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PROBLEM OF MODELING OF THE INTERACTION OF NONSTATIONARY ELECTRIC, THERMAL, AND DIFFUSION FIELDS IN LAYERED MEDIA

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The interaction of nonstationary electric and thermal fields in a layered medium with allowance for mass transfer is considered. The heating of an electrochemical cell is modeled numerically; the influence of an electric double layer at the metal–electrolyte interface is investigated.

Introduction. Investigation of the interaction of electric and thermal fields with allowance for mass transfer and contact phenomena is a complex and topical problem of the theory and practice of various fields of natural science and technology.

Two approaches are applicable to solution of the problem posed. One can consider in detail the action of an electric field on electric charges that exist independently or are incorporated into the molecules or atoms of a medium. The computations required in this case are cumbersome because it is necessary to take into account the action not only of the incident wave but also of the secondary waves from all the remaining charges [1, p. 302].

The other way of solving the problem relies on phenomenological electrodynamics whose propositions provide the basis for the investigations of the present work.

Let us consider the interface *S* of two media with different electrophysical properties. Under the action of the external electric field, surface charges σ and surface currents $\mathbf{i}$ (vectors lying in the tangential plane to the interface *S*) occur on the contact. On both sides of the interface, the vectors of the magnetic-field strength $\mathbf{H}$ and the magnetic inductance $\mathbf{B}$ and of the electric field $\mathbf{E}$ and the electric displacement $\mathbf{D}$ are finite and continuous but they can undergo a discontinuity of the first kind at the phase boundary.

In considering the electromagnetic field which interacts with a material medium, we use the Maxwell equations [1, p. 299]

$$\frac{\partial \mathbf{D}}{\partial t} + \mathbf{I}_q = \nabla \times \mathbf{H} \,, \quad \nabla \mathbf{D} = \rho \,, \quad -\frac{\partial \mathbf{B}}{\partial t} = \nabla \times \mathbf{E} \,, \quad \nabla \mathbf{B} = 0 \,, \tag{1}$$

where $\mathbf{I}_q$ is the conduction current and ρ is the space electric charge. At the interface *S*, the system of equations (1) is supplemented with the conditions [1, p. 401]

$$D_{\text{n}1} - D_{\text{n}2} = \sigma \,, \quad B_{\text{n}1} - B_{\text{n}2} = 0 \,, \quad E_{\tau 1} - E_{\tau 2} = 0 \,, \quad H_{\tau 1} - H_{\tau 2} = \frac{4\pi}{c} (\mathbf{i} \cdot \mathbf{n}) \,. \tag{2}$$

Here c is the electrodynamic constant, the subscripts n and τ denote the normal and tangential components of the vectors to the interface *S*, and the subscripts 1 and 2 denote the adjacent media with different electrophysical properties.

The surface charge σ is formed due to the spontaneous redistribution of ions or electrons at the interface of a layered medium for equalization of Fermi energy levels [2, p. 425], which results in the occurrence of an electric double layer and a spatial distribution of electric charges at the boundary of contact of different substances.

Nonstationary thermal and diffusion processes influence the structure of the electric double layer, which makes the problem of modeling of electric fields much more difficult. The reasons for the charge distribution can be different: in the case of the electrolyte–metal contact it is attributed to the transition of the ions from the electrode to the solution, specific adsorption of the ions of one sign on the electrode surface, and orientation of polar molecules near the

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electrode surface [3, p. 39]. Other factors are responsible for the structure of the electric double layer in the case of contact of two solid conductors or a dielectric and a conductor; this structure has its distinctive features [4, p. 490; 5].

We note that the structure of the electric double layer substantially influences electrokinetic phenomena, the rate of electrochemical processes, and the stability of colloidal systems.

For the indicated reasons, the electric double layer produces fundamental difficulties in modeling electric fields in a layered medium.

Construction of the equivalent circuits to take into account the electric double layer through introduction of the surface capacitance [5] which is determined experimentally is worthwhile only for the range of those conditions under which it has been determined.

The work seeks to construct a physicomathematical model of the interaction of nonstationary electric fields in a layered medium with allowance for nonstationary thermal phenomena and mass transfer without explicit separation of charge carriers. The media in contact are considered to be homogeneous. For the sake of clear representation, we will consider two-layer, one-dimensional models.

Interaction of Electric and Thermal Fields. In different substances, the processes of transfer of charge and energy are interrelated. The quantity of the released heat is determined by Joule heating and by the effects of Thomson and Peltier. The problem of interaction of nonstationary thermal phenomena has been considered in [6] without taking into account the Thomson effect.

According to [7, p. 153], the expressions for the conduction-current density and the energy-flux density in the absence of an external magnetic field or in the case of its slight influence have, respectively, the form

$$\mathbf{I}_q = -\lambda\,(\alpha\,(T)\ \mathrm{grad}\ T + \mathrm{grad}\ \varphi)\ , \quad \mathbf{I}_T = -k\,(T)\ \mathrm{grad}\ T + \mathbf{I}_q\,(\Pi + \varphi)\ ,$$

where $\alpha(T)$ is the specific thermoelectromotive force, $\Pi = \alpha T$ is the Peltier coefficient, φ is the potential, and $k(T)$ is the thermal-conductivity coefficient. We note that the problem is always nonlinear.

Having eliminated the magnetic-field strength from system (1) according to one method of [8–10], we obtain the equation for the electric-field strength

$$\frac{\varepsilon}{c^2}\frac{\partial^2 \mathbf{E}}{\partial t^2} + \mu_0 \frac{\partial \mathbf{I}_q}{\partial t} = \frac{1}{\mu}\nabla^2 \mathbf{E}\ . \tag{3}$$

The heat-balance equation has the form [7, p. 154]

$$c_p\rho\,\frac{\partial T}{\partial t} = \mathrm{div}\,(k\,(T)\ \mathrm{grad}\ T) + \mathbf{I}_q \cdot (\mathbf{E} - (\alpha\,(T) + T\beta\,(T))\ \mathrm{grad}\ T)\ . \tag{4}$$

At the interface, the following relation [11, p. 420] holds:

$$\mathrm{div}\,\mathbf{i} + I_{qn1} - I_{qn2} = -\frac{\partial \sigma}{\partial t}\ . \tag{5}$$

Here ε is the permittivity, $c^2 = 1/(\varepsilon_0\,\mu_0)$, μ is the magnetic permeability, ε_0 is the electric constant, μ_0 is the magnetic constant, $c_p\rho$ is the product of the specific heat of the medium by its density, and $\beta(T) = \partial\alpha(T)/\partial T$.

In deriving Eq. (4), use has been made of the condition of local electroneutrality of the substance. We give a differential formulation of the problem and the method of its solution in greater detail.

Let us investigate, on the segment $[0 \le x < l] = [0 \le x < \xi] \cup [\xi \le x \le l]$, the contact of homogeneous media 1 and 2 with different electrophysical properties. The quantities ε, λ, μ, E, and φ have discontinuities of the first kind at the point of the interface $x = \xi$. We will consider the case of plane contact where the influence of surface currents can be disregarded and the thickness of the electric double layer is much smaller than the characteristic dimension of the object. We set $E = -\mathrm{grad}\,\varphi$. Equations (3) and (4) for the one-dimensional problem will take the form

$$\frac{\varepsilon}{c^2}\frac{\partial^2 E}{\partial t^2} + \mu_0 \frac{\partial}{\partial t}\left(-\lambda\left(\alpha\,(T)\frac{\partial T}{\partial x} - E\right)\right) = \frac{1}{\mu}\frac{\partial^2 E}{\partial x^2}\ , \tag{6}$$

$$c_p\rho\,\frac{\partial T}{\partial t}=\frac{\partial}{\partial x}\left(k\,(T)\,\frac{\partial T}{\partial x}\right)-\lambda\left(\alpha\,(T)\,\frac{\partial T}{\partial x}-E\right)\left(E-(\alpha\,(T)+T\beta\,(T))\,\frac{\partial T}{\partial x}\right). \tag{7}$$

Conditions (2) and (5) will be written, respectively, in the form

$$\varepsilon_1\varepsilon_0E_1\,\big|_{x=\xi-0}-\varepsilon_2\varepsilon_0E_2\,\big|_{x=\xi+0}=\sigma\,\big|_{x=\xi}\,, \tag{8}$$

$$\lambda_1\left(E_1-\alpha_1\,(T)\,\frac{\partial T}{\partial x}\right)\bigg|_{x=\xi-0}-\;\lambda_2\left(E_2-\alpha_2\,(T)\,\frac{\partial T}{\partial x}\right)\bigg|_{x=\xi+0}=-\;\frac{\partial\sigma}{\partial t}\bigg|_{x=\xi}\,. \tag{9}$$

By differentiating (8) with respect to time and taking into account (9), at the phase boundary we obtain the condition of equality of the total currents

$$\left[\lambda\left(E-\alpha\,(T)\,\frac{\partial T}{\partial x}\right)+\varepsilon\varepsilon_0\,\frac{\partial E}{\partial t}\right]\bigg|_{x=\xi}=0\,. \tag{10a}$$

Here and in what follows, for the arbitrary function f we adopt the notation

$$[f]\,\big|_{x=\xi}=f\,\big|_{x=\xi+0}-f\,\big|_{x=\xi-0}\,.$$

Equality (8) is a corollary of the relation $\lim\limits_{x\to\varepsilon\pm0}\operatorname{div}\mathbf{D}=\rho$ [11, p. 40]. Taking into consideration the finiteness of the value of the space electric charge and the continuity of it, in the one-dimensional case we obtain

$$\left[\varepsilon\varepsilon_0\,\frac{\partial E}{\partial x}\right]\bigg|_{x=\xi}=0\,. \tag{10b}$$

Relations (10) reflect the laws of conservation and continuity of electric charge [7, p. 156; 11, p. 420].

At the contact point, we also have the equality of the temperatures and the energy fluxes [7, p. 256]:

$$[T]\,\big|_{x=\xi}=0\,, \tag{11a}$$

$$\left[k\,(T)\,\frac{\partial T}{\partial x}+\lambda\left(\alpha\,(T)\,\frac{\partial T}{\partial x}-E\right)(\alpha\,(T)\,T+\varphi)\right]\bigg|_{x=\xi}=0\,. \tag{11b}$$

Thus, in the presence of the interaction of the electric and thermal fields in a layered medium, the equality of the charge fluxes (10a), the equality of the charges (10b) (when the conditions of quasineutrality of the contacting media beyond the electric double layer are satisfied), the equality of the temperatures (11a), and the equality of the energy fluxes must be fulfilled at the interface of the media. In the relations for the charge and energy fluxes, we take into account cross thermoelectrical phenomena.

The process of charge of the electric field is considered on the finite time interval $[0\le t\le t_0]$. The initial conditions have the form

$$T\,(x,0)=T_0\,(x)\,,\quad E\,(x,0)=f_1\,(x)\,,\quad \frac{\partial E\,(x,0)}{\partial t}=f_2\,(x)\,. \tag{12}$$

We give the boundary conditions, for example, for the value $x=0$. We set the value of the total-current density to be known

$$\left(\lambda\left(\alpha\,(T)\,\frac{\partial T}{\partial x}-E\right)-\varepsilon\varepsilon_0\,\frac{\partial E}{\partial t}\right)\bigg|_{x=0}=j_1\,(t) \tag{13}$$

and take into account the heat exchange at the boundary using the Newton relation

$$\left. \left(k(T) \frac{\partial T}{\partial x} + \lambda \left(\alpha(T) \frac{\partial T}{\partial x} - E \right) (\alpha(T) T + \varphi) \right) \right|_{x=0} = \gamma_1 (T - T^*) \,|_{x=0} \,, \tag{14}$$

where γ_1 is the coefficient of heat exchange and T^* is the ambient temperature.

We consider the analogous relations at the right-hand boundary.

Interaction of Electric and Thermal Fields with Allowance for Mass Transfer. Let us consider the nonstationary model of heat and mass transfer for electrochemical systems with the example of electrolysis. For this problem, an electric double layer occurs at the site of contact of the electrolyte with the metallic anode and cathode. Let us set the density of the space charge beyond the electric double layer of the metal–electrolyte contact to be zero at the initial instant and hence, according to [12, p. 27], remain constant in the future while the voltage drop in the electrodes and leads is small. The influence of the electrodes on the temperature field of the electrolyte will be taken into account in terms of the coefficient of heat exchange.

Two approaches to modeling of diffusion-electrical phenomena have currently been developed, each of which has certain disadvantages and advantages. The first approach [13–15] is characterized by consideration of the flows of ions of the corresponding sort in a completely or partially dissociated electrolyte. The equations derived contain many parameters, the reliable procedure of determination of which is absent in the majority of cases. Furthermore, the proposed theory fails to provide for taking into account the interaction of the cations and the anions of the dissolved substance with each other and with the molecules of the solvent. It must also be borne in mind that, apart from the dissociation reaction, we have the association of simple molecules which in turn are dissociated into complex and simple ions. It is difficult to determine the diffusion coefficients of complex ions; it is also difficult to take into account the force interaction of complex cations and anions with each other and with the external electric field. The difficulties in question become more serious when a multicomponent electrolyte is described. We note that such difficulties also arise in modeling, for example, certain problems of plasma physics.

The second approach [16–19] fails to provide for the introduction and determination of the coefficients of molecular diffusion of cations and anions and the degree of dissociation of the electrolyte. It is based on the interaction of the mass and charge fluxes. The ionic flows are not determined.

The system of equations describing the electrodynamic processes in the electrolyte is as follows:

$$\frac{\varepsilon}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} + \mu_0 \frac{\partial \mathbf{I}_q}{\partial t} = \frac{1}{\mu} \nabla^2 \mathbf{E} \,, \quad \frac{\partial n}{\partial t} = - \operatorname{div} (\mathbf{I}_{\mathrm{m}}) \,, \quad c_0 \rho_0 \frac{\partial T}{\partial t} = - \operatorname{div} (\mathbf{I}_T) \,, \tag{15}$$

Here $\mathbf{I}_q$, $\mathbf{I}_{\mathrm{m}}$, and $\mathbf{I}_T$ are the densities of the conduction current and of the mass and heat fluxes which (in the case of the nonequilibrium state of the medium) have, respectively, the form [7, p. 153]

$$\begin{gathered} \mathbf{I}_q = \lambda (\mathbf{E} - \lambda_{\mathrm{A}}^* \operatorname{grad} n) - \lambda \alpha (T) \operatorname{grad} T \,, \\ \mathbf{I}_{\mathrm{m}} = - D_{\mathrm{M}} \operatorname{grad} n + D_{\mathrm{A}}^* \lambda \mathbf{E} - D_T \operatorname{grad} T \,, \\ \mathbf{I}_T = - k \operatorname{grad} T + \mathbf{I}_q (\Pi + \varphi) - D_T^* \operatorname{grad} n \,, \end{gathered} \tag{16}$$

where D_{M} is the molecular diffusion coefficient, D_{A}^* is the ambipolar diffusion coefficient, D_T is the thermal diffusion coefficient, D_T^* is the coefficient taking into account the transfer of heat due to the motion of the impurity, and n is the concentration of the impurity.

We give the initial

$$E(x, 0) = E_0(x) \,, \quad \frac{\partial E(x, 0)}{\partial t} = \tilde{E}(x) \,, \quad T(x, 0) = T_0(x) \,, \quad n(x, 0) = n_0(x) \tag{17}$$

and boundary conditions (anode–electrolyte, $x = 0$)

$$I_q(t) = \lambda \left(\lambda_{\mathrm{A}}^{*} \frac{\partial n}{\partial x} - E \right) + \lambda \alpha (T) \frac{\partial T}{\partial x} - \varepsilon \varepsilon_0 \frac{\partial E}{\partial t} ,$$

$$\gamma_1 (T - T^{*}) = k \frac{\partial T}{\partial x} - I_q (\Pi + \varphi) + D_T^{*} \frac{\partial n}{\partial x} , \tag{18}$$

$$k_{\mathrm{a}} I_q(t) = D_{\mathrm{M}} \frac{\partial n}{\partial x} - D_{\mathrm{A}}^{*} \lambda E + D_T \frac{\partial T}{\partial x}$$

in modeling the one-dimensional problem in the region $G = [0 \le x \le l] \times [0 \le t \le T]$. Here k_{a} is the electrochemical equivalent of a substance which deposits at the anode. The analogous conditions hold for $x = l$ as well.

The constructed system of differential equations makes it possible to model the processes of transfer in the electrochemical system with allowance for the influence of an electric double layer; this influence directly determines the thermoelectrical and ambipolar electrodiffusion phenomena at the boundary of the metal–electrolyte contact.

Numerical Modeling of Electrochemical Systems. Heating of an Electrochemical Cell in the Case of Constant-Current and Pulse Electrolysis. In electrochemical systems, the electrodes are metals as a rule. The electrical conductivity of metals is hundreds of times higher than the specific electrical conductivity of electrolytes; therefore, one can disregard the voltage drop in electrodes and leads. In highly conductive metals, one can also disregard the displacement currents and the coefficients of ambipolar diffusion and thermal diffusion and restrict oneself to consideration of the system of equations (15)–(18) just in the electrolyte itself.

Let us consider the process of heating of an electrochemical medium in the case of passage of an electric current with density $I_q(t)$ through a solution of copper sulfate $CuSO_4 \cdot 5H_2O$. We use copper (99.78%) as the anode. The copper from the solution is deposited at the cathode. We set the current efficiencies for the copper equal to 100% and the electrochemical equivalent k_{e} equal to $0.6588 \cdot 10^{-6}$ kg/C. The dependence of the specific electrical conductivity $\lambda(n)$ of the copper-plating electrolyte on the concentration of the copper sulfate in water is given in [20]; the dependence of λ and k on the temperature will be considered to be insignificant. In the calculations for $CuSO_4$, we set $D_{\mathrm{M}} = 5 \cdot 10^{-10}$ m^2/sec, $D_{\mathrm{A}}^{*} = 10^{-11} \lambda$, $\lambda_{\mathrm{A}}^{*} = 10^{-4} \lambda$, $\varepsilon = 70$, and $\mu = 1$ and disregard the effects of thermal diffusion. The distance between the cathode and the anode is $L = 0.05$ m. The heat capacity, the density, and the thermal-conductivity coefficient of the electrolyte are taken to be $4.2 \cdot 10^3$ J/(kg·K), 10^3 kg/m^3, and 0.6 V/(m·K) respectively. We note that the regimes of pulse action have been considered in [21] as applied to the processes of electrodeposition of alloys. This investigation has been carried out on the basis of a separate description of the transport of ions and the employment of the Kirchhoff law for quasistationary currents in a cell. With such an approach, the displacement currents are taken into account indirectly through introduction of the capacitive current of the electrode and determination of the experimental dependence of the polarization capacitance of the electrode on the character of the pulse action. In [21], it has been noted that neglect of the capacitive current in pulse electrolysis involves significant errors. The employment of system (15)–(18) to model the process of copper plating makes it possible to consider the problems without introducing the concepts of inductance and capacitance.

The obtained nonlinear system of equations is solved by the finite-difference method analogously to [17].

We have modeled numerically the process of copper plating for a constant current and a pulse current (Fig. 1). The density of the constant current was equal to 300 A/m^3. The maximum density of the pulse current was also 300 A/m^3, while the period of traversal of the current and the break were equal to 0.01 sec.

Figure 1 gives the results of modeling the distributions of the concentration of $CuSO_4$ and the temperature for different regimes of electrolysis. The measurements were carried out within 60 sec after switching on the current. It is seen from the figure that the concentration gradients near the cathode surface ($x = 1$) are different and depend on the regime of copper plating. In the case of nonstationary action they are substantially smaller than in the case of electrolysis in the regime of constant current. As has been shown in [21], this tendency also holds in the case where one and the same total charge passes through the electrochemical cell.

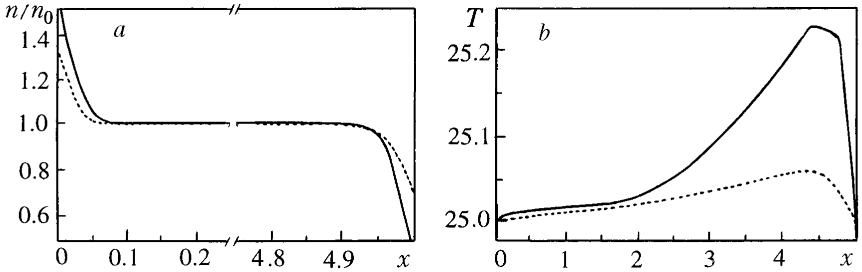

Fig. 1. Distribution of the normalized concentration of $CuSO_4$ (a) and the temperature (b) in an electrochemical cell within 60 sec after switching on the current. The solid curve, at constant current; the dashed curve, at pulse current. T, ^oC; x, mm.

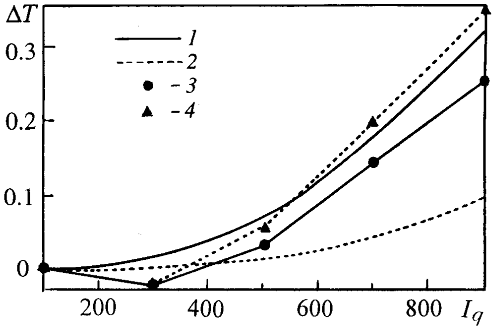

Fig. 2. Heating of the electrolyte in the cathode region as a function of the value and kind of polarizing current: 1) constant current; 2) pulse current; 3, 4) corresponding experimental data. ΔT, ^oC; I_q, A/m^2.

The analysis of the temperature field (Fig. 1b) shows that the temperatures of the electrolyte in the vicinity of the anode and the cathode differ significantly. Nonsymmetric heating of the electrochemical cell attributed to the ambipolar diffusion and electrical ambipolar conductivity occurs. The modeling results are in agreement with the data of [22, p. 29].

With the aim of measuring the temperature in the surface layer of the electrolyte solution near the cathode and comparing the results to the calculated data, we developed and manufactured an experimental setup. Copper anode and cathode were placed in an electroplating bath with a sulfuric-acid copper-plating electrolyte. In the 1-mm-thick cathode, a hole was made into which we placed a temperature-sensitive element. As the latter we employed a thermal resistor. The current strength in the bath was prescribed by a variable resistor.

Figure 2 gives the results of numerical modeling and the experimental data on heating of the cathode region in relation to the regime of current. It is seen that the increase in the density of the current (both constant and pulse) causes the temperature near the cathode surface to increase. In the anode, no increase in the temperature was observed either in the calculations or in the experiment, which is in agreement with the data of [22]. The surface temperature obtained in solving Eqs. (15)–(18) in the regime of constant current is 10–15% higher than the temperature recorded in the experiments.

A more complicated situation occurs when we compare the experimental data and the results of numerical modeling in the regime of pulse electrolysis. The results of the experiments demonstrate that in the regime of pulse current the temperature of the electrolyte in the cathode region increases more than in the regime of constant current. At the same time, the numerical calculations of the pulse electrolysis show that the heating of the cathode region here is smaller than for the regime of constant current. The difference is, apparently, attributed to concentration convection [23], whose influence is substantial for high densities of the current for pulse electrolysis.

In a longer term, one must take into account natural and concentration convection and heat exchange with the ambient medium. Nonetheless, despite the errors in determination of the coefficients and the assumptions made in the model, the calculation results are in qualitative agreement with the experimental data, which makes it possible, in certain cases, to use the proposed approach for modeling of nonstationary processes.

Conclusions. For the first time, we have constructed a consistent physicomathematical model of interaction of nonstationary electric and thermal fields in a layered medium with allowance for mass transfer. The model is based on the methods of thermodynamics and on the equations of an electromagnetic field and is formulated without explicit separation of the charge carriers and the charge of an electric double layer. We have obtained the relations for the electric-field strength and the temperature, which take into account the equality of the total currents and the energy fluxes, to describe the electric and thermal phenomena in layered media where the thickness of the electric double layer is small as compared to the dimensions of the object under study.

We have modeled numerically the heating of an electrochemical cell with allowance for the influence of the electric double layer at the metal–electrolyte interface. The calculation results are in satisfactory agreement with experimental data.

NOTATION

D_{M}, molecular diffusion coefficient, $\mathrm{m}^2/\mathrm{sec}$; L, length, m; D_{A}^{*}, ambipolar diffusion coefficient, kg/(A·sec); λ, specific electrical conductivity, $1/(\Omega\cdot\mathrm{m})$; $\lambda_{\mathrm{A}}^{*}(n)$, coefficient of specific electrical ambipolar conductivity, $\mathrm{V}\cdot\mathrm{m}^3/\mathrm{kg}$; k_{e}, electrochemical equivalent of copper, kg/C. Subscripts: n, normal; q, charge; m, impurity mass; a, anode; A, ambipolar; M, molecular; e, equivalent.

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