean value theory [49-51], the measured mean concentration in the specified section is as following:

$$\bar{C}(t) = \frac{\int_0^l C(x,t)dx}{\int_0^l dx} \dots\dots.(25)$$

Carrying out the integration process, we obtain equation (26):

$$\bar{C}(t) = C_e + (C_0 - C_e)e^{\frac{-\pi^2}{4l^2}Dt}.d.\frac{2l}{\pi} \dots.(26)$$

Because the integration above is equivalent to the normalization of the function (in the sense that the amount of material scattered along the space must be 100% or 1 in all sections) and because the constant d contributes to this condition, the remainder of the extent of trigonometric functions after achieving this condition must be equal to 1, which means that $\frac{d.2l}{\pi} = 1$. Thus the value of the constant is $(d = \frac{\pi}{2l})$. The use of the average concentration method that we performed practically allows simplifying the (x) dependent part of the concentration function to just sin and cos trigonometric functions that appear in equation (19), which is required to satisfy the Fick's second law and results in no need to insert a Fourier series as in reference [47].

It remains to be noted that the primary concentration here, C_0 replaced with concentration C_i of acquired from the equations (8), (14) and (17). This is similar to the case in [27, 28] that qualifies $\frac{\partial C}{\partial t}$ term in the diffusion equation with $\frac{dC}{dt}$ in the kinetic equation, but the qualifying step in this work was performed after integration. This will give the concentration at the three sections with the contribution of the diffusion process so resulting in the following:

$$\bar{C}(t) = C_e + (C_i - C_e)e^{\frac{-\pi^2}{4l^2}Dt} \dots.(27)$$

$$\overline{(A)_{B1}}(t) = (A)_{B1}{}^{\infty} + \{(A)_{B1}{}^{0}e^{-k_1 t} + k_2(A)_H{}^{max}\left\{\frac{[1-e^{-k_1 t}]}{k_1} - \frac{[e^{-2k_2 t}-e^{-k_1 t}]}{(k_1-2k_2)}\right\} - (A)_{B1}{}^{\infty}\}e^{\frac{-\pi^2}{4.l_{B1}{}^2}D_B t} \ldots.(28)$$

$$\overline{(A)_H}(t) = (A)_H{}^{\infty} - (A)_H{}^{\infty}e^{-2k_2 t}e^{\frac{-\pi^2}{4.l_H{}^2}D_H t} \ldots.(29)$$

$$\Longrightarrow \overline{(A)_H}(t) = (A)_H{}^{\infty}\{1 - e^{-\left(2k_2+\frac{\pi^2}{4.l_H{}^2}D_H\right)t}\} \ldots\ldots.(30)$$

$$\overline{(A)_{B2}}(t) = (A)_{B2}{}^{\infty} + [k_2(A)_H{}^{\infty}\left\{\frac{[1-e^{-k_1 t}]}{k_1} - \frac{[e^{-2k_2 t}-e^{-k_1 t}]}{(k_1-2k_2)}\right\} - (A)_{B2}{}^{\infty}]e^{\frac{-\pi^2}{4.l_{B2}{}^2}D_B t}.(31)$$

The linear form of equation (30) can be obtained by rearranging and taking the logarithm and is as following:

$$\ln\left[\frac{(A)_H{}^{\infty}}{(A)_H{}^{\infty}-\overline{(A)_H}(t)}\right] = \left(2k_2 + \frac{\pi^2}{4.l_H{}^2}D_H\right)t \ldots\ldots(32)$$

By drawing $\ln\left[\frac{(A)_H{}^{\infty}}{(A)_H{}^{\infty}-\overline{(A)_H}(t)}\right]$ versus time, a straight line going through the origin point will result in its slope being equal to $\left(2k_2 + \frac{\pi^2}{4.l_H{}^2}D_H\right)$. In contrast, the mean concentration of dissolved atropine in benzene in both arms of the tube does not give a simple expression that can be used to calculate the constants in an experimental way to compare the proposed kinetic model with experimental results, but it can simplify equations (28) and (31) through the study of these equations during the initial time periods (primary sections of the curves), where in the initial time periods, the exponential terms can be replaced by $e^{(-z)} \approx 1 - z$ according to the Taylor series [52]. Therefore, equation (28) becomes the following:

$$\overline{(A)_{B1}}(t) = (A)_{B1}{}^{\infty} + \{(A)_{B1}{}^{0}(1-k_1 t) - (A)_{B1}{}^{\infty}\}(1 - \frac{\pi^2}{4.l_{B1}{}^2}D_B t) \ldots(33)$$

This can be arranged into the form

$$\overline{(A)_{B1}}(t) = (A)_{B1}{}^{0} - \left[(A)_{B1}{}^{0}k_1 + \frac{\pi^2}{4.l_{B1}{}^2}D_B\left((A)_{B1}{}^{0} - (A)_{B1}{}^{\infty}\right)\right]t + (A)_{B1}{}^{0}\frac{\pi^2}{4.l_{B1}{}^2}D_B k_1 t^2 \ldots.(34)$$

The linear form becomes as follows:

$$\frac{(A)_{B1}{}^{0}-\overline{(A)_{B1}}(t)}{t} = \left[(A)_{B1}{}^{0}k_1 + \frac{\pi^2}{4.l_{B1}{}^2}D_B\left((A)_{B1}{}^{0} - (A)_{B1}{}^{\infty}\right)\right] - (A)_{B1}{}^{0}\frac{\pi^2}{4.l_{B1}{}^2}D_B k_1 t..(35)$$

By plotting $\frac{(A)_{B1}{}^{0}-\overline{(A)_{B1}}(t)}{t}$ versus time, a straight line is obtained with the intercept $=\left[(A)_{B1}{}^{0}k_1 + \frac{\pi^2}{4.l_{B1}{}^2}D_B\left((A)_{B1}{}^{0} - (A)_{B1}{}^{\infty}\right)\right]$, and its slope is $(A)_{B1}{}^{0}\frac{\pi^2}{4.l_{B1}{}^2}D_B k_1$.

Applying the same procedure to equation (31), we obtain equation (36):

$$\overline{(A)_{B2}}(t) = (A)_{B2}{}^{\infty}\frac{\pi^2}{4.l_{B2}{}^2}D_B t \ldots(36)$$

The above equation states that when the primary section of the concentration $\overline{(A)_{B2}}(t)$ is plotted versus time, we will obtain a straight line through the origin point that has the slope $(A)_{B2}{}^{\infty}\frac{\pi^2}{4.l_{B2}{}^2}D_B$. Equations (32), (35) and (36) in addition to the equilibrium constant relationship which is:

$$K_e = \frac{k_1}{k_2} = \frac{(A)_H{}^{\infty}}{(A)_B{}^{\infty}} \ldots\ldots\ldots(37)$$

All of the kinetic constants for this system can be determined, thus allowing the supposed kinetic model to be compared with the experimental results for the purpose of determining the efficiency of this model to describe the overall process.

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نموذج حركي لانتشار المذاب في انظمة الاغشية السائلة

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الخلاصة:

في هذه الدراسة تم صياغة نموذج حركي لانتقال المذاب في انظمة الاغشية السائلة. دمج هذا النموذج بين حركيتي العمليات المتتابعة و الانعكاسية مع تعبير شبه مشتق للانتشار، منتجا معادلات تصف تركيز المذاب في المقاطع الثلاثة (الواهب و المستقبل و الغشاء السائل). تم تعديل المعادلات الناتجة الى الشكل الخطي و الذي يتحقق تحت ظروف خاصة لغرض تبسيط الحصول على الثوابت الحركية المهمة للعملية بصورة تجريبية.

الكلمات المفتاحية: نموذج حركي، الانتشار، الاغشية السائلة، نموذج رياضي، انتشار المذاب في LMS