FTD-TT-63-722

64-5

TRANSLATION

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By

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FOREIGN TECHNOLOGY
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UNEDITED ROUGH DRAFT TRANSLATION

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English Pages: 13

SOURCE: Russian Periodical, Inzhenerno-Fizicheskiy
Zhurnal, Vol. 6, Nr. 2, 1963, pp 60-68

S/170-63-6-2

	PREPARED BY: TRANSLATION DIVISION FOREIGN TECHNOLOGY DIVISION WP-AFB, OHIO.

Date 22 July 19 63

Performance of Porous Electrodes in a Diffusion Condition of Feeding Reagents

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I. G. Gurevich and V. S. Bagotskiy

Under consideration is a problem concerning the distribution of the electochemical process by the depth of a porous electrode of finite thickness functioning in a diffusion condition iwth consideration of electrotechnical polarization and ohmic losses. Given is a general solution in closed form.

The use in applied electrochemistry of porous electrodes, having developed internal surface, is connected with the desire for maximum intensification of electrode processes and for the obtainment of maximum specific characteristics.

In this connection of greater importance is the problem of effectiveness of employing such electrodes, due to the macrokinetic nature of the electrode process, leading to nonequilibrium distribution of its intensity over the volume of the electrode.

In a majority of published reports (1-16), directly or indrectly pertaining to the given problem, the problem of distribution of electrode process intensity over the volume of the electrode is solved (with these or any other approximations) analytically, on the basis of the general theory of field distribution in an electrolytic cell (the Poisson equation is used), with consideration of mass transfer process [13-15] or without such a consideration [4-10]. In reports [11-12] the electrode is considered equipotential and intensity distribution of the electrochemical process over the depth of the electrode is bound only with the mass transfer process. In some reports [1-3, 16] intensity distribution of the process in the electrode is investigated with the replacing of the electrode with an equivalent electric system, allowing to reduce the problem to the regaining of currents in elements of this system; the shortcomings of such a method lie in the difficulties of modeling transportation processes, which under conditions of the discussed problem are described (shown below) by nonlinear equations [3].

In this report in contrast to the report by O. S. Ksenzhek [10,12] and L.G. [illegible] in [11] the problem concerning the function of a porous electrode in a diffusion condition is considered in a more general arrangement: all kinds of losses are taken into consideration (polarization and ohmic) for an electrode of finite thickness.

We will discuss the following system (fig.1). The porous electrode with thickness L is submerged in an electrolyte chamber. In this report is discussed a case, when in the role of reagents, on one hand appear electroneutral molecules, and on the other hand - corresponding electrolyte ions. It is assumed, that the transfer of electroneutral molecules of the reagent and product in the examined system electrode-electrolytic chamber is realized only by the way of molecular diffusion, and the transfer of electrolyte ions - by diffusion and migration in the electric field of the electrode.

It is further assumed, that the concentration of ion components of the working mixture, filling the electrode, considerably exceeds the concentration of electroneutral reagents and product. This condition allows to consider the conductivity of the mixture as constant and, in addition, to disregard the diffusion potential, forming on account of nonuniform distribution of concentrations of ion components.

Secured are conditions of maintaining constant volumetric (in electrolytic chamber at a certain distance from the electrode) concentrations of the reagent c_p^v and of the product c_p^v or on account of chamber dimensions, or on account of continuous feeding into it the reagent and removal of the forming product.

In stationary state into the electrode from the electrolytic chamber is passed a quantity of reagents equivalent to the current magnitude I (amperage), and from the electrode is removed a corresponding amount of products forming during the reaction.

In this report when examining the transportation stage of the electrode process into consideration are taken streams of electroneutral substances (reagent or product)

$$\overline{q_j} = -D_j' \,\mathrm{qrad}\, c_j. \qquad (1)$$

Assuming in the electrode takes place a reaction of the type

$$\nu_p P + \ldots \rightarrow \nu_n \Pi + \ldots + ne. \qquad (2)$$

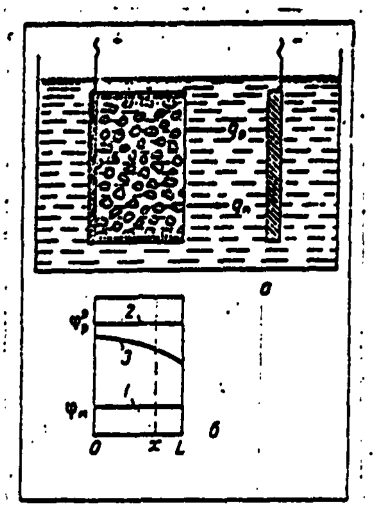

Fig. 1, a-scheme of electrolytic cell; b-distribution of potentials in depth of porous electrode metallic skeleton of the electrode (1) working solution of electrolyte in the absence of current (2) and at current (3).

Assuming the visible surface of the electrode is sufficiently large, in order to disregard the boundary effects, and the structure as sufficiently finely porous, in order to gain the possibility of getting away from a concrete structure and consider the electrochemical reaction taking place over the entire volume of the electrode (as it was made for the first time by Ya.B.Zel'dovich [17]), it is possible to begin deliberating a monodimensional problem.

Since the distribution of intensity of the electrode process (true current density i) over the depth of the porous electrode is connected, on one hand, with distribution of reagent and product concentrations, and on the other - with the distribution of the potential, then we arrive at a system of two equations, describing the performance of the electrode: it is evident from the Fick equation

$$c_p'' = \frac{\nu_p s}{nFD_p} i, \qquad (3)$$

the second - Poisson equation

$$\eta'' = sR_\rho i. \qquad (4)$$

Before we begin analyzing the systems of equations (3),(4) it is advisable to examine the laws (validities) of employing the Poisson equation in form (4), or, generally speaking, it describes the distribution of solution potential.

Disregarding the resistance of the metallic skeleton in comparison with the effective resistance of the mixture in pores of the electrode R_3 =const, we obtain an independent from coordinate x course of the metal potential φ_M (straight line 1, fig. 1,b); if in the absence of current the potential of the solution (equilibrium) φ_p^p is also independent from x (straight line 2), then under load φ_p=f (x) (curve 3).

The jump of the potential on the surface of the electrode in the absence of current

$$\Delta\varphi^p = \varphi_M - \varphi_p^p. \qquad (5)$$

and the sudden change of the potential for the electrode under load

$$\Delta\varphi = \varphi_M - \varphi_p. \qquad (6)$$

The polarization of the electrode

$$\eta = \Delta\varphi^p - \Delta\varphi = \varphi_p - \varphi_p^p. \qquad (7)$$

Differentiating (7), we obtain

$$\frac{\partial\eta}{\partial x} = \frac{\partial\varphi_p}{\partial x}. \qquad (8)$$

From equation (8) follows the rule of using the Poisson equation in form (4) whereby the condition of such a validity lies in the fact, that the conductivity of the metallic skeleton should considerably exceed the conductivity of the working mixture.

For kinetic characteristic of the process (i) we accept the expression from the theory of the slowed down process

$$i = j_0\left\{\frac{c_p^x}{c_p^v}\exp\left[\frac{\alpha F}{RT}\eta\right] - \frac{c_n^x}{c_n^v}\exp\left[-\frac{(n-\alpha)F}{RT}\eta\right]\right\}. \qquad (9)$$

In order that the system of equations (3),(4) should be complete, we will find the relationship between local values of product and reagent concentrations, which should enable to eliminate from equation (9) the concentration of the product.

Writing down the Fick equation (1) twice - once for the reagent and the second time for the product - and expressing the stream of the substance through electric values and reactions coefficients (2), we will obtain respectively

$$\frac{dc_p}{dx} = \frac{\nu_p}{nFD'_p} I_x, \qquad (10)$$

$$\frac{dc_n}{dx} = -\frac{\nu_n}{nFD'_n} I_x. \qquad (11)$$

Excluding from (10) and (11) I_x, we obtain after integration the interesting relation

$$c_n^x = c_n^L + \frac{\nu_n}{\nu_p}\frac{D'_p}{D'_n}(c_p^L - c_p^x). \qquad (12)$$

In order to change over in equation (12) from unknown concentration values on the frontal (with respect to the counter electrode, i.e. at x=L) surface of the electrode c_r^L and c_p^L to given volumetric concentrations c_r^V and c_p^V, we will discuss the conditions of reagent diffusion from the electrolytic chamber to the electrode. Assuming the distribution of reagent and product concentrations in the diffusion layer at the frontal surface of the electrode as linear, we obtain the following expression for current per unit of visible surface of the electrode:

$$I = \frac{nFD_p}{\nu_p}\frac{(c_p^v - c_p^L)}{\delta}. \qquad (13)$$

Using then the expression for the ultimate current

$$I_{\text{пред}} = \frac{nFD_p}{\nu_p \delta} c_p^v, \qquad (14)$$

we obtain $C_r^l = \frac{}{nFD_r} I_{forw} (1-\theta)$, where $\theta = \frac{I}{I_{forw}}$ -coefficient of "charging" the electrode ($0 \leqslant \theta \leqslant 1$). Finally

$$c_p^L = c_p^v(1-\Theta). \qquad (15)$$

Analogously we obtain

$$c_n^L = c_n^v + \frac{\nu_n}{\nu_p}\frac{D_p}{D_n}c_p^v\Theta. \qquad (16)$$

With consideration of equation (12), (15) and (16) equation (9) can be rewritten in following form:

$$i = j_0\left\{\frac{c_p}{c_p^v}\exp\left[\frac{\alpha F}{RT}\eta\right] - \left[1 + \frac{c_p^v}{c_n^v}\frac{\nu_n}{\nu_p}\frac{D'_p}{D'_n} + \right.\right.$$

$$\left.\left. + \frac{c_p^v}{c_n^v}\left(\frac{D_p}{D_n} - \frac{D'_p}{D'_n}\right)\frac{\nu_n}{\nu_p}\Theta - \frac{c_p}{c_n^v}\frac{\nu_n}{\nu_p}\frac{D'_p}{D'_n}\right]\exp\left[-\frac{(n-\alpha)F}{RT}\eta\right]\right\}. \qquad (9')$$

We will introduce the following designations: $\nu = \frac{p}{r}$; $xi = \frac{c_p^v}{c_r^v}$; $d_{r.p} = \frac{D_r}{D_p}$;

$_{r.p} = \frac{D'_r}{D_p}$; $d_f = \frac{D'_j}{D_j}$ (for many porous electrodes d = 10^{-1} -10^{-2}[12]). If the lectrode weakens diffusion only on account of its structure regardless of the operties of the diffused substance, then it is apparent, that $d_r = d_p$, i.e. $\frac{D'_r}{D_r} = \frac{D'_p}{D_p}$ The expression for i will then be simplified:

$$i = j_0 \left\{ \frac{c_p}{c_p^v} \exp\left[\frac{\alpha F}{RT} \eta \right] - \left(1 + \frac{\nu d'_{p.n}}{\xi} - \frac{\nu d'_{p.n}}{\xi} \frac{c_p}{c_p^v} \right) \times \exp\left[-\frac{(n-\alpha) F}{RT} \eta \right] \right\} \tag{17}$$

and the system of equations (3), (4) will be rewritten:

$$c_p'' = \frac{\nu_p s j_0}{nFD'_p} \left\{ \frac{c_p}{c_p^v} \exp\left[\frac{\alpha F}{RT} \eta \right] - \left(1 + \frac{\nu d'_{p.n}}{\xi} - \frac{\nu d'_{p.n}}{\xi} \frac{c_p}{c_p^v} \right) \exp\left[-\frac{(n-\alpha) F}{RT} \eta \right] \right\}.$$
$$\eta'' = sR_s j_0 \left\{ \frac{c_p}{c_p^v} \exp\left[\frac{\alpha F}{RT} \eta \right] - \left(1 + \frac{\nu d'_{p.n}}{\xi} - \frac{\nu d'_{p.n}}{\xi} \frac{c_p}{c_p^v} \right) \exp\left[-\frac{(n-\alpha) F}{RT} \eta \right] \right\}. \tag{18}$$

Taking into consideration, that by the conditions of the investigated problem (fig. 1a) the flow of the substance and the current through the rear surface of the electrode equal zero, and through the frontal one they are respectively equal $\frac{\nu_r}{nF}$ I and I, we obtain boundary conditions of following form:

$$\eta'|_{x=0} = 0,\ \eta'|_{x=L} = R_s I;$$
$$c'_p|_{x=0} = 0,\ c'_p|_{x=L} = \frac{\nu_p}{nFD'_p} I. \tag{19}$$

System (18) appears to be a system of two nonlinear differential equations of second order.

We shall rewrite the system of equations (18) and boundary conditions (19), changing preliminarily to dimensionless coordinate from condition zeta = $\frac{x}{L}$ ($0 \leqslant$ zeta $\leqslant 1$), dimensionless concentration from condition $c = \frac{C_r}{C_r}$ ($0 \leqslant c \leqslant 1$) and to dimensionless polarization from condition $u = \frac{1}{R_{з}\ LI} \eta$.

Then

$$c'' = A\psi\left\{c\exp\left[\frac{\theta}{\Omega_{forw}}u\right] - (E - Mc)\exp\left[-\frac{(n-\alpha)\theta}{\alpha\Omega_{forw}}u\right]\right\},$$

$$u'' = \frac{A}{\theta}\left\{c\exp\left[\frac{\theta}{\Omega_{forw}}u\right] - (E - Mc)\exp\left[-\frac{(n-\alpha)}{\alpha}\frac{\theta}{\Omega_{forw}}u\right]\right\} \qquad (18')$$

and the boundary conditions

$$u'|_{\zeta=0} = 0,\ u'|_{\zeta=1} = 1;$$
$$c'|_{\zeta=0} = 0,\ c'|_{\zeta=1} = \psi\theta, \qquad (19')$$

where

$$A = \frac{I_0}{I_{forw}};\ I_0 = sLj_0;\ \ \psi = \frac{D_p/\beta}{D_p'/L};$$
$$\Omega_{forw} = \frac{1}{R_s L}\frac{RT/\alpha F}{I_{forw}};\ E = 1 + \frac{\nu d_{p.n}'}{\xi};\ M = \frac{\nu d_{p.n}'}{\xi}. \qquad (19a)$$

Making the first equation of system (18') into the second one, we obtain

$$\frac{c''}{u''} = \psi\theta. \qquad (20)$$

After double integration (20) with consideration of boundary conditions at zeta = 1 we have

$$u = \frac{c}{K} + C_2. \qquad (21)$$

where K = $\psi\theta$, and C_2 - constant of second integration.

Substituting (21) in first equation of the system (18'), we will obtain

$$c'' = A\psi\left\{K_1 c\exp(K_2 c) - (E - Mc)\,K_1^{\frac{\alpha-n}{\alpha}}\exp\left[\frac{\alpha-n}{\alpha}K_2 c\right]\right\}, \qquad (22)$$

where K_1 = exp $(C_2\theta/\Omega_{forw})$; $K_2 = \frac{1}{\Omega_{forw}}\psi$

We will change the variables in accordance with

$$c = \ln z. \qquad (23)$$

Then equation (22) will be rewritten

$$0 = (z)\,{}^{2}\!f + {}_{1}z\,(z)\,{}^{1}\!f - {}_{2}z \qquad (24)$$

with boundary conditions $z'|_{\zeta=0} = 0;\ z'|_{\zeta=1} = Kz_1\ \ (z_1 = z|_{\zeta=1})$. (24a)

Here

$$f_1(z) = \frac{1}{z}; \qquad (24b)$$

$$f_2(z) = A\psi\left\{-K_1\ln z\,z^{K_2+1} + (E - M\ln z)\,K_1^{\frac{\alpha-n}{\alpha}}z^{\frac{\alpha-n}{\alpha}K_2+1}\right\}. \qquad (24c)$$

The substitution of p(z) = z' (ζ) allows to reduce the order of equation (24) and bring it into form of the Bernoulli equation

$$p' - f_1(z)p + f_2(z)p^{-1} = 0. \qquad (24')$$

The boundary conditions respectively will be written:

$$p(z)|_{z=z_0} = 0; \; p(z)|_{z=z_1} = Kz_1. \qquad (24'a)$$

The following substitution q (z)=p^2 allows to reduce equation (24') to linear equation with variable coefficients

$$q' - 2f_1(z)q + 2f_2(z) = 0. \qquad (24")$$

Integrating (24") by known methods and determining the integration constant from boundary condition at ζ = 1:

$$q|_{z=z_1} = p^2|_{z=z_1} = K^2 z_1^2, \qquad (24"a)$$

we obtain a solution in following form:

$$q = z^2\left\{2A\psi\left\{K_1 z^{K_2}\left[\frac{\ln z}{K_2} - \frac{1}{K_2^2}\right] - EK_1^{\frac{\alpha-n}{\alpha}}\frac{z^{\frac{\alpha-n}{\alpha}K_2}}{\frac{\alpha-n}{\alpha}K_2} + \right.\right.$$
$$\left.\left. + MK_1^{\frac{\alpha-n}{\alpha}} z^{\frac{\alpha-n}{\alpha}K_2}\left[\frac{\ln z}{\frac{\alpha-n}{\alpha}K_2} - \frac{1}{\left(\frac{\alpha-n}{\alpha}K_2\right)^2}\right]\right\} + \right. \qquad (24"b)$$
$$+ K^2 - 2A\psi\left\{K_1 z_1^{K_2}\left[\frac{\ln z_1}{K_2} - \frac{1}{K_2^2}\right] - EK_1^{\frac{\alpha-n}{\alpha}}\frac{z_1^{\frac{\alpha-n}{\alpha}K_2}}{\frac{\alpha-n}{\alpha}K_2} + \right.$$
$$\left.\left. + MK_1^{\frac{\alpha-n}{\alpha}} z_1^{\frac{\alpha-n}{\alpha}K_2}\left[\frac{\ln z_1}{\frac{\alpha-n}{\alpha}K_2} - \frac{1}{\left(\frac{\alpha-n}{\alpha}K_2\right)^2}\right]\right\}\right\}.$$

To determine K_1 (more accurately, the integration constant of equation (20) C_2) included in it, we utilize boundary condition at ζ = 0:$q/_{z0} = 0$.

The solution of the obtained equation relative to K_1 in general form is difficult. For values alpha and n, satisfying the formula $\frac{\text{alpha} - n}{\text{alpha}} = -1$, the equation becomes quadratic, and for K_1 is obtained the following expression:

$$K_1 = Q + \sqrt{Q^2 - B}, \tag{25}$$

where

$$Q = \frac{K^2 K_2^2}{4A\psi}\left(z_1^{K_2}[K_2 \ln z_1 - 1] - z_0^{K_2}[K_2 \ln z_0 - 1]\right)^{-2};$$

$$B = \frac{EK_2\left(\frac{1}{z_1^{K_2}} - \frac{1}{z_0^{K_2}}\right) - M\left\{\frac{1}{z_1^{K_2}}[K_2 \ln z_1 + 1] - \frac{1}{z_0^{K_2}}[K_2 \ln z_0 + 1]\right\}}{z_1^{K_2}[K_2 \ln z_1 - 1] - z_0^{K_2}[K_2 \ln z_0 - 1]} \tag{25a}$$

Hence

$$C_2 = \frac{\Omega f_{forw}}{\theta} \ln\left(Q + \sqrt{Q^2 - B}\right). \tag{26}$$

Making a reverse transformation from variable z to variable c we obtain

$$\zeta = \int_{c_0}^{c}\left\{K^2 + 2A\psi\left[\frac{K_1}{K_2^2}\left(\exp[K_2c][K_2c - 1] - \exp[K_2c_1][K_2c_1 - 1]\right) + \right.\right.$$
$$+ \frac{1}{K_1}\left(\frac{E}{K_2}\left(\exp[-K_2c] - \exp[-K_2c_1]\right) - \frac{M}{K_2^2}\left(\exp[-K_2c] \times \right.\right.$$
$$\left.\left.\left.\left. \times [K_2c + 1] - \exp[-K_2c_1][K_2c_1 + 1]\right)\right)\right]\right\}^{-1/2} dc. \tag{27}$$

The relationship between c and ζ with known approximation can be simplified. Taking into consideration, that for a majority of real systems the magnitude

$$K_2c_1 = (1 - \theta)\frac{1}{\Omega_{пред}\psi} \ll 1 \quad (K_2c \ll K_2c_1), \tag{27a}$$

and by changing thanks to that exp (K_2c) by two first members of decomposition into series, we obtain instead of (27) the folloing expression:

$$\zeta \approx \int_{c_0}^{c}\left\{2A\psi\left(\frac{M}{K_1} + K_1\right)c^2 - 2A\psi\frac{E}{K_1}c + K^2 - \right. \tag{27b}$$

$$\left. - 2A\psi\left(\frac{M}{K_1} + K_1\right)c_1^2 + 2A\psi\frac{E}{K_1}c_1\right\}^{-1/2} dc. \tag{27'}$$

Integrating (27') within mentioned limits, we obtain

$$\zeta \approx \frac{1}{\sqrt{a}}\left[\operatorname{Arch}\frac{2ac + b}{\sqrt{-\Delta}} - \operatorname{Arch}\frac{2ac_0 + b}{\sqrt{-\Delta}}\right], \tag{28}$$

where

$$a = 2A\psi\left(\frac{M}{K_1} + K_1\right); \quad b = -2A\psi\frac{E}{K_1};$$

$$\Delta = 8A\psi\left(\frac{M}{K_1} + K_1\right)\left\{K^2 - 2A\psi\left(\frac{M}{K_1} + K_1\right)c_1^2 + 2A\psi\frac{E}{K_1}c_1\right\} - $$
$$- 4(A\psi)^2\frac{E^2}{K_1^2}. \tag{28a}$$

Then for dimensionless concentration can be written the following expression

$$c \approx \frac{\sqrt{-\Delta}\,\mathrm{ch}\left[\varsigma\sqrt{a}+\mathrm{Arch}\,\frac{2ac_0+b}{\sqrt{-\Delta}}\right]-b}{2a}. \qquad (29)$$

The concentration at the rear surface of the electrode c_0 included in (29) can be determined by solving the transcentent equation (29) with the use of boundary condition at $\varsigma = 1$:

$$c_1 = -\theta$$

The expression for dimensionless polarization (21) can then be written in following form

$$u \approx \frac{\sqrt{-\Delta}\,\mathrm{ch}\left[\varsigma\sqrt{a}+\mathrm{Arch}\,\frac{2ac_0+b}{\sqrt{-\Delta}}\right]-b}{2aK}+ \\ +\frac{\Omega_{forw}}{\theta}\ln\left(Q'+\sqrt{Q'^2-B'}\right), \qquad (30)$$

where Q' and B' - values Q and B with consideration of the above adopted assumption.

Distribution of polarization and current in depth of the porous electrode is described consequently by the following expressions:

$$\eta = \frac{RT}{\alpha F}\frac{\theta}{\Omega_{forw}}\mu \approx \frac{RT}{\alpha F\,\Omega_{forw}}\theta \times \\ \times\left\{\frac{\sqrt{-\Delta}\,\mathrm{ch}\left[\varsigma\sqrt{a}+\mathrm{Arch}\,\frac{2ac_0+b}{\sqrt{-\Delta}}\right]-b}{2aK}+ \right. \\ \left. +\frac{\Omega_{forw}}{\theta}\ln\left(Q'+\sqrt{Q'^2-B'}\right)\right\}, \qquad (31)$$

$$i = \frac{j_0}{A\psi}c'' \approx \frac{1}{2}\frac{j_0\sqrt{-\Delta}}{A\psi}\,\mathrm{ch}\left[\varsigma\sqrt{a}+\mathrm{Arch}\,\frac{2ac_0+b}{\sqrt{-\Delta}}\right]. \qquad (32)$$

The solution of the problem regarding distribution of intensity of the electrochemical process obtained in close form for the depth of the working and diffusing conditions of the porous electrode (27) and the expressions for polarization (31) and current (32) emanating therefrom (at known assumptions) represent greater difficulties for the analysis. Possibilities for their overcoming will be discussed in the next report.

Designations

D_j - diffusion coefficient of j-component in free electrolyte; D'_j - effective coefficient of diffusion of j-component in porous electrode; d- coefficient of weakening the diffusion in the electrode in comparison with diffusion in free electrolyte; ν_j - stoichiometric coefficient at j-component of reaction; s-specific surface of porous electrode; R_g - effective resistance of working mixture in the pores of the electrode; n-number of electrons participating in the reaction; F-Faraday number; alpha-kinetic coefficient of electrode reaction; δ -thickness of diffusion layer at frontal surface of electrode; ϑ - coefficient of "charge" of the electrode; xi-coefficient of "contamination" of the reagent by the product of its oxidation (reduction); $j_o = j'_o\ (c^v_p)\ \alpha/p\ (c^w_r)\ 1-\alpha/n$ - interchange current; I_o - effective value of interchange current on the porous electrode; ψ-parameter (introduced for the first time by [11]), characterizing the ratio between the weakening of the transfer in diffusion layer, adjoining the frontal surface of the electrode, and in the electrode; $\varkappa_{forw}$-diffusibility factor, characterising the ratio between rolarisability of the electrode, loaded with a specific current, and ohmic resistance filling its working mixture.

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