SUMMARY

For the prediction of the process of separation of emulsions and selection of optimal conditions for these processes an appropriate mathematical description is required. Thereby, a mathematical model of the convective diffusion of the continuous process in the space between the membrane and the impermeable wall, and the molecular diffusion in membrane was developed in the form of an analytical solution of the corresponding differential equations of transient diffusion for substance transfer in the moving fluid in the membrane, where due to membrane’s selective properties the constant separation of substances takes place. In this study authors addressed and analytically solved a diffusion boundary-value problem describing the mechanism of baromembrane emulsions separation. This value problem may find an application in the study of confectioner's cream production cycle.

Keywords: mathematical description, membrane, emulsion, convection and molecular mass transfer, microfiltration, permeate.

1. INTRODUCTION

The separation of substances using semipermeable membranes (ultrafiltration, microfiltration and reverse osmosis) is widely used in the food industry, including the processing of dairy products [1–16].

Membrane separation of emulsions allows to efficiently solve the problem of skimmed milk and whey processing in the production of the diet and dessert foods.

Scheme of the separation of solutions (emulsions) can be represented as follows: a solution moves between an impermeable wall and a semipermeable membrane. A liquid boundary layer is formed on the surface of this membrane. Separated from the flow core substances, the fat globules are transferred by convection and molecular diffusion through the boundary layer to the membrane. Ultimately, the permeate is filtered through a semipermeable membrane by Darcy’s law.

For the prediction of the processes of the separation of emulsions and selection of optimal conditions for these processes, an appropriate mathematical description is required.

Known methods for solving the tasks of the separation of emulsions using baromembrane techniques include the solution of unsteady mass conductivity in an unstirred medium of batch cell [17–20], which is commonly used in laboratory practice, or solving the problems of steady mass transfer in laminar flow in a channel with one semipermeable wall [21–24]. However, the mentioned problems do not correspond to real conditions and do not contain common formulations of the problem. In Refs. [25,
26] by considering the concentration of substances in the boundary diffusion layer, the theory of the separation of solutions by baromembranes methods has been developed and the general solution to the problem and its particular solution important for practice have been obtained. However, problems considered in these studies are solved for the boundary layer, within which the molecular transfer of substances takes place. In Ref. [27], a mathematical model is developed, applicable to the ultrafiltration of whey, in the form of analytical solutions of the convective diffusion equation to coordinate the initial distribution of the concentration of the separated protein product.

A quantitative description of diffusion processes occurring in the membrane – the technological partition, ensuring, due to its selective permeability, properties the separation of substances mostly without chemical transformations – is based on three main approaches: statistical, thermodynamic and phenomenological [9, 28]. On the basis of the phenomenological approach some problems are posed and solved by Nikolayev [9].

The combined corresponding boundary-value problem for a continuous process of the convective diffusion in the space between the wall and the impermeable membrane and the molecular diffusion in the membrane can become a development of the ideas mentioned above.

2. MODELLING

The mathematical description of the observable process corresponds to the solution to a one-dimensional non-stationary diffusion equation with drift (convective diffusion equation for the transport of matter in moving with speed \( \omega \) liquid):

\[
\frac{\partial c_1}{\partial t} = D_1 \frac{\partial^2 c_1}{\partial x^2} - \omega \frac{\partial c_1}{\partial x} \quad (0 < x < l_1, \ \tau > 0)
\]  

(1)

with the initial condition:

\[
c_1(x,0) = c_0 = \text{const}
\]

(2)

and with solution to the equation of unsteady one-dimensional diffusion in the membrane:

\[
\frac{\partial c_2}{\partial t} = D_2 \frac{\partial^2 c_2}{\partial x^2} - \omega \frac{\partial c_2}{\partial x} \quad (0 < x < l_2, \ \tau > 0)
\]  

(4)

and with following initial and boundary conditions:

\[
c_2(x,0) = 0
\]

(5)

\[
c_2(l_1,\tau) = c_2(l_1,0)
\]

(6)

\[
-D_2 \frac{\partial c_2}{\partial x}(0,\tau) = -D_1 \frac{\partial c_1}{\partial x}(0,\tau)
\]

(7)

\[
c_2(l_2,\tau) = c_c = \text{const}
\]

(8)

At this point we introduce the following notation: \( c_i = c_i(x, \tau), i = 1, 2 \) – concentration of a separated substance in the solution and the membrane, respectively; \( c_i(x, \tau) = c(x, \tau), c_0 \) – initial concentration; \( c_c \) – concentration of the medium, in which comes the substance diffused through the membrane; \( x \) – current coordinate, \( l_1 \) and \( l_2 \) – thickness of the liquid layer and the membrane, respectively; \( \tau \) – time; \( D_1 \) and \( D_2 \) – molecular diffusion coefficients.

Equation (5) is the initial condition, referring to a situation where, at the beginning of the process, the flat membrane does not contain a diffusing agent; the boundary conditions of the fourth kind, Eqs. (6) and (7), express the equality of concentration and mass flow of substance at the membrane-solution border.

The boundary condition of the first kind, Eq. (8), reflects the fact that the substance diffused through the membrane is transferred in a sufficiently large volume without changing its concentration \( c_c \), and quickly moved away from the membrane.

Posed boundary-value problem, Eqs. (1) – (8), is solved analytically by the Laplace integral transformation. Concentration distribution of the substance separated in the membrane have been obtained as follows:

\[
\theta(X,F_0) = \theta_0 - \theta_c X - \frac{\theta_c (1 - e^{-\mu_0 F_0})}{1 - e^{-\mu_0}} + \theta_0 K_D K_i P_e - \frac{\theta_0 K_D K_i P_e}{1 - e^{-\mu_0}}
\]

\[
-2 \sum_{m=1}^{\infty} \frac{c_m \mu_m}{\mu_m^2} \sin(\mu_m X) e^{-\mu_m^2 F_0}
\]

\[
-2 \sum_{m=1}^{\infty} (-1)^m (1 - \frac{1}{\mu_m^2}) \sin(\mu_m X) e^{-\mu_m^2 F_0}
\]

(9)

where:

\[
\theta(X,F_0) = c_0 - c(x, \tau) - \text{dimensionless concentration;}
\]

\[
\theta_0 = -\frac{c_0 - c_c}{c_0 - c_c}; \quad \theta_c = c_0 - c_c;
\]

\[
X = \frac{x}{\delta} - \text{dimensionless coordinate;}
\]

\[
F_0 = \frac{D_2 \tau}{\delta} - \text{Fourier number;}
\]

\[
P_e = \frac{\omega l_1}{D_1} - \text{Peclet number;}
\]

\[
K_D = \frac{D_1}{D_2}; \quad K_i = \frac{l_2}{l_1}; \quad \mu_n = n \pi
\]

(10)

\[
\mu_m - \text{successive positive roots of the characteristic equation:}
\]

\[
\frac{P_e}{2} + \frac{\gamma_m}{\sqrt{K_D K_i}} \text{cth} \frac{\gamma_m}{\sqrt{K_D K_i}} = -\frac{\mu_n}{K_D K_i}
\]

(10a)
Concentration of the dispersed phase in liquids can vary from very low (droplets float freely in a dispersion medium) to very high – 95-99% (dispersion medium is only a thin layer between the droplets) [5].

The most promising method for the separation of emulsions, especially for the fine-dispersed ones, is microfiltration [6–8]. It can be used for the increase of the content of the emulsified oil in an aqueous emulsion from 1–10 to 90%, practically yielding pure water in permeate [7].

To describe this process the diffusion boundary-value problem, complementing the author’s study [9] is considered.

From a limited volume $V$ containing a substance with a concentration $c_0$, the substance diffuses into a volume with a constant concentration $c_0$ through a flat membrane of defined dimensions (of a thickness $L$ and an area $S$).

It is assumed that there are no diffusion fluxes from the ends of the membrane.

Mathematical description of the observable process equals to the solving of the one-dimensional non-stationary diffusion equation:

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial x^2}; \quad (0 < x < L, \; \tau > 0) \quad (11)$$

with the following initial and boundary conditions:

$$c(x,0)=0 \quad (12)$$

$$V \frac{\partial C(0,\tau)}{\partial \tau} = -DS \frac{\partial C(0,\tau)}{\partial x} \quad (13)$$

$$c(L,\tau) = C_c = \text{const} \quad (14)$$

Equation (12) is an initial condition based on the premise that, at the beginning of the process, the flat membrane does not contain a diffusing agent.

The boundary condition, Eq. (13), shows that the concentration changing intensity of the diffusing substance at the border membrane-volume $V$ causes the mass flow of the diffusing agent inbye the membrane.

The boundary condition of the first kind, Eq. (14), reflects the fact that the substance diffused through the membrane is transferred in a sufficiently large volume without changing its concentration $c_c$, and quickly moved away from the membrane.

Solution to the boundary diffusion, Eqs. (11) – (14), by the Laplace integral transformation leads to the following expression for the diffusing substance concentration distribution in the membrane:

$$\theta(X,F_\tau) = \frac{c(x,\tau)}{c_c} = 1 + \sum_{n=1}^{\infty} \frac{2(\delta \cos(\mu_n X) + \mu_n \sin(\mu_n X))}{\mu_n^2 - \delta(1-\delta)\cos\mu_n} \exp\left(-\mu_n^2 F_\tau\right) \quad (15)$$

where:

$$\mu_n - \text{successive positive roots of the characteristic equation:}$$

$$\frac{1}{\delta} = -\frac{\mu^2}{\delta} \quad (16)$$

Averaging over the volume of the dimensionless concentration $\theta(X,F_\tau)$ using the following formula:

$$\bar{\theta}(F_\tau) = \int_{0}^{1} \theta(X,F_\tau) dX \quad (17)$$

leads to the expression for the mass of the diffusing substance absorbed by the membrane:

$$\bar{\theta}(F_\tau) = \frac{c(\tau)}{c_c} = 1 + \sum_{n=1}^{\infty} \frac{2\left(1 + \left[1 + \frac{\delta}{\mu_n}\right] \cos \mu_n \right)}{\mu_n^2 - \delta(1-\delta)\cos\mu_n} \exp\left(-\mu_n^2 F_\tau\right) \quad (18)$$

For small values of $F_\tau$ the solution described with Eq. (15) is a bit inconvenient. Most suitable for small values of $F_\tau$ solution is obtained as follows:

$$\theta(X,F_\tau) = \frac{c(x,\tau)}{c_c} = \text{erfc}\frac{1-X}{2\sqrt{F_\tau}} + \text{erfc}\frac{1+X}{2\sqrt{F_\tau}}$$

where:

$$\text{erfc} = 1 - \text{erfc} = \frac{2}{\sqrt{\pi}} \int_{0}^{z} e^{-Z^2} dZ \quad (19)$$

$$\text{erfc}Z = 2 \sqrt{\frac{2}{\pi}} \int_{0}^{z} e^{-Z^2} dZ \quad \text{Gauss error function (tabulated)}$$

The resulting solution obtained with Eq. (15) describes the mechanism of baromembrane separation of emulsions components and can be used for further research of the cream confectionery production process serving as a basis for its optimization.

The symbols used in the last several equations have the following meaning:

$c_c$ – the concentration of the medium into which the diffused through the membrane substance comes,

$x$ – the current coordinate,

$L$ – thickness of the membrane,

$\tau$ – time,
3. CONCLUSIONS

The following conclusions have been reached from the performed analyses:

1. For the prediction of the processes of the separation of emulsions and selection of optimal conditions for these processes, a mathematical model of the convective diffusion of the continuous process in the space between a membrane and an impermeable wall, and the molecular diffusion in membrane was developed in the form of an analytical solution of the corresponding differential equations of transient diffusion for substance transfer in the moving fluid in the membrane, where due to membrane’s selective properties the constant separation of substances is provided.

2. Posed and analytically solved, the diffusion boundary-value problem describes the mechanism of separation of baromembrane emulsion components passing through a flat membrane from a limited volume into a volume with a constant concentration of the substance.

3. The developed mathematical models can be applied to the study of the processes in the food industry, in particular to the production process of the confectionery’s cream.

4. REFERENCES


Kako bi se mogli predvidjeti procesi odvajanja emulzija te odabrati optimalni uvjeti za ostvarivanje istih, potreban je prikladan matematički model koji bi opisao te procese. Stoga, razvijen je matematički model konvektivne difuzije neprekidnih procesa u prostoru između membrane i nepropusnog zida te molekularne difuzije u membrani u obliku analitičkog rješenja i odgovarajućih diferencijalnih jednadžbi nestacionarne difuzije za prijenos tvari u pokretnom fluidu u membrani, gdje se zbog selektivnih svojstava membrane događa neprekidno odvajanje supstanci. Autori su se u ovome radu bavili i iznašli analitičko rješenje rubne zadaće za difuziju koje opisuje mehanizam baromembranskog odvajanja emulzija. Rješenje ove zadaće može naći primjenu u proučavanju proizvodnog procesa slitka tučenog vrhnja.

**Ključne riječi:** matematički opis, membrane, emulzija, konvekcija i prijenos mase na razini molekule, mikrofiltracija, filtrat.