Model and numerical experiment for calculating the theoretical current-voltage characteristic in electro-membrane systems

M Kh Urtenov¹, A V Kovalenko¹, A I Sukhinov², N O Chubyr³, and V A Gudza¹

¹Kuban State University, 149 Stavropolskaya St, Krasnodar 350040, Russia
²Don State Technical University, Gagarin sq. 1, Rostov-on-Don 344000, Russia
³Kuban State Technological University, 2 Moskovskaya St, Krasnodar 350042, Russia

Abstract. This work is devoted to problems of numerical calculation of the theoretical current-voltage characteristics (CVC) and galvanography of electromembrane systems in the potentiometric mode. The purpose of this work is the derivation of the formula for calculating the CVC and galvanography of electromembrane systems in the potentiometric mode, which allows calculating stable relative to random errors and rounding errors in over-limiting current densities. To use the formulas, it is necessary to create a mathematical model for calculating the local current density. In this paper, as this model model is used transport of ions in binary salt in the desalination channel, taking into account electroconvection. The electromembrane systems have been used for water medium preparing in mobile mini systems for generating heat and electrical energy as well as vapour producing. The quality of clean water influences on reliability and life time of these systems, including vapour generator and dam-vapour turbine. The obtained formula in the form of a double integral of the local current density is derived, which allows calculating them steadily with respect to random errors and rounding errors. The formulas allowing considering CVC stably with respect to random errors and rounding errors are obtained, and the physical meaning of these formulas is clarified. The calculation of the theoretical CVC with the use of a mathematical model of ion transfer binary salts given electroconvection and shows that it qualitatively coincides with the experimental current-voltage curves. The quantitative difference can be explained by the fact that the mathematical model does not take into account the reaction of water dissociation / recombination, gravitational convection and other transport mechanisms.

1. Introduction

The current-voltage characteristic (CVC) is a common, integral and most important characteristic of heat and mass transfer processes in electromembrane systems. The CVC is used to evaluate and select the optimal design and effective operating modes. In theoretical studies, using the CVC, the fundamental concept of the limiting current is introduced; various modes of functioning of the electromembrane systems are introduced and analyzed. Qualitative and quantitative correspondence of theoretical CVC calculated using mathematical models and experimental CVC is the most important argument in favor of the adequacy of the corresponding models. However, to date, the only way to study the CVC is to analyze experimental data. The study of the experimental CVC in the over-limiting regime, carried out in [1], by the methods of Fourier analysis, wavelet analyzes [2], and dynamic chaos methods, shows the complex, unsteady, and unstable behavior of the CVC. For a
theoretical study of this behavior of the CVC, special mathematical methods and models are required. The absence of such models does not allow us to identify the relationship between the behavior of the CVC and the properties of the salt ion transfer process in electromembrane systems, for example, to adequately interpret the growth and vibrations, the instability of the CVC, with the size and velocity of the vortex structures, their interaction and bifurcations, etc. As a result of this, it is impossible to carry out engineering calculations to optimize the designs of electro-membrane systems, including optimizing their geometric and technological characteristics in order to intensify transfer processes in electro-membrane systems.

Here, for the first time, a formula is proposed that allows one to calculate the theoretical CVC based on some mathematical model. As an example, we use a 2D model of salt ion transport in the desalination channel of an electrodialysis apparatus (EDA) with homogeneous, perfectly selective ion-exchange membranes in an over-limiting current mode taking into account electroconvection, in the form of a boundary value problem for the system of Nernst-Planck-Poisson and Navier-Stokes equations [3].

Let the flow channel of desalination in the electrical circuit is indicated in the form:

Figure 1. 2D scheme - images of the desalination channel in the electrical circuit.

Here AEM is an anion-exchange membrane, CEM is a cation-exchange membrane, the sign $\uparrow \vec{V}$ shows the direction of forced convection, the sign $\rightarrow$ shows the directions of motion of anions, shows the directions of motion of cations. Suppose that the OY axis passes through the AEM, and the OX axis perpendicular to it is directed toward the CEM. Let the channel width be equal $H$ and the length $L$. The current density at an arbitrary point in the channel at some point in time is denoted $\vec{I}(t,x,y)$. The rectangle $\Omega$ is used below when applying the Gauss theorem.

Electromembrane processes can operate in the potentiodynamic mode, when the potential jump is set and the current density is calculated, or in the galvanodynamic mode, when the current density passing through the electromembrane system is set and the corresponding potential jump is calculated. We will consider the potentiodynamic mode in this paper. For figure 1 shows a diagram of the flow channel desalination. With some accuracy, it can be assumed that the surfaces of ion-exchange membranes are equipotential, and then it is possible to combine the longitudinal walls of the rectangle with the surface of the membranes. In comparison with the experiment, it is necessary in this case to take into account the jumps of potential through the membranes. For the theoretical calculation of CVC potentiometric mode, assume that a known current density $\vec{I}(t,x,y)$ at each point $(x,y)$ region at time $t$, given a jump in potential. We will denote

$$i_A = \frac{1}{L} \int_0^L I_x(t,0,y) \, dy,$$

$$i_K = \frac{1}{L} \int_0^L I_x(t,H,y) \, dy,$$

In the stationary case, $i_A$ and $i_K$ are averaged densities of currents passing through AEM and CEM, so when the potential jump changes, they represent the current – voltage characteristic of AEM and CEM, respectively. In the unsteady case, at a fixed potential jump, they are averaged galvanic grams
of AEM and CEM. The arithmetic mean of these values can be considered in many cases as a characteristic of the entire channel. It is \( i_A \) and \( i_K \) that are the required values. At first glance, \( i_A \) and \( i_K \) are easy to calculate using these formulas if \( \tilde{I}(t, x, y) \) is known. However, these formulas use the ordinates values of the current density at the boundary, which, as will be shown below, in turn depend on the concentration derivatives and the potential jump over the variable \( x \) at the boundary. Small errors in the computation of these derivatives lead to large errors in the computation of \( i_A(t, x, y) \) at \( x = 0 \) and \( x = H \). It would seem that integrals will correct these errors, but this does not happen since integrals are taken by the variable \( y \). Therefore, it is necessary to derive a formula for calculating \( i_A \) and \( i_K \), stable with respect to rounding errors.

2. Formula for calculating CVC

The formula for calculating CVC is based on Gauss' law[4-5], the formulation of which depends on whether the current density is a solenoid field or not.

2.1. Gauss' Law

In the two-dimensional case, the Gauss' law is written as [4]:

\[
\oint_{\partial \Omega} \mathbf{I} \cdot d\mathbf{l} = \iint_{\Omega} \text{div} \mathbf{I} \, dS
\]

Here \( d\mathbf{l} = \vec{n}dl \) is the element of the oriented boundary, i.e. \( \vec{n} \) – normal to the boundary, \( dl \) is an infinitesimal element of the boundary, and \( dS \) infinitesimal element of the area.

If we consider separately the potential jumps through the membranes and consider them constant as in [3,5,6], we can assume \( \Omega = \{ (x, y) : 0 \leq x \leq H, 0 \leq y \leq L \} \), then:

\[
\oint_{\partial \Omega} \mathbf{I} \cdot d\mathbf{l} = \int_{0}^{L} I_x(t, H, y) \, dy - \int_{0}^{L} I_x(t, 0, y) \, dy + \int_{0}^{H} I_y(t, x, L) \, dx - \int_{0}^{H} I_y(t, x, 0) \, dx
\]

and the integral Gauss' law is written as:

\[
\int_{0}^{L} I_x(t, H, y) \, dy - \int_{0}^{L} I_x(t, 0, y) \, dy +
\]

\[
+ \int_{0}^{H} I_y(t, x, L) \, dx - \int_{0}^{H} I_y(t, x, 0) \, dy = \iint_{\Omega} \text{div} \mathbf{I}(t, x, y) \, dxdy
\]

2.2. Derivation of the formula for calculating the CVC in the electrical circuit for the solenoid case

For solenoidal current density \( \text{div} \mathbf{I} = 0 \) and the integral Gauss' law, as follows from the previous formula becomes: \( \iint_{\partial \Omega} \mathbf{I} \cdot d\mathbf{l} = 0 \) or

\[
\int_{0}^{L} I_x(t, H, y) \, dy - \int_{0}^{L} I_x(t, 0, y) \, dy + \int_{0}^{H} I_y(t, x, L) \, dx - \int_{0}^{H} I_y(t, x, 0) \, dy = 0
\]

Let us take an arbitrary point \( s \in (0, H) \) and consider the section \( S = \{ (x, y) : x = s, 0 < y < L \} \) and region \( \Omega_1 = \{ (x, y) : 0 \leq x \leq s, 0 \leq y \leq L \} \) and \( \Omega_2 = \{ (x, y) : s \leq x \leq H, 0 \leq y \leq L \} \). It is obvious that \( \Omega = \Omega_1 \cap \Omega_2 \).

Since the field current density is solenoidal in a region \( \Omega \), then \( \text{div} \mathbf{I} = 0 \) in each of the areas \( \Omega_1 \) and \( \Omega_2 \), and therefore: \( \iint_{\Omega_i} \text{div} \mathbf{I} \, dxdy = 0, \ i = 1,2. \)

From the Gauss' law applied to each of the domains \( \Omega_1 \) and \( \Omega_2 \), it follows:

\[
\int_{0}^{L} I_y(t, x, 0) \, dx + \int_{0}^{L} I_x(t, 0, y) \, dy - \int_{0}^{s} I_y(t, x, L) \, dx - \int_{0}^{s} I_x(t, s, y) \, dy = 0
\]

\[
\int_{0}^{L} I_y(t, x, 0) \, dx + \int_{0}^{L} I_x(t, 0, y) \, dy - \int_{0}^{H} I_y(t, x, L) \, dx - \int_{0}^{H} I_x(t, x, 0) \, dy = 0
\]
We multiply the first equation by \(-1\) and add it to the second, then we get the equality valid for any \(s \in [0, H]\):

\[
\int_0^H I_y(t, x, 0)dx - \int_0^L I_x(t, H, y)dy - \int_0^H I_y(t, x, L)dx + \int_0^L I_x(t, s, y)dy = 0
\]

We integrate this identity in \(s\) from 0 to \(H\), and then we get:

\[
2 \int_0^H \int_0^L I_x(t, s, y)dy \, ds = -\int_0^H \int_0^L I_y(t, x, 0)dx \, ds + H \int_0^L I_x(t, H, y)dy + \int_0^H \int_0^L I_y(t, x, L)dx \, ds + H \int_0^L I_x(t, 0, y)dy - \int_0^H \int_0^L I_y(t, x, L)dx \, ds
\]

Changing the order of integration in integrals:

\[
\int_0^H \int_0^L I_y(t, x, 0)dx \, ds, \int_0^H \int_0^L I_y(t, x, L)dx \, ds, \int_0^H \int_0^L I_y(t, x, 0)dx \, ds, \int_0^H \int_0^L I_y(t, x, L)dx \, ds
\]

We got:

\[
\int_0^H \int_0^L I_y(t, x, 0)dx \, ds = \int_0^H \int_0^L I_y(t, x, L)dx \, ds = \int_0^H xI_y(t, x, 0)dx, \int_0^H \int_0^L I_y(t, x, L)dx \, ds = \int_0^H xI_y(t, x, L)dx, \int_0^H \int_0^L I_y(t, x, 0)dx \, ds = \int_0^H \int_0^L I_y(t, x, 0)dx \, ds = \int_0^H (H - x)I_y(t, x, 0)dx, \int_0^H \int_0^L I_y(t, x, L)dx \, ds = \int_0^H \int_0^L I_y(t, x, L)dx \, ds = \int_0^H (H - x)I_y(t, x, L)dx.
\]

Therefore:

\[
2 \int_0^H \int_0^L I_x(t, s, y)dy \, ds = -\int_0^H \int_0^L I_y(t, x, 0)dx \, ds + H \int_0^L I_x(t, H, y)dy + \int_0^H \int_0^L I_y(t, x, L)dx \, ds + H \int_0^L I_x(t, 0, y)dy - \int_0^H \int_0^L I_y(t, x, L)dx \, ds
\]

Further:
\[
2 \int_0^H \int_0^L I_x(t,s,y) dy \, ds = H \int_0^L I_x(t,H,y) dy + H \int_0^L I_x(t,0,y) dy + \int_0^H (H-2x)I_y(t,x,0) dx - \int_0^H (H-2x)I_y(t,x,L) dx
\]

Thus:
\[
\frac{1}{2L} \left( \int_0^L I_x(t,H,y) dy + \int_0^L I_x(t,0,y) dy \right) = \frac{1}{HL} \int_0^H \int_0^L I_x(t,s,y) dy \, ds + \frac{1}{2HL} \left( \int_0^H (H-2x)I_y(t,x,L) dx - \int_0^H (H-2x)I_y(t,x,0) dx \right)
\]

Denote by \( i_{av} = \frac{1}{2} (i_A + i_K) \) the average current. Then from (1) follows:
\[
i_{av} = \frac{1}{HL} \int_0^L I_x(t,s,y) dy \, ds + \frac{1}{2L} \left( \int_0^H (H-2x)I_y(t,x,L) dx - \int_0^H (H-2x)I_y(t,x,0) dx \right)
\]

We denote \( i_{in} = \frac{1}{H} \int_0^H I_y(t,x,0) dy \), \( i_{out} = \frac{1}{H} \int_0^H I_y(t,x,L) dx \), which are averaged incoming and outgoing current densities.

Simple estimates show that
\[
\left| \frac{1}{H} \int_0^H (H-2x)I_y(t,x,L) dx - \frac{1}{H} \int_0^H (H-2x)I_y(t,x,0) dx \right| \leq |i_{in}| + |i_{out}|
\]

Because \( \frac{1}{2HL} \left( \int_0^H (H-2x)I_y(t,x,L) dx - \int_0^H (H-2x)I_y(t,x,0) dx \right) \leq \frac{1}{L} \left( \frac{|i_{in}| + |i_{out}|}{2} \right) \).

If the input and output currents are small, or (and) the channel is long enough (in practice, the channel length is usually tens or hundreds of times longer than the width), then
\[
i_{av} \approx \frac{1}{HL} \int_0^H \int_0^L I_x(t,s,y) dy \, ds
\]

Unlike integrals (1) in the double integral (2), errors in the calculation of the derivative with respect to the variable \( x \) are compensated by integration with respect to the same variable.

Thus, the double integral (2) makes sense of the averaged current density (for the solenoid case) and allows calculating CVC accurately and adequately.

Formula (2) is quite accurate for long channels, but for short channels gives the value of the current with a significant error.

2.3. Derivation of the formula for calculating the CVC in the electrical circuit for the non-solenoid case

In general, for non-solenoid current density output formula for calculating the CVC is similar to the previous one and it has the form:
\[
i_{av} \approx \frac{1}{HL} \int_0^H \int_0^L I_x(t,s,y) dy \, ds - \frac{1}{2HL} \int_0^L \int_0^H (H-2x) div\vec{\nabla}dxdy
\]

where the averaged current density \( i_{av} \) is sum of the solenoidal part of the average current density \( i_{sd} = \frac{1}{HL} \int_0^H \int_0^L I_x(t,s,y) dy \, ds \) and non-solenoidal part of the average current density \( i_{acd} \):
\[
i_{av} = i_{sd} + i_{acd}
\]

It is easy to show that
\[
|i_{acd}| \leq \frac{1}{2HL} \int_0^L \int_0^H (H-2x) div\vec{\nabla}dxdy \leq \frac{1}{HL} \int_0^H \int_0^H |div\vec{\nabla}|dxdy,
\]
therefore, if \( \max_{(x,y) \in \Omega} |div\mathbf{f}| \) is small, i.e. the current density is almost solenoidal, then again, get
\[
i_{av} \approx \frac{1}{H \ell} \int_{L_0}^{H_0} \int_{L_0}^{H_0} I(t, s, y) \, dy \, ds.
\]

3. Mathematical model
For the theoretical calculation of CVC in the potentiodynamic mode, it is necessary to create a mathematical model to calculate the current density \( \mathbf{I}(t, x, y) \) at each point \((x, y)\) of the \( \Omega \) region at any time \( t \), for a given potential jump. The following secondary or conjugate phenomena of concentration polarization occur in over-limiting current regimes in membrane systems:

- the spatial electric charge occupies a macroscopic region, smaller, but comparable to the thickness of the diffusion layer [11],
- intensive generation of hydrogen and hydroxyl ions begins [12-17],
- microconvective flows, gravitational and electroconvection intensifying mass transfer appear in the system [18-22]

To assess the impact of each of these phenomena on the transport of salt ions, it is necessary to conduct a theoretical study using CVC, and to calculate the CVC it is necessary to use mathematical models of salt ion transport.

Currently, it is considered that electroconvection is the main mechanism of over-limiting mass transfer, so in this article we will limit the study to the calculation of CVC using the binary salt ion transport model taking into account electroconvection [3]. This model consisting of a system of partial differential equations is given below.

3.1. System of equations
The Nernst-Planck-Poisson and Navier-Stokes equations are used to describe the transfer process [4]:
\[
\mathbf{j}_i = \frac{F}{RT} z_i D_i \mathbf{E} - D_i \nabla C_i + C_i \mathbf{V}, \quad i = 1, 2
\]
\[
\frac{\partial C_i}{\partial t} = -div \mathbf{j}_i, \quad i = 1, 2
\]
\[
\varepsilon \Delta \Phi = -F (z_1 C_1 + z_2 C_2)
\]
\[
\mathbf{I} = F(z_1 \mathbf{j}_1 + z_2 \mathbf{j}_2)
\]
\[
\frac{\partial \mathbf{V}}{\partial t} + (\mathbf{V} \cdot \nabla) \mathbf{V} = -\frac{1}{\rho_0} \nabla P + \nu \Delta \mathbf{V} + \frac{1}{\rho_0} \mathbf{f} + \frac{\partial \mathbf{f}}{\partial t}.
\]
\[
\text{div} (\mathbf{V}) = 0.
\]

where \( C_1, C_2 \) – concentrations of cations and anions in solution, respectively, \( z_1, z_2 \) – charge numbers of cations and anions, \( D_1, D_2 \) – diffusion coefficients of cations and anions, respectively, \( \Phi \) – electric field potential, \( \varepsilon_0 \) – dielectric permittivity of the electrolyte, \( F \) – Faraday constant, \( R \) – universal gas constant, \( T \) – absolute temperature. In this case \( P, \mathbf{V}, \Phi, C_1, C_2 \) – is the unknown function depending on time \( t \) and coordinates \( x, y \), \( f \) – is the density of electric force.

The density of the electric field has the form [6]:
\[
\mathbf{f} = \rho \mathbf{E}.
\]

Where \( \rho = F(z_1 C_1 + z_2 C_2) \) the density distribution of space charge, and \( \mathbf{E} = -\nabla \Phi \) is the electric field strength. Formula (5) can be written in another way, using the Poisson equation (3):
\[
\mathbf{f} = \rho \mathbf{E} = -\varepsilon \Delta \Phi = \varepsilon \Delta \Phi \nabla \Phi = \varepsilon \mathbf{E} \text{div} \mathbf{E}.
\]
3.2. Current density divergence:
Multiply by $z_i$, and fold, then
\[
\frac{\partial}{\partial t} (z_1 j_1 + z_2 j_2) = -\text{div} (z_1 j_1 + z_2 j_2),
\]
or
\[
\frac{\partial \rho}{\partial t} = -F \text{div} (z_1 j_1 + z_2 j_2),
\]
where $\rho = F(z_1 C_1 + z_2 C_2)$ is the density of the charge distribution. Given (4), we obtain Gauss's law in differential form:
\[
\frac{\partial \rho}{\partial t} = -F \text{div} \vec{I}
\]
Therefore, $\text{div} \vec{I} = 0$ only if the condition of local electroneutrality $\rho = F(z_1 C_1 + z_2 C_2) = 0$, or is considered a stationary process $\rho = F(z_1 C_1 + z_2 C_2) = \text{constant}$.

Thus, the current density is a solenoid field only for the stationary problem and (or) in those areas where the local electroneutrality condition is satisfied.

3.3. Formula for calculating the current density
The current density in the desalination channel must be considered taking into account the cation and anion fluxes, according to the equation:
\[
\vec{I} = F(z_1 j_1 + z_2 j_2)
\]
Hence, taking into account the formula (6):
\[
\vec{j}_i = \frac{F}{RT} z_i D_i C_i \vec{E} - D_i \vec{V} C_i + \vec{V}_i, \quad i = 1, 2
\]
We have
\[
\vec{I} = \frac{F^2}{RT} (z_1^2 D_1 C_1 + z_2^2 D_2 C_2) \vec{E} - F(z_1 D_1 \vec{V} C_1 + z_2 D_2 \vec{V} C_2) + F(z_1 C_1 + z_2 C_2) \vec{V}
\]
or
\[
\vec{I} = -\frac{F^2}{RT} (z_1^2 D_1 C_1 + z_2^2 D_2 C_2) \vec{V} \phi - F(z_1 D_1 \vec{V} C_1 + z_2 D_2 \vec{V} C_2) + F(z_1 C_1 + z_2 C_2) \vec{V}
\]

3.4. Results of numerical experiment
In the paper the analysis of theoretical CVC and its comparison with experimental current-voltage curves (figure 2). The qualitative correspondence of theoretical and experimental CVC is seen. The numerical difference is explained by the idealized nature of the calculated CVC, since in the mathematical model membranes are considered ideal selective planes that have no thickness. In addition, the mathematical model does not take into account the gravitational convection and the effect of dissociation/recombination of water.

Figure 2 – Comparison of theoretical CVC (NPP/NS) and experimental CVC (other curves [23])
4. Discussion and conclusion

Formulas are obtained that allow us to count CVC stably with respect to random errors and rounding errors and the physical meaning of these formulas is elucidated. The calculation of the theoretical CVC using a mathematical model of ion transport of a binary salt taking into account electroconvection is given and it is shown that it qualitatively coincides with the experimental CVC. The quantitative difference can be explained by the fact that the mathematical model does not take into account the dissociation/recombination reaction of water, gravitational convection and other transport mechanisms. In this regard, it is necessary to formulate the appropriate models, to conduct their numerical study, to calculate the CVC for them and to conduct their comparative analysis.

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