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COUPLED HEAT AND MASS TRANSFER IN MULTISTAGE ELECTROMEMBRANE FACILITIES FOR SEPARATING LIQUID SYSTEMS

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An analysis of works dealing with investigations of mass and heat transfer in electromembrane systems (EMS) is presented. Basic approaches to the mathematical description of the phenomena of transfer in the solutions and membranes of EMS and mathematical models of the mass and heat transfer processes are considered.

In recent years membrane separation of liquid media has been utilized increasingly extensively in various branches of industry. This has stimulated a considerable amount of research into the theoretical and practical problems of membrane processes and apparatus at academic and industrial scientific-research institutes, universities, design bureaus, scientific-industrial associations, etc.

At the present time, a wide variety of membrane facilities are being built in the countries of the CIS [1, 2]. In Russia, the largest plants manufacturing membrane equipment are the Tambov. Factory "Komsomolets" and Industrial Association "Tambovmash." The tables that follow list the main types of membrane apparatus produced by these enterprises and their basic characteristics. In the last few years there appears to be a tendency toward a significant expansion of the sphere of application of membrane methods for separation of liquid media. Along with the traditional field of application of membrane technology for water demineralization [3-13], membrane methods are finding use for sewage treatment [10, 14-16], in the chemical [14, 15, 17-19], food-manufacturing [7, 20-23], and pharmaceutical industries, biotechnology [24-27], etc.

Distinctive attributes of the application of membrane technology methods in industries that are unconventional for them are a wide range of treated solutions, multiplicity of components, wide ranges of the concentrations of desired components, temperatures, flow rates, etc.

When treating liquid media (especially industrial sewage) by membrane methods, various final objectives can be set out: (1) the production of just demineralized water; (2) the production of just a concentrated solution (concentrate); (3) the simultaneous production of demineralized water and concentrate (the problem of demineralization-concentration). In this case the demineralized water (dialyzate) is used, as a rule, in the systems of closed water recycling, whereas the concentrate is either used as a finished product or subjected to further treatment (evaporation, crystallization, and so on).

To realize these tasks, use is made of various hydraulic schemes of joining chambers and apparatus, but practically always one has to pass solutions through several chambers connected in series, groups of chambers or even through several series-connected apparatuses. Moreover, for attaining the required concentrations of solutions, one often has to form circuits. Thus, the processes of membrane technology are taken to proceed, as a rule. in multichamber apparatuses or facilities and often with the closed circulation of solutions.

The present work considers problems of mass and heat transfer in multistage electromembrane facilities. For the sake of definiteness, we shall consider multichamber electrodialyzing and electroosmofiltrational facilities (see Figs. 1 and 2).

Generally, an electrodialyzing facilities (Fig. 1) includes electrodialyzer 1, pumps 2-5, tanks for solutions 6-9, and electric power supply unit 10. In turn, the electrodialyzer consists of electrode and working chambers. The working chambers are formed by alternating anion-exchange (AEM) and cation-exchange (CEM) membranes

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TABLE 1. Membrane Facilities Built by "Komsomolets" Factory (the data are provided by "Komsomolets")

Type of facility	Membrane process	Capacity, m^3/h		Pressure, MPa	Selectivity, %
		original solution	permeate		
UMR-2.5-2-1	RO	2.5	1.3-2.0	4.0	90-97
UMR-4-1-1	RO	4.0	2.0-3.0	5.0	90-97
UMR-4-2-1	RO	4.0	2.0	4.0	90-94
UMR-6.5-1-2	RO	6.5	4.0-5.0	4.0-5.0	>90
UMR-10-2-1	RO	10.0	6.0-8.0	5.0	>90
UMR-16-1-1	RO	16.0	10.0-14.4	5.0	>90
UMR-25-2-2	RO	25.0	12.5-20.0	3.5	>96
UMR-25-2-1	RO	25.0	15.0-21.0	5.0	>90
UMR-50-2-2	RO	50.0	45.0	3.0-4.0	90-97
UMR-4-2-2	RO	4.0	2.5-3.5	Technological lines for water demineralization	
UMR-10-2-2	RO	10.0	6.0-8.0		
UMK-P-10	RO	10.0	Technological lines for producing feed water for boiler installations		
UMK-P-16	RO	16.0			
UNK-P-25	RO	25.0			
UMK-P-50	RO	50.0			
UMT-3-1	UF	0.16	0.15	0.45	95-99
UMT-6-1	UF	0.32	0.30	0.45	95-99
UMT-9-1	UF	0.48	0.45	0.45	95-99
UMT-18-1	UF	0.96-2.0	0.9-1.5	0.45	95-99
UMT-36-1	UF	1.96-2.4	1.8-2.2	0.45	95-99
UMT-108-1	UF	5.76-6.3	5.4-6.0	0.45	95-99
UMT-36-2	UF	-	1.8	0.50	99
UMT-108-2	UF	-	5.4	0.50	99

Note: 1. In UMR-series facilities coiled membrane elements are employed, and tubular in UMT-series facilities. 2. For UMT-series fasilities the selectivity is given on the basis of suspended particles (99%) and oil products (95%). 3. In UMT-36-2 and UMT-108-2 facilities flotational removal of concentrate is envisaged.

(there may be a different combination of membranes, say, ion-exchange and bipolar ones, etc.). The chambers are equipped with turbulizers. The electrode chambers contain electrodes to which a constant is fed voltage is fed from the power supply unit. With respect to the dialyzate and concentrate circuits, the working chambers can be connected in different ways. In the scheme of Fig. 1 their series connection is shown. The concentrate and dialyzate move successively through all the working chambers of the electrodialyzing apparatus, enter intermediate tanks, and go again into the electrodialyzer. Under the action of the electric field forces the ions present in the solution move through the membranes from the demineralization chambers to the concentration chambers. To wash out the gases formed as a result of electrode reactions, the solutions circulate through the electrode chambers.

The electroosmofiltrational facility (Fig. 2) consists of electroosmofiltrational apparatus 1, pump 2, filtrate collectors 3 and 4, intermediate tank 5, and electric power supply unit 6.

The working chambers of the electroosmofiltrational apparatus are formed by a pair of semipermeable membranes (reverse-osmotic, ultrafiltrational, etc.) located on porous metallic substrates that also act as electrodes. The substrate is mounted on a dielectric plate. The outer chambers contain electrodes to which a constant voltage is fed from the power supply unit. By an excess-pressure pump the solution is driven successively through the working chambers of the apparatus. Under the action of the pressure difference, the solvent (water) passes through the membrane, enters the substrate, and leaves the system. Under the action of the electric field forces and because of imperfections in the membrane, the ions and molecules of the dissolved substance move through the membrane,

TABLE 2. Membrane Facilities Built by Industrial Association "Tambovmash" (the data are provided by the Tambov Special Technological Design Office)

Type of facility	Membrane process	Capacity, m^3/h	Working pressure, MPa	Initial and final salt content, g/liter	Remark
Desalination of salty water for drinking water supply					
EOU-NIIPM-25M	ED	1.0	0.3	6.0-0.5	
EOU-2-1-R21U-250	ED	10.4	0.3	6.0-0.5	
EOU-M-1000	ED	0.04	0.05	6.0-0.5	
EOU-R11-40	ED	1.25	0.3	6.0-0.5	
EOU-2R11-75	ED	3.1	0.3	6.0-0.5	
EOKS-R11-40	ED	1.25	0.3	6.0-0.5	Container stations
EOKS-R11-75	ED	3.1	0.3	6.0-0.5	
Preparation of process water					
EOU-2-1-R21K-50	ED	50.0	0.3	1.2-0.6	
EOU-4-P-R21K-50	ED	50.0	0.3	1.2-0.3	
EOU-6-Sh-R21K-50	ED	50.0	0.3	1.2-0.15	
Special-purpose facilities					
Electrodialyzer for cleaning glyoxol	ED	12.0	0.3		Three-stage facility
RM-7.2	UF	7.2	0.45		
Ultrafiltrational for silver extraction	UF	4.8	0.45		

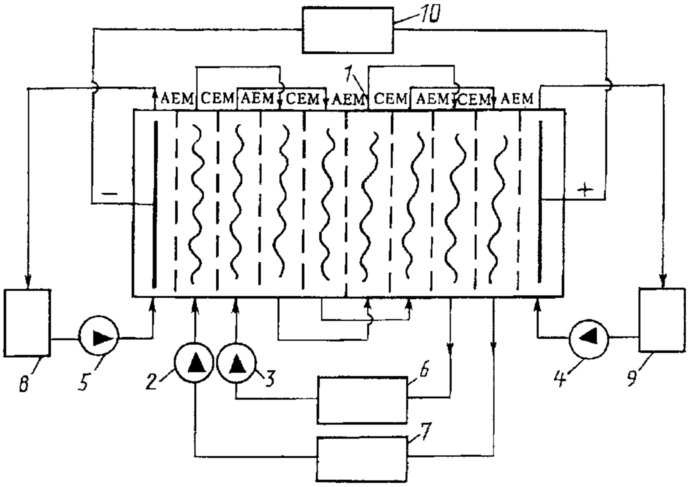

Fig. 1. Schematic of an electrodialyzing facility.

enter the substrate, undergo electrochemical transformations, and, together with the filtrate, leave the apparatus. By using chambers of different constructions, one can realize different schemes of the separation of solutions in these apparatuses [5, 6].

Electromembrane facilities of the types described are used in both industry and laboratory practice. We used them for demineralization-concentration of sulfate-, aniline-, and morpholine-containing sewage from the production of chemicals-additives for polymer materials on laboratory and pilot-plant scales.

In the process of electromembrane separation of solutions a change in the concentrations, volumetric flow rates, and temperatures of solutions occurs both over the chambers of the apparatus and in time. The changes in the concentrations and volumetric flow rates result from mass transfer phenomena (transportation of the ions of

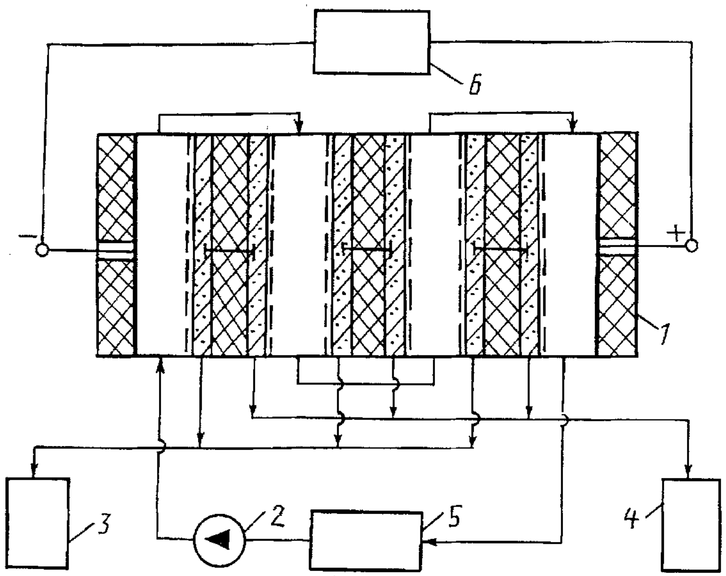

Fig. 2. Schematic of an electroosmofiltrational facility.

dissolved substances and solvent through membranes), whereas the changes in the temperatures of the solutions and membranes are caused by passage of electric current through them and Joule heat release.

As illustrative examples, Figs. 3 and 4 present kinetic curves of electrodialyzing and electroosmofiltrational separation of sulfate-, aniline- and morpholine-containing aqueous solutions.[*]

From the graphs given it is seen that the concentrations, temperatures, and volumes of the solutions may change within wide ranges in the process of separation. The range of these changes depends substantially on the operational parameters of the process (current density, initial concentrations of the solutions, etc.).

The concentration and temperature changes in the solutions and membranes lead to a change in the kinetic characteristics of electro-, mass, and heat transfer (electric conductivity, transference numbers, diffusional, osmotic, and electroosmotic permeabilities, etc.), which ultimately has a strong effect on the kinetics of the process itself; i.e., the processes of electro-, mass, and heat transfer are interrelated, and this should be taken into account when developing mathematical models and engineering methods for designing electromembrane systems (EMS). However, until recently the majority of reseach workers concentrated mainly on problems of mass transfer in these systems and much less on problems of heat transfer.

We shall take a brief look at the basic concepts of the theory of mass and heat transfer in EMS. First, we shall dwell on mass transfer phenomena.

The mechanism of mass transfer in EMS has been given rather detailed consideration in the literature [3, 5-9, 22, 28-34].

The basic principles underlying the mechanism of mass transfer through a membrane separating the relinquishing and receiving solutions (dialyzate and concentrate, respectively) are traditional for mass transfer theory (see Fig. 5a; for definiteness a scheme of mass transfer through an ion-exchange membrane (IEM) is given). Here, just as for many processes of mass transfer in heterogeneous systems, it is assumed that the mass transfer is composed of the following stages: mass transfer from the core of the relinquishing flow to the surface M of the membrane; transfer through the phase interface; transfer through the membrane; transfer through the phase boundary; mass transfer from the phase interface to the receiving flow core. Depending on which of the phases (external, internal or both) limits the mass transfer, one distinguishes processes with external, internal, and mixed-diffusional kinetics. In electromembrane processes, mass transfer is most often limited by the external phase, thus

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[*] The kinetic relations were obtained for the electrodialyzing separation by S. V. Mukin and for the electroosmofiltrational separation by S. I. Lazarev

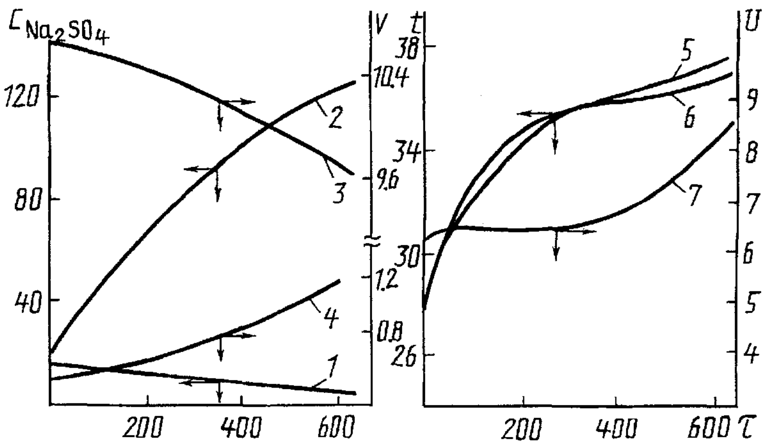

Fig. 3. Concentrations (1, 2), volumes (3, 4), and temperatures (5, 6) of dialyzate and concentrate, respectively, and voltage across electrodes (7) vs time in the course of electrodialyzing demineralization and concentration of a solution of sodium sulfate ($i = 65.4$ A/m^2, $n = 8$, $S = 0.06$ m^2). $C_{Na_2SO_4}$, g/liter; V, liter; t, °C; U, V; τ, min.

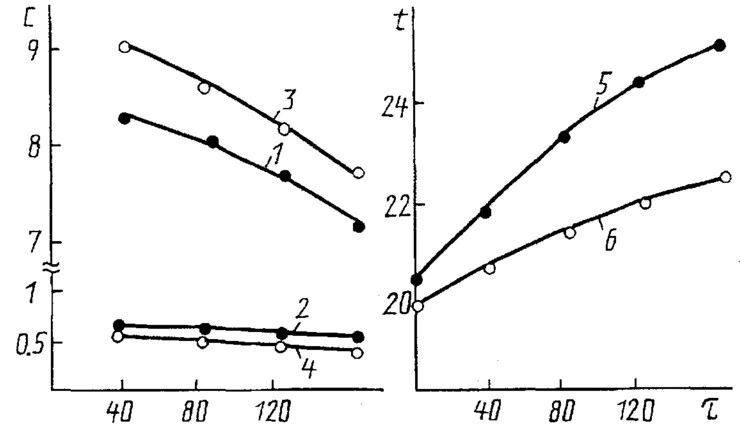

Fig. 4. Changes of the concentrations of aniline (1, 2) and morpholine (3, 4) in a tank (1, 3) and in the filtrate (2, 4) and of the temperatures of aniline (5) and morpholine (6) solutions in a tank in the course of electroosmofiltrational separation of aniline- and morpholine-containing solutions ($i = 18.6$ A/m^2, $n = 7$, $S = 0.008$ m^2, $\Delta P = 1$ MPa).

giving rise to the phenomenon of concentrational polarization. Mass fluxes through the membrane in an EMS are composed (Fig. 5) of dissolved-substance fluxes (migrational N_{mig}, diffusional N_{dif}, and convective N_{conv}) and solvent fluxes (electroosmotic m_{eo}, osmotic m_{osm}, convective m_{conv}). Aside from those listed, hydroxyl (OH^-) and hydroxonium (H_2O^+) ions can participate in electro- and mass transfer (especially in regions near and beyond the limit).

When specific processes are considered, some of the components of the mass fluxes may be ignored because of their smallness.

As noted above, mass transfer in the solution phase is characterized by concentrational polarization, which in many ways determines the kinetics of the electromembrane process as a whole. The essential information about concentrational polarization for neutral and charged membranes in the presence and absence of an electric field can be found in a number of monographs by domestic and foreign authors [3-10, 14, 20, 22, 27-30, 32]. There are many periodicals devoted to experimental and theoretical investigations of the phenomena of concentrational polarization in EMS. The most popular methods used for investigating concentrational polarization in EMS are the

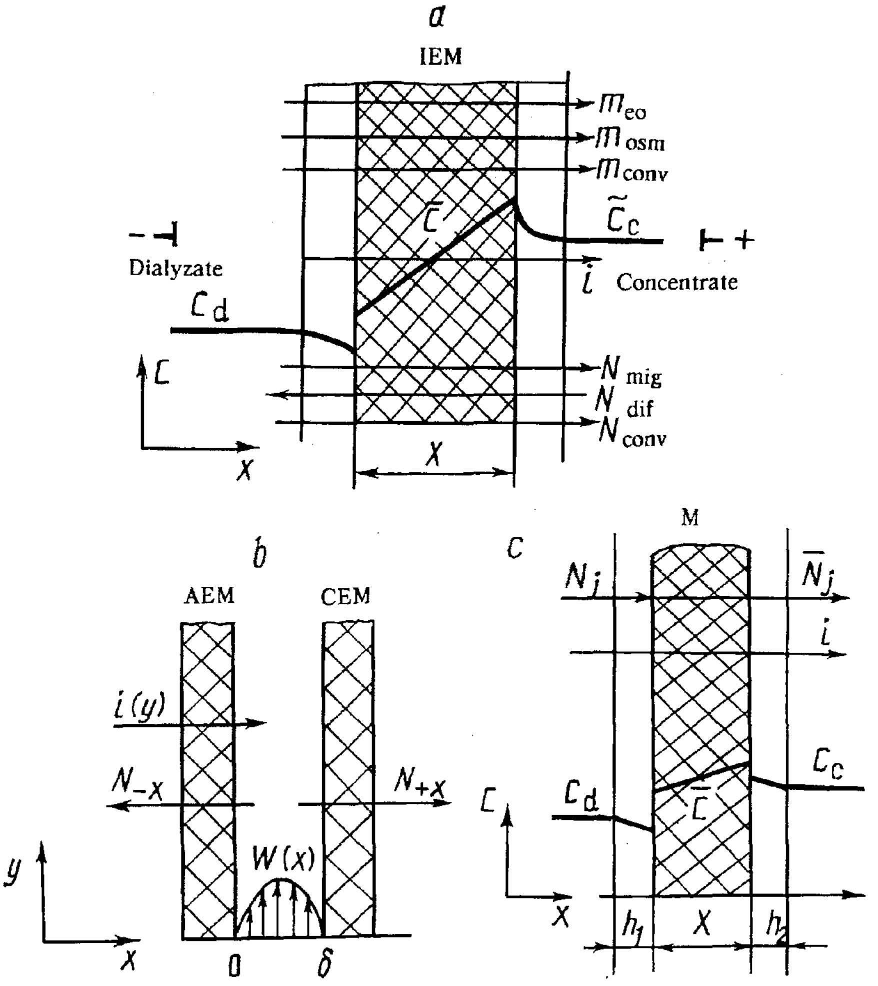

Fig. 5. Schemes of mass transfer through a membrane (a), in an electromembrane chamber (b), and in diffusional layers and in a membrane (c).

methods of recording volt-ampere characteristics in continuous-flow cells [30] and of laser interferometry [32]. Relatively recently, the method of recording volt-ampere characteristics with a rotating membrane disk has been applied for this purpose [32, 35].

When we consider concentrational polarization in EMS, the most important, and frequently the ultimate, problem is that of the critical (limiting) current density i_{cr} that delineates the pre- and trans-limiting regions of operation of EMS.

The intensity of mass transfer in the precritical region is not high, but the current efficiency is high and the expenditures of energy are relatively small. In the postcritical region the intensity of mass transfer grows, but in this case the current efficiency falls and energy expenditures increase.

The limiting density is determined by many factors: the nature and composition of the electrolyte, kind of membranes, temperature, etc. The most popular relation used for determining the critical current density [30, 36, 37] is

$$\mathrm{Sh} = k\,\mathrm{Re}^{n}\,\mathrm{Sc}^{m}, \tag{1}$$

where $\mathrm{Sh} = i_{cr}(\bar{t} - t)\delta/FDC$ is the Sherwood number; $\mathrm{Re} = W\delta/\nu$ is the Reynolds number; $\mathrm{Sc} = \nu/D$ is the Schmidt number; k, n, and m are numerical coefficients.

The numerical values of the coefficients k, n, and m depend on the geometry of the system and the hydrodynamic situation in the apparatus. In [38], with reference to various literature sources, the following limits on the variation of these coefficients are given: n = 0.33–0.8; m = 0.25–0.38 (the lower limit corresponds to a laminar moving flow and the upper to a turbulent flow). These limits encompass the coefficients n and m obtained in [36, 37] for different types of turbulizers. Moreover, it follows from these works that an equation of type (1) can be used successfully in a rather wide temperature range. It should also be noted that there are works dealing specifically with the influence of temperature on the limiting current density [39-41]. Besides the form of Eq. (1) given, there are also other forms of relations available for determining the critical current density [32, 42, 43]. However, they are chiefly used for single-component electrolytes. At the same time, problems of concentrational polarization in multicomponent electrolytes, which are most frequently encountered in practice, have been developed inadequately as yet, even though there are investigations devoted to these problems [44, 45].

We shall briefly consider problems dealing with the mathematical description of the phenomena of mass transfer in EMS. It includes the description of the phenomena of transfer in each of the phases (in the solution and the membrane) and of interactions at the boundary and initial conditions (for unsteady-state processes), i.e., a rigorous statement of the problems of mass transfer in EMS requires the examination of conjugate problems. The mathematical description of processes of electro- and mass transfer in EMS is based on the Nernst-Planck equation [28, 29]. For describing mass transfer in a liquid phase, two approaches have gained acceptance [13].

The first approach, implemented in works [46-52], is distinguished by the analysis of mass transfer in EMS consisting of a pair of AEM and CEM and a solution moving between them (Fig. 5b). An electric current of density i passes through the system perpendicularly to the membranes. To simplify the description, a number of assumptions are made: the equivalence of all the chambers of the apparatus; the absence of solvent (water) flow; the one-dimensionality of the flow; a certain (most commonly, quadratic) law of velocity change between the membranes; flow isothermicity.

As basic equations for describing the flow of ions of the j-th species use is made of the generalized Nernst-Planck equation and equations of electroneutrality and current density

$$\mathrm{N} = -D_j(\mathrm{grad}\, C_j + z_jC_j\,\mathrm{grad}\,\varphi) + C_j\mathbf{W}; \tag{2}$$

$$\sum_j z_jC_j = 0; \tag{3}$$

$$i = F\sum_j z_jN_j. \tag{4}$$

Equations (2)-(4) are transformed to an equation of convective diffusion, generally with boundary conditions of the second kind, which is then solved by numerical methods (with the most common choice being a square law of velocity profile distribution [46-48, 50, 51], but other relations may also be employed [49]).

The second approach, successfully used in [52-55], is characterized by the fact that the mass transfer phenomena are considered in diffusional layers of definite thickness which depend on the flow hydrodynamics and which are adjacent to the membrane (Fig. 5c). In this case, the basic equation used is also the Nernst-Planck equation with boundary conditions of the first (from the side of the flow core) and second (from the side of the membrane) kinds.

The second approach offers the possibility of considering a wider range of problems than the first one.

The description of electro- and mass transfer in ion-exchange membranes is based either on the homogeneous or heterogeneous model of the structure of membranes.

The homogeneous model [7] presupposes a homogeneous structure for the membrane. The heterogeneous model is based on a structurally inhomogeneous composition for the membrane. It is assumed in this model that the membrane consists of several phases (most often two: a gel phase and an intergel solution).

When the homogeneous model is employed for describing the phenomena of electro- and mass transfer in a membrane, use is made of the Nernst-Planck equation (very often neglect of the convective component) supplemented with an equation of electroneutrality (in the given case it is written as $\sum z_j\overline{C}_j + \omega\overline{Q} = 0$, where $\overline{Q}$ is the concentration of fixed ions in the membrane; ω is the charge sign of the fixed ions ($\omega = -1$ for cationites and $\omega = 1$ for anionites) and current density equation.

When the heterogeneous model of the structure of ion-exchange membranes is used for describing electro- and mass transfer, the authors of [56-58] suggest the following equation:

$$N_j = -L_j^* \left(RT \frac{d \ln C_j}{dx} + z_j F \frac{d\varphi}{dx} \right) = -L_j^* \left(RT \frac{d \ln \bar{C}_j}{dx} + z_j F \frac{d\bar{\varphi}}{dx} \right), \tag{5}$$

where L_j^* is the effective kinetic coefficient of a component in the heterogeneous membrane

$$L_j^* = [f_1 \bar{L}_j^{\alpha} + (1 - f_1) L_j^{\alpha}]^{\frac{1}{\alpha}}; \tag{6}$$

$\bar{L}_j = \bar{D}_j \bar{C}_j/RT$; $L_j = D_j C_j/RT$; f_1 is the volumetric fraction of the gel phase; α is a parameter taking into account the mutual arrangement of phases.

At the same time, the description of mass transfer phenomena in membranes used in electroosmofiltration (ultrafiltrational, reverse-osmotic) is the concern of a much smaller number of works [5, 6, 59]. In [59], a system of differential equations of transfer is described for baromembrane processes when an electric field is imposed on them, i.e., for electroosmofiltrational processes.

To couple the equations of electro- and mass transfer in the liquid and membrane phases, use is made of the equations of equilibrium for the membrane-solution phase boundary. The most common forms of the equilibrium equation for ion-exchange membranes were treated in [53-58, 60] and for reverse-osmotic and ultrafiltrational membranes in [61].

The approaches considered above allowed many authors [44, 46-58, 60] to pose and solve many general and specific problems of electro- and mass transfer in EMS (to a large extent, applicable to systems with ion-exchange membranes). For example, the solutions were obtained and explanations were furnished for the mechanisms underlying the distribution of concentrations and current densities along the length and width of the intermembrane channel, and the volt-ampere characteristics, limiting currents, and diffusional layers were calculated.

On the basis of the approaches described, mathematical models of separate types of electrodialyzing apparatus were developed in [62-65].

In conclusion, noting the indubitable usefulness of these results for understanding the mechanism of the processes of electro- and mass transfer and for evaluating its individual characteristics, let us also note that because of certain limitations imposed on the mathematical models (the absence of solvent flow, isothermicity, stationarity, and so on), it is rather difficult to apply them to engineering calculations of multichamber electromembrane facilities, especially those operating under unsteady-state or nonisothermal conditions.

From the viewpoint of engineering calculations of electromembrane processes, a mathematical model of an electrodialyzing facility operating in the continuous flow regime of desalination suggested in [66] is of interest. However, due to a number of assumptions made when posing the problem (isothermicity of the process, independence of the resistance of membranes of the concentration, etc.), it is also very difficult to extend this model to the electromembrane facilities we are considering. Nevertheless, the general approach to the mathematical simulation of electrodialyzing processes suggested in this work seems to be quite reasonable and suitable for practical calculations.

Let us consider the phenomena and processes of heat transfer in EMS. A much smaller number of research workers have been concerned with the study of heat transfer in EMS than of mass transfer. Problems of heat transfer were treated in the works of N. M. Smirnova, A. M. Kuzavskiy, V. P. Shulik, V. A. Shaposhnik, N. P. Gnusin, et al. [32, 67-74]. However, there are no systematic or completed investigations along these lines although their importance is self-evident, since an increase in the temperature of solutions and membranes even by several degrees may alter the mass transfer characteristics substantially, having in turn an effect on both the speed of the process as a whole and its individual characteristics. Thus, for exameple, A. F. Mazanko et al. [12] note that at elevated temperatures in membrane electrolysis there is an increase in both the diffusion of chlorine ions (by about 5% per deg C) and the current efficiency. Problems of heat transfer in electrochemical systems were considered in [75, 76], and specifically in electrodialysis in [32, 67-74]; the present authors are unaware of publications dealing with heat transfer in electroosmofiltrational processes.

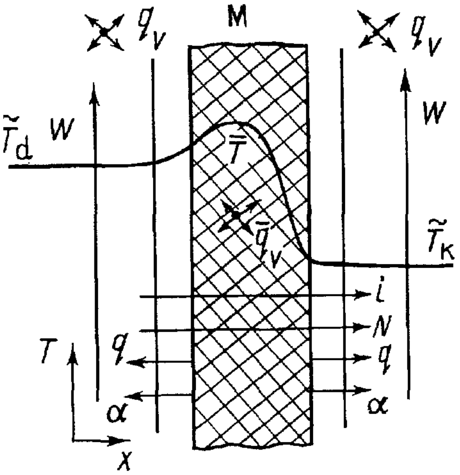

Fig. 6. Schematic of heat transfer in an EMS.

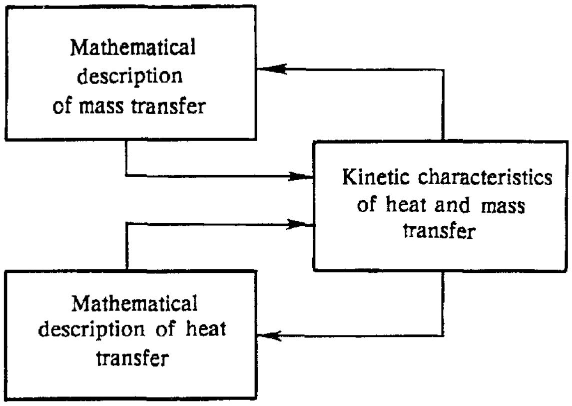

Fig. 7. Interrelation between mass/heat transfer and the kinetic characteristics of the process.

The mechanism underlying the heating of solutions and membranes can be presented in the following way (Fig. 6). When an electric current of density i is passed through solutions and membranes, Joule heat is released. The quantity of heat released may vary because of different electric conductivities (different chambers may contain solutions with different concentrations; in the case of a large number of series-connected chambers and at large current densities this difference can be appreciable and, consequently, the electric conductivity of the solutions and membranes may differ considerably from that for those located in other chambers of the apparatus), and therefore the temperatures of the solutions and membranes will differ in every chamber and, consequently, heat exchange between the solutions and membranes will take place. Moreover, it should be borne in mind that, flowing from chamber to chamber, the solution will be heated repeatedly and will repeatedly interact (in the sense of heat transfer) with the membranes. When heat transfer phenomena are described, it is most often assumed (for the purpose of simplification) that there are uniformly distributed volumetric heat sources of power q_V and $\bar{q}_V$ in solutions and membranes, respectively equal to

$$q_V = (i\eta)^2/\varkappa; \quad \bar{q}_V = (i\eta)^2/\bar{\varkappa}, \tag{7}$$

where η is a coefficient taking into account the decrease in the current efficiency due to current leaks [66, 72]. (Strictly speaking, volumetric heat sources are distributed nonuniformly in membranes and solutions because of the nonuniform distribution of concentrations in solutions (especially in diffusional layers) and membranes; it would be advisable to take into account this fact when considering the phenomena of heat and mass transfer in EMS and their mathematical description.) Next, generally, the problem of heat conduction in a membrane is considered under boundary conditions of the third kind and the influence of temperature on the individual char-

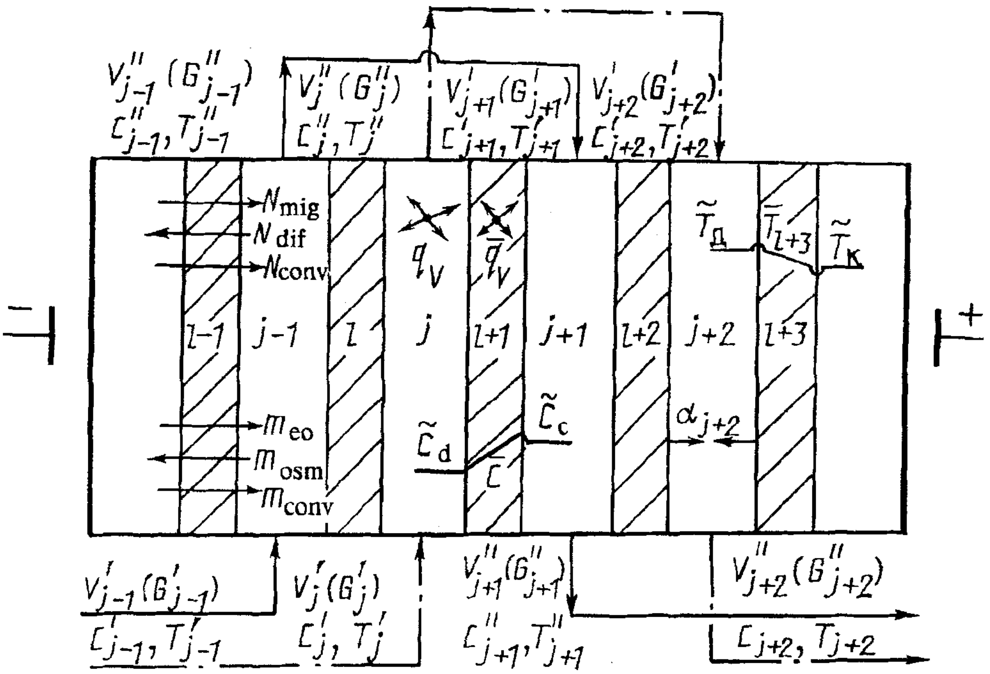

Fig. 8. Schematic of mass and heat transfer in a multichamber electromembrane apparatus.

acteristics of electrodialysis is analyzed. In our opinion, it seems reasonable to consider precisely the interrelation between the kinetics of mass transfer and the kinetics of heating of EMS. However, consideration of the interrelation between the phenomena of mass and heat transfer at the level of conjugate equations of electro-, heat, and mass transfer [59, 75] is rather a challenge because of the unwieldiness of these equations. Therefore it seems appropriate to "decouple" these equations and consider the kinetics of mass and heat transfer separately and then to correct them with allowance for the change in the kinetic characteristics as a functrion of the concentration and temperature (Fig. 7). We considered such an approach in [74] when we developed a mathematical model of mass and heat transfer in an electrodialyzing facility with closed circulation of solutions along the dialyzate and concentrate circuits with series connection of the chambers along them (see Fig. 1). The scheme of mass and heat transfer for such EMS's is given in Fig. 8.

The description of mass transfer is based on the following assumptions: 1) there is equilibrium of phases at the solution-membrane boundary; 2) there is intense mixing of solutions in the working chambers (no concentration gradients along the chamber width); 3) mass fluxes are quasistationary. It is further assumed that mass transfer through a membrane is composed (Fig. 8) of fluxes of dissolved substance N and solvent (water) m. The flux of the dissolved substance flux is composed of two components: migrational N_{mig} and diffusional N_{dif} (the convective component is ignored):

$$N = N_{mig} + N_{dif}; \tag{8}$$

$$N_{mig} = \bar{t} i S \eta / F; \tag{9}$$

$$N_{dif} = P_d (\tilde{C}_c - \tilde{C}_d) S/X. \tag{10}$$

The solvent (water) flux is composed of three components (see Fig. 5a): electroosmotic m_{eo}, osmotic m_{osm}, convective (hydraulic) m_{conv}:

$$m = m_{eo} + m_{osm} + m_{conv}; \tag{11}$$

$$m_{eo} = \beta i S \eta; \tag{12}$$

$$m_{\mathrm{osm}} = D_W (\tilde{C}_{\mathrm{c}} - \tilde{C}_{\mathrm{d}})\, S/X; \tag{13}$$

$$m_{\mathrm{conv}} = k \Delta P S. \tag{14}$$

Besides the above components, one can also take into account the fluxes of H_3O^+ and OH^- ions in electro- and mass transfer (especially in the regions near and beyond the limit).

By writing the mass balance for the dissolved substance and the solvent for an arbitrary chamber of an electromembrane apparatus and assuming that the mean concentration of the dissolved substance in the chamber is equal to the arithmetic mean one at the entrance and exit, we can obtain equations that would describe the variation in the concentrations and volumetric flow rates in arbitrary chambers of the apparatus.

For example, for multichamber electrodialyzers the following equations were obtained for the j-th chamber:

$$\begin{gathered} \frac{V_{\mathrm{c}}}{2}\left(\frac{dC_j''}{d\tau} + \frac{dC_j'}{d\tau}\right) = \pm E_j + K_l C_{j\pm 1}'' + K_{l+1} C_{j\pm 1}'' - \\ - (K_{\mathrm{s}j} + V_j'')\, C_j'' + K_{l+1} C_{j\pm 1}' + K_l C_{j\pm 1}' - (K_{\mathrm{s}j} - V_j')\, C_j'; \end{gathered} \tag{15}$$

$$\begin{gathered} -W_l C_{j\pm 1}'' - W_{l+1} C_{j\pm 1}'' + W_{\mathrm{s}j} C_j'' - W_l C_{j\pm 1}' - W_{l+1} C_{j\pm 1}' + \\ + W_{\mathrm{s}j} C_j' \pm H_j + V_j' - V_j'' = 0, \end{gathered} \tag{16}$$

where E_j, K_l, K_s, H_j, W_l, and W_{sj} are coefficients composed of the coefficients of electro- and mass transfer $\bar{t}$, P_d, β, D_W, etc. [74].

Next, the equations of mass balance for the dissolved substance and the solvent are written for all the intermediate tanks available in the scheme (Figs. 1 and 2) (with allowance for the scheme of their connection with the electromembrane apparatus):

$$V_{\mathrm{E}_1} \frac{dC_1''}{d\tau} = V_n'' (C_n'' - C_1'); \tag{17}$$

$$\frac{dV_{\mathrm{E}_1}}{d\tau} = V_n'' - V_n'. \tag{18}$$

And finally, the equality of the concentrations and volumetric flow rates at the exit from the preceding chamber and the entrance to the next one is written down:

$$C_j' = C_{j-2}''; \tag{19}$$

$$V_j' = V_{j-2}''. \tag{20}$$

Thus, we obtain a closed system of differential-algebraic equations consisting of n + 2p (n is the number of chambers, p is the number of intermediate tanks) equations which describe the time dependence of concentrations and flow rates in the chambers of the apparatus and of the volumes of solutions in the intermediate tanks.

Description of Heat Transfer. The main features of heat transfer in an EMS were considered earlier. Here, we shall set out briefly the procedure used to calculate the thermal regime of an electromembrane facility. The essence of it is as follows. A heat balance is set up for an arbitrary chamber of the apparatus. The overall quantity of heat acquired by the solution in the j-th chamber is

$$\begin{gathered} Q_j = \left[\alpha (\bar{T}_l^{\mathrm{s}} - \tilde{T}) + \alpha (\bar{T}_{l+1}^{\mathrm{s}} - \tilde{T}) + \frac{(i\eta)^2 \delta}{\varkappa_j}\right] S = \\ = Gc (T_j'' - T_j') + \rho c \delta \frac{dT_j}{d\tau} S. \end{gathered} \tag{21}$$

Then, to determine the surface temperature of the membranes $\overline{\text{T}}_l^s$, the nonstationary heat conduction equation is considered with nongradient initial conditions and boundary conditions of the third kind.

By averaging this equation [74], we obtain an expression for the mean temperature of the membrane

$$\bar{\bar{T}}_l = T_0 e^{-a\tau} + b e^{-a\tau} \int_0^\tau (\tilde{T}_{j-1} + d\tilde{T}_j) e^{a\tau} d\tau + \frac{(i\eta)^2 (1 - e^{-a\tau})}{\overline{ac\rho\varkappa}}, \tag{22}$$

where a, b, and d are coefficients representing relationships between the heat transfer coefficients and the thermophysical coefficients of the membrane [74].

Having substituted the values for the mean temperature of the membrane (22) into the balance equation (21), we shall write an equation for finding the temperature of the solution in a chamber:

$$\begin{gathered} \sigma_{1j} \sum_{k=0}^{1} b_{l+k} e^{-a_{l+k}\tau} \int_0^\tau (T''_{j-1+k} + T'_{j-1+k} + d_{l+k} T''_{j+k} + d_{l+k} T'_{j+k}) e^{a_{l+k}\tau} d\tau - \\ - \frac{dT''_j}{d\tau} - \frac{dT'_j}{d\tau} - \sigma_{2j} T''_j - \sigma_{3j} T'_j = \sigma_{4j}, \end{gathered} \tag{23}$$

where σ is a coefficient composed of thermophysical characteristics and time functions [74].

Next, we write equations of heat balance for all intermediate tanks (see Fig. 1) in the form

$$V_{E_1} \frac{dT''_1}{d\tau} = V''_n \left(\frac{\rho_n c_n}{\rho_1 c_1} T''_n - T'_1 \right) \tag{24}$$

as well as equations for the equality of the temperatures at the exit from the preceding chamber and the entrance to the next one

$$T'_j = T''_{j-2}. \tag{25}$$

Thus, we obtain a closed system of n + 1 integrodifferential equations to determine the dependence of the temperatures of the solutions in the chambers and intermediate tanks on time.

Using a similar approach, we developed a mathematical model of mass transfer for a multichamber electroosmofiltrational facility (Fig. 2). But in the equations for the mass flux of dissolved material we took into account one other component, namely, the transfer of mass through a membrane due to the hydraulic permeability. Therefore, the equations constituting the mathematical model of mass transfer in an electroosmofiltrational facility also involve such characteristics as the membrane selectivity, the specific capacity of the membranes and the osmotic pressure of the solutions.

A mathematical model of thermal phenomena in an electroosmofiltrational facility was constructed on the basis of heat balance equations for a separate cell, the whole apparatus, and an intermediate tank. As a result, a system of ordinary differential equations was obtained for determining the temperatures of solutions in intermediate tanks and at the exit from the apparatus at an arbitrary time.

In order to be able to realize the mathematical models numerically, it is necessary to know various kinetic characteristics and their dependence on the operational parameters of the process. Let us consider the available information on the most important kinetic characteristics.

The transference numbers in membranes depend on the nature of the electrolyte and the membranes, the concentration of the external solution, and the temperature. Generally these are determined experimentally. Data on transference numbers can be found in [77-84]. As the concentration of the solutions increases, the transference numbers decrease [77, 81, 84]; they also decrease with increase in the current density [80, 83] and the solution temperature [79]. The values of the transference numbers depend to a great extent on the presence of competing ions [78, 82].

Duffusional, electroosmotic, osmotic, and hydraulic permeabilities. Diffusional permeability was considered in [70, 85-90]. There, its dependence on concentration was noted (in some cases it increases with increase in concentration, and in others it decreases). As the temperature increases, the diffusional permeability increases exponentially [70, 89, 90].

In [70, 87, 88], empirical dependences of diffusional permeability on concentration are given, and in [70, 89, 90] also its dependence on the temperature of solutions. These relations have the form

$$P_{\mathrm{d}} = aC^n \exp\left(-\frac{A}{T}\right), \tag{26}$$

where a, n, and A are experimental coefficients.

The problems of the electroosmotic permeability of water in ion-exchange membranes have evoked a rather large number of publications [91-97]. N. P. Gnusin, N. P. Berezina , et al. made important contributions to the study of electroosmotic permeability and its dependence on various factors [92-96]. On the basis of the heterogeneous structure of membranes, they derived formulas to calculate the transference numbers of water. From these formulae one can easily calculate the coefficients of electroosmotic permeability. They also showed that the transference numbers of water depended on the physicochemical properties of a polymer material and moisture capacity.

The electroosmotic permeability is independent of current density [91, 96] and is a rather complicated function of the solution concentration. For heterogeneous and some types of homogeneous membranes, the electroosmotic permeability first remains constant and then falls as the concentration of the external electrolyte grows [91, 96]. For other types of homogeneous membranes the electroosmotic permeability of solutions falls with rise in their concentration [91, 96].

Unfortunately we failed to locate information concerning the effect of temperature on the electroosmotic permeability of membranes.

The osmotic permeability of membranes was considered in [3, 87, 88, 90]. As the concentration of solutions rises, the osmotic permeability falls. For heterogeneous membranes this dependence is close to a linear one [87] and for homogeneous membranes the authors of [88] suggest the following relation for its description:

$$D_W = a - b \ln C, \tag{27}$$

where a and b are empirical coefficients. That work also contains data on the dependence of the osmotic permeability on temperature. It has the form

$$D_W = A \exp(-E/RT). \tag{28}$$

Most often the hydraulic permeability is termed simply permeability. For reverse-osmotic and ultrafiltrational membranes permeability problems have been rather well studied (for example, [4-7, 10, 14, 20]).

There is little information on the permeability of ion-exchange membranes. We will only mention the data given in [8]. It is noted there that the (hydraulic) permeability of homogeneous membranes falls linearly with increase in the concentration of solutions, whereas for heterogeneous membranes, conversely, the permeability increases with increase in the concentration of solutions. No information is given concerning the dependence of permeability in ion-exchange membranes on temperature.

The electrical conductivity of solutions and membranes. The electrical conductivity of solutions has been investigated sufficiently well. In numerous monographs [98, 99], handbooks [85, 100], and reviews [102] one can find various information on the electrical conductivity of solutions, including also its dependence on concentrations, temperatures, pressures, etc. Therefore, we shall not consider this problem in detail.

The electrical conductivity of membranes has also been investigated by various research workers and can be found in numerous works [3, 8, 103-109] but, obviously, in a much smaller volume than the electrical conductivity of solutions. In some works [103, 105], theoretical problems of the electrical conductivity of ionites and ionite membranes are considered, and in others [104, 106-109] specific data on the electrical conductivity of membranes is given.

As the concentration of the external solution rises, the electrical conductivity of membranes increases [104, 106]; with a rise in temperature, it increases exponentially [109].

Thermophysical characteristics of solutions and membranes. The required information on the thermophysical properties of solutions (ρ, c, ϑ) can be rather easily found in reference books [101, 110]. However, there are relatively few studies on the thermophysical properties of membranes [32], and it does not appear possible to say anything definite about their dependence on various parameters.

Heat transfer coefficients are calculated from ordinary dimensionless equations presented in numerous references (for example, in [111]).

In concluding this work we wish to note that numerical realization of the proposed mathematical models of the processes of electrodialysis and electroosmofiltration demonstrated their adequacy to real processes. At the same time, for numerical realization of mathematical models it is necessary to have a large choice of reliable kinetic characteristics and to know their dependence on the operational parameters of the process.

NOTATION

C, concentration; T, temperature; φ, electric potential; i, current density; ΔP, pressure drop on membrane; W, velocity of solution; z, number of elementary charges; N, m, fluxes of dissolved substance and solvent; V, G, volumetric and mass flow rates of solution; V_E, volume of solution in intermediate tank; x, τ, coordinate and time; X, S, thickness and area of membrane; δ, intermembrane distance; D, coefficient of diffusion; P_d, β, D_W, k, diffusional, electroosmotic, osmotic, and hydraulic permeabilities; t, transference number; κ, electrical conductivity; ρ, ν, c, density, kinematic viscosity and heat capacity; α, heat transfer coefficient; R, F, universal constant and Faraday number. Subscripts: j, ion species, number of arbitrary chamber; n, last chamber; l, membrane number; d, c, dialyzate, concentrate; s, membrane surface; RO, reverse osmosis; UF, ultrafiltration; ED, electrodialysis; $'$, $''$, value of parameter at entrance and exit; $-$, $\sim$ association of parameter with membrane, mean value.

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