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Identification of unknown diffusion and convection coefficients in ion transport problems from flux data: an analytical approach

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Abstract   This article presents an analytical approach for identification problems related to ion transport problems. In the first part of the study, relationship between the flux $\varphi_L := (D(x)u_x(0, t)_{x=0}$ and the current response $\mathcal{I}(t)$ is analyzed for various models. It is shown that in pure diffusive linear model case the flux is proportional to the classical Cottrelian $\mathcal{I}_C(t)$. Similar relationship is derived in the case of nonlinear model including diffusion and migration. These results suggest acceptability of the flux data as a measured output data in ion transport problems, instead of nonlocal additional condition in the form an integral of concentration function. In pure diffusive and diffusive-convective linear models cases, explicit analytical formulas between inputs (diffusion or/and convection coefficients) and output (measured flux data) are derived. The proposed analytical approach permits one to determine the unknown diffusion coefficient from a single flux data given at a fixed time $t_1 > 0$, and unknown convection coefficient from a single flux data given at a fixed time $t_2 > t_1 > 0$. Linearized model of the nonlinear ion transport problem with variable diffusion and convection coefficients is analyzed. It is shown that the measured output (flux) data can not be given arbitrarily.

Keywords   Ion transport problem · Coefficient identification · Fluxdata · Cottrelian · Analytical relationship

1 Introduction

We study inverse and optimal control problems related to determination of unknown diffusion and convection coefficients in ion transport problems. Theoretical and

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computational modeling of electroanalytical experiments, performed under conditions of semi-infinite diffusion-migration transport has a growth interest in recent years (see, for example, [1,2] and references therein). Mathematical modeling of such problems in electrochemistry, in general, and in chronoamperometry, in particular, is usually based on linear Nernst-Planck equations [3–6]. In many cases these models are quite complicated due to their nonlinearity. More realistic models, in particular, models in real time and space domains, certainly require numerical simulations [5,7–10]. This is one of reasons why most of the results have been obtained for simplest or/and steady state experiments. Analytical solutions obtained for these simplest models permit one to understand experiments. Moreover, some relationships obtained by this way, may play a key role in finding out some distinguished features of ion transport problems, which can not be estimated experimentally.

Consider one-dimensional model of mass and charge transport in a controlled potential experiment, i.e. in chronoamperometry, given by Pfabe [4] and Cohn et al. [5]. We assume that there is an electrode at $x = 0$, and a medium containing mobile ions and electroactive species in the one-dimensional motion from the electrode to $x = \infty$. A potential $E = E(x, t)$ introduced at the initial time $t = 0$ causes a *reduced fraction* of the *oxidized species* at the surface of the electrode. As oxidized species are reduced at the surface of the electrode, its concentration decreases, and the concentration $u(x, t)$ of the reduced species at the electrode increases. As a result there arises two diffusion processes: oxidized species diffuse in toward $x = 0$, and the reduced species, out into the medium. Hence ion transport here can be regarded as a superposition of diffusion and migration. Exchange of electrons between the surface of the electrode and electroactive species in the time $t > 0$ gives rise to the *current response* $\mathcal{I} = \mathcal{I}(t)$, which is related to the concentration of reduced species by the equation

$$\int_0^\infty u(x, t)dx = \frac{1}{nFS_e} \int_0^t \mathcal{I}(\tau)d\tau. \tag{1}$$

Here n is the number of electrons gained by an ion upon reduction, F is Faraday's constant and S_e is the surface of the electrode. The total charge carried by the reduced species is

$$Q(t) = \int_0^t \mathcal{I}(\tau)d\tau. \tag{2}$$

Definitions (1, 2) permit one to define the total charge $Q(t)$ and the current response $\mathcal{I}(t)$ via the concentration $u(x, t)$ of the reduced species as follows:

$$Q(t) = nFS_e \int_0^\infty u(x, t)dx, \quad \mathcal{I}(t) = nFS_e \int_0^\infty \frac{\partial u(x, t)}{\partial t}dx. \tag{3}$$

We introduce also the scaled total charge, defined as

$$q(t) = \int_0^\infty u(x,t)dx. \tag{4}$$

Then the current response $\mathcal{I}(t)$ can be defined via the scaled total charge $q(t)$ as follows:

$$\mathcal{I}(t) = n\mathcal{F}S_e q'(t). \tag{5}$$

In view of the above physico-chemical model, pure diffusive and diffusive-convective linear models can be described by the following initial-boundary value problems

$$\begin{cases} \frac{\partial u}{\partial t} = D\frac{\partial^2 u}{\partial x^2}, & x > 0, \ t > 0; \\ u(x,0) = 0, & x > 0; \\ u(0,t) = u_0/z_r, & t > 0; \end{cases} \tag{6}$$

$$\begin{cases} \frac{\partial u}{\partial t} = \frac{\partial^2 u}{\partial x^2} - v\frac{\partial^2 u}{\partial x^2}, & x > 0, \ t > 0; \\ u(x,0) = 0, & x > 0; \\ u(0,t) = u_0/z_r, & t > 0, \end{cases} \tag{7}$$

respectively. Here $D > 0$, $c > 0$ are the diffusion and convection coefficients, and z_r is the valences of the reduced species.

The scaled nonlinear analogue of the above linear models leads to the following nonlinear identification (optimal control) problem [5]

$$\begin{cases} u_t = (g(u)u_x)_x + q'(t)h(u)_x, & x > 0, \ t > 0, \\ u(x,0) = 0, & x > 0, \\ u(0,t) = 1, & t > 0, \\ q(t) = \int_0^\infty u(x,t)dx, & t \geq 0, \end{cases} \tag{8}$$

with respect to the concentration $u(x,t)$ of the reduced species. Under the assumptions

(A1) $u(\infty,t) = u_x(\infty,t) = 0, \ \forall t > 0$;

(A2) $z_r D_r = z_o D_o$,

the nonlinear model (8) has been analyzed in [10,11]. In this case the functions $g(u)$ and $h(u)$ in Eq. (7) have the following forms:

$$g(u) := 1 + \left(\frac{z_r}{z_0} - 1\right)u, \quad h(u) := \frac{1}{z_r}u. \tag{9}$$

The aim of this study is to analyze the above models from points of view optimal control and inverse problems. In view of experiments, for identification of unknown diffusion or/and diffusion and convection coefficients, most acceptable additional

condition (measured output data) is the flux $\varphi_L(t) := (D(x)u_x(0, t))_{x=0}$. The identification problem (8) does not contain this data; instead it contains the additional nonlocal condition in the form of the integral of concentration function $u(x, t)$, which means the scaled total charge. Thus, the first problem here is to find out a relationship between the left data and the current response/scaled total charge, which is given in the nonlinear model (8).

The paper is organized as follows. In Sect. 2, relationships between the flux and the current response/scaled total charge are established for above linear and nonlinear models. An inverse problem for pure diffusive linear model with output flux data is analyzed in Sect. 3. A simple formula for determination of the unknown diffusion coefficient D from the single flux data $\varphi_L^{(1)} = \varphi_L(t_1)$ given at a fixed time $t_1 > 0$, is derived. In Sect. 4 the inverse problem of simultaneous determination of constant diffusion and convection coefficients in (7), from two data $\varphi_L^{(1)} = \varphi_L(t_1)$ and $\varphi_L^{(2)} = \varphi_L(t_2)$ is considered. The explicit analytical formulas for determination of these coefficients are proposed. Linearized version of the nonlinear model (8) is analyzed in the final Sect. 5. Here the distinguished features and differences of the identification (optimal control) and inverse coefficient problems are discussed. In particular, it is shown that the measured output (flux) data $\varphi_L(t)$ can not be given arbitrarily.

2 Relationships between the flux and the current response in linear and nonlinear models

To derive the relationship between the flux and the current response for pure diffusive linear model, we use the analytical solution

$$u(x, t) = \frac{u_0}{z_r} erfc\left(\frac{x}{2\sqrt{Dt}}\right), \quad x > 0, \ t > 0 \tag{10}$$

of the parabolic problem (6). This solution, with formulas (4) and (5) implies:

$$\mathcal{I}_C(t) = \frac{n\mathcal{F}S_e u_0}{z_r}\sqrt{\frac{D}{\pi t}}, \quad t > 0, \tag{11}$$

where $\mathcal{I}_C(t)$ is the Cottrellian.

This results, derived in [5], is an analytical approval of well-known Cottrell's result [12].

Let us calculate the flux at the electrode surface $x = 0$. We have

$$[u_x(x, t)]_{x=0} = \frac{u_0}{z_r}\frac{\partial}{\partial x}\left[1 - \frac{2}{\sqrt{\pi}}\int_0^{\frac{x}{2\sqrt{Dt}}} e^{-y^2}dy\right]_{x=0}, \quad t > 0,$$

and hence

$$u_x(0, t) = -\frac{u_0}{z_r} \frac{1}{\sqrt{\pi D t}}, \quad t > 0.$$

According to the definition $\varphi_L(t) := -D u_x(0, t)$ of the left flux, this implies:

$$\varphi_L(t) = \frac{u_0}{z_r} \sqrt{\frac{D}{\pi t}}, \quad t > 0. \tag{12}$$

Hence for pure diffusive linear ion transport the analytical formula for the flux is defined by formula (12).

Comparing formulas (11) and (12) we obtain the following relationship between the flux $\varphi_L(t)$ and the current response $\mathcal{I}_C(t)$ (Cottrellian)

$$\varphi_L(t) = (n\mathcal{F}S_e)^{-1}\mathcal{I}_C(t), \quad t > 0, \tag{13}$$

in the case of the pure diffusive linear model (6). This formula, with (5), means that the flux is the derivative of the scaled total charge:

$$\varphi_L(t) = q'(t), \quad t > 0. \tag{14}$$

Note that the same result can be obtained by integrating Eq. (6) on $(0, \infty)$, and then taking into account the condition $u_x(\infty, t) = 0$.

Let us consider now the diffusive-convective linear model (7). To derive the relationship between the flux and the current response, we use the analytical formula for the solution $u(x, t)$ of problem (7), given in [13]:

$$u(x, t) = \frac{u_0 x}{2z_r\sqrt{\pi D}} \exp\left(\frac{c}{2D}x - \frac{c^2}{4D}t\right) \int_0^t \exp\left(\frac{x^2}{4D(t-\tau)}\right)$$

$$\times \exp\left(\frac{c^2\tau}{4D}\right) \frac{d\tau}{(t-\tau)^{3/2}}, \quad x, t > 0. \tag{15}$$

From this formula we conclude $u(x, t) \to 0$, $u_x(x, t) \to 0$, as $x \to \infty$. Now we integrate Eq. (7) on $(0, \infty)$:

$$\int_0^\infty u_t(x, t)dx = D \int_0^\infty u_{xx}(x, t)dx - v \int_0^\infty u_x(x, t)dx, \quad t > 0.$$

On the left hand side we use Eqs. (4) and (5), and on the right hand side we use the boundary conditions $u_x(0, t) = u_0/z_r$, $u(\infty, t) = 0$, $u_x(\infty, t) = 0$. Then we get:

$$\varphi_L(t) = (n\mathcal{F}S_e)^{-1}\mathcal{I}(t) - \frac{u_0}{z_r}v. \tag{16}$$

Comparison with formula (13) shows contribution of the convection term $-vu_x$ in Eq. (7) to the relationship between the flux and the current response, as the second term on the right hand side of (16).

Since the diffusive-convective linear model (7) is most idealized one, the real contribution of the migration in the ion transport problem can not be observed from relationship (16). To investigate this phenomenon we need to analyze the nonlinear model (8). Let us integrate Eq. (8) on $(0, \infty)$:

$$\int_0^\infty u_t(x, t)dx = \int_0^\infty (g(u)u_x)_x(x, t)dx + \frac{1}{z_r}q'(t)\int_0^\infty u_x(x, t)dx. \qquad (17)$$

Here we used the form of $h(u)$, given by (9), which follows from the assumption (A2). The left hand side integral is the function $q'(t)$, by definition (4). Calculating the integrals on the right hand side we conclude

$$q'(t) = g(u(\infty, t))u_x(\infty, t) - g(u(0, t))u_x(0, t)$$
$$+\frac{1}{z_r}q'(\tau)[u(\infty, t) - u(0, t)], \quad t > 0.$$

We use here the conditions $u(\infty, t) = u_x(\infty, t) = 0$, $u_x(0, t) = 1$, and also the definition $\varphi_L(t) := -g(u(0, t))u_x(0, t) = g(1)u_x(0, t) = u_x(0, t)/\kappa$ of the flux (due to (9), $g(u(0, t)) = g(1) = 1/\kappa$). The diffusivity ratio $\kappa = D_0/D_r$ has the property $\kappa = z_r/z_0 = D_0/D_r$, by the assumption (A1). Then we get

$$q'(t) = \varphi_L(t) - \frac{1}{z_r}q'(\tau), \quad t > 0,$$

and hence

$$\varphi_L(t) = \frac{1 + z_r}{z_r}q'(\tau), \quad t > 0. \qquad (18)$$

Therefore in the case of the nonlinear ion transport model (8) the relationship between the flux $\varphi_L(t)$ and current response $\mathcal{I}(t)$ is as follows:

$$\varphi_L(t) = \frac{1 + z_r}{n\mathcal{F}S_e z_r}\mathcal{I}(t), \quad t > 0. \qquad (19)$$

This result completely agrees with the result (13), obtained for the linear pure diffusive model in the sense that, *in both cases the flux is proportional to the current response*. From inverse problems point of view this result implies that giving as a measured output data $\varphi_L(t)$ means knowing the current response, and vice versa.

3 Determination of the unknown diffusion coefficient $D > 0$ in pure diffusive model

Consider *the inverse problem* of determining the unknown diffusion coefficient $D > 0$ in the linear model (6) from the measured output (flux) data $\varphi_L(t)$

$$\varphi_L(t) := -Du(0, t), \quad t > 0, \tag{20}$$

at the electrode surface $x = 0$. Although due to various distances between closest cations and anions in lattice, the diffusion coefficient usually depends on the space variable $x > 0$ (see [14]), as a first step, here the diffusion coefficient is assumed to be constant.

Since the diffusion coefficient $D > 0$ is a constant, the measured output data $\varphi_L(t)$ given for all $t > 0$ (or in some time interval $(0, T]$), is more than necessary. As we will see below, only one single data $\varphi_L(t_1)$ for some $t_1 > 0$ is enough to determine one-to-one the unknown diffusion coefficient $D > 0$.

Let us assume that the single data $\varphi_L^{(1)} := \varphi_L(t_1)$ is an experimentally given value of the flux at $t = t_1 > 0$. Then due to formula (12) we get

$$\frac{u_0}{z_r} \sqrt{\frac{D}{\pi t_1}} = \varphi_L^{(1)}.$$

Thus the unknown diffusion coefficient $D > 0$ in pure diffusive model (6) can be determined from the single data $\varphi_L^{(1)}$ by the formula:

$$D = \frac{z_r^2 \pi}{u_0} t_1 \varphi_L^{(1)}. \tag{21}$$

4 Simultaneous determination of unknown diffusion and convection coefficients in linear model (7)

Consider *the inverse problem* of determining the unknown diffusion and convection coefficients $D > 0$ and $c > 0$ in the linear model (7) from the measured output (flux) data. We will show that two experimentally given single data $\varphi_L^{(1)} := \varphi_L(t_1)$, $\varphi_L^{(2)} := \varphi_L(t_2)$, $t_2 > t_1 > 0$, is enough for determination of the unknown coefficients.

Using analytical formula (15) we calculate

$$u_x(0, t) = \frac{u_0}{2z_r \sqrt{\pi D}} \exp\left(-\frac{c^2}{4D} t\right), \quad t > 0.$$

Hence the theoretical value of the flux is as follows:

$$-Du_x(0, t) = \frac{u_0}{2z_r} \sqrt{\frac{D}{\pi}} \exp\left(-\frac{c^2}{4D} t\right), \quad t > 0.$$

Substituting here $t = t_1$ and then $t = t_2$, and taking into account the measured output data $\varphi_L^{(1)}$ and $\varphi_L^{(2)}$, we obtain the following nonlinear system of equations

$$\begin{cases} \frac{u_0}{2z_r} \sqrt{\frac{D}{\pi}} \exp\left(-\frac{c^2}{4D} t_1\right) = \varphi_L^{(1)}, \\ \frac{u_0}{2z_r} \sqrt{\frac{D}{\pi}} \exp\left(-\frac{c^2}{4D} t_2\right) = \varphi_L^{(2)}, \end{cases} \tag{22}$$

with respect to the unknown coefficients $D > 0$ and $c > 0$.

To solve this system, we introduce the parameter $\tau = t_2 - t_1 > 0$. Then the second equation of system (23) implies:

$$\varphi_L^{(1)} \exp\left(-\frac{c^2}{4D} \tau\right) = \varphi_L^{(2)}.$$

This equation yields:

$$-\frac{c^2}{4D} = \frac{1}{\tau} \ln \frac{\varphi_L^{(2)}}{\varphi_L^{(1)}}. \tag{23}$$

Substituting this in the first equation of system (22) we get:

$$\frac{u_0}{2z_r} \sqrt{\frac{D}{\pi}} \left(\frac{\varphi_L^{(2)}}{\varphi_L^{(1)}}\right)^{t_1/\tau} = \varphi_L^{(1)}.$$

Solving this equation with respect to the unknown diffusion coefficient $D > 0$ we find:

$$D = \pi \left[\frac{2z_r}{u_0} \left(\frac{\varphi_L^{(1)}}{\varphi_L^{(2)}}\right)^{t_1/\tau} \varphi_L^{(1)} \right]^2. \tag{24}$$

Having $D > 0$, we may find the unknown convection coefficient $c > 0$ from Eq. (23):

$$c = 2 \left(\frac{D}{\tau} \ln \frac{\varphi_L^{(1)}}{\varphi_L^{(2)}} \right)^{1/2}. \tag{25}$$

Formulas (24, 25) define one-to-one the unknown diffusion and convection coefficients from the measured output data $\varphi_L^{(1)}$ and $\varphi_L^{(2)}$.

5 Identification and inverse problems for linearized ion transport problem

Under the above assumptions (A1, A2), we may rewrite the nonlinear identification problem (8) as follows:

$$
\begin{cases}
u_t = (g(u)u_x)_x + q'(t)u_x/z_r, & x > 0, \ t > 0, \\
u(x,0) = 0, & x > 0, \\
u(0,t) = 1, & t > 0, \\
q(t) = \int_0^\infty u(x,t)dx, & t \geq 0,
\end{cases}
\tag{26}
$$

taking into account the form of the coefficient $h(u)$, given by (9). Linearizing the nonlinear Eq. (26), as $u_t^{(n)} = (g(u^{(n-1)})u_x^{(n)})_x + q'(t)u_x^{(n)}/z_r$, and assuming $v(x,t) = u^{(n)}(x,t)$, we obtain the following linearized ion transport problem:

$$
\begin{cases}
v_t = (D(x,t)v_x)_x + q'(t)v_x/z_r, & x > 0, \ t > 0, \\
v(x,0) = 0, & x > 0, \\
v(0,t) = 1, & t > 0, \\
q(t) = \int_0^\infty v(x,t)dx, & t \geq 0.
\end{cases}
\tag{27}
$$

This is a special case of the more general optimal control problem

$$
\begin{cases}
w_t = (D(x,t)w_x)_x + v(t)w_x, & x > 0, \ t > 0, \\
w(x,0) = 0, & x > 0, \\
w(0,t) = 1, & t > 0, \\
v(t) = \int_0^\infty w(x,t)dx, & t \geq 0.
\end{cases}
\tag{28}
$$

Here the coefficient $v(t)$ can be treated as a control. Note that such kind of optimal control problems, related to contaminant flow have also been considered in [15,16]. These problems are defined to be nonlocal nonlinear identification problems for parabolic equations.

To analyze this problem, let us integrate Eq. (28) on $(0, \infty)$:

$$
\int_0^\infty w_t(x,t)dx = \int_0^\infty (D(x,t)w_x(x,t))_x dx + v(t) \int_0^\infty w_x(x,t)dx, \quad t > 0.
$$

Taking into account here the additional nonlocal condition for the coefficient $v(t)$, the boundary condition $w(0,t) = 1$, compatibility condition $v(0) = 0$ and the assumption (A1) we get:

$$
v'(t) = -D(0,t)w_x(0,t) - v(t), \quad t > 0.
\tag{29}
$$

By the definition $\varphi_L(t) := -D(0, t)w_x(0, t)$ of the left flux, and by the initial condition $w(x, 0) = 0$ (which implies $v(0) = 0$), this leads to the following Cauchy problem:

$$\begin{cases} v'(t) + v(t) = \varphi_L(t), & t > 0. \\ v(0) = 0. \end{cases} \tag{30}$$

Thus we get the following necessary condition for optimality of the pair $\langle w, v \rangle$.

Proposition 5.1 *If the pair $\langle w, v \rangle$ is the solution of the optimal control problem (28), then the convection coefficient $v(t) \in C[0, T]$, $T > 0$, satisfies the Cauchy problem (30).*

The solution of the Cauchy problem (30) can easily be derived via the left flux:

$$v(t) = \exp(-t) \int_0^t \exp(\tau)\varphi_L(\tau)d\tau, \quad t > 0. \tag{31}$$

Let us apply now this result to the special case, i.e. to the problem (27). In this case, we will have the equation

$$q'(t) = \varphi_L(t) - q'(t)/z_r, \quad t > 0,$$

instead of the differential Eq. (29). Hence the relationship between the flux and the scaled total charge for the linearized model (27) has the following form:

$$\varphi_L(t) = \frac{1 + z_r}{z_r}q'(t), \quad t > 0. \tag{32}$$

By definition (5), in terms of the current response $\mathcal{I}(t)$ this relationship is as follows:

$$\varphi_L(t) = \frac{1 + z_r}{z_r}q'(t), \quad t > 0. \tag{33}$$

This result completely agrees with relationship (18), obtained for the nonlinear ion transport model (26). Namely, independently on linear or nonlinear ion transport models, the relationships between the flux and the current response/scaled total charge are same. Moreover, the above results show that, in presence of the nonlocal condition (2), the additional condition $\varphi_L(t) := -(Du_x)_{x=0}$, with the measured output data $\varphi_L(t)$, has no additional information, since the theoretical value of the flux $\varphi_L(t)$ can be determined via the scaled total charge, by (32). Therefore, an inverse problem of determining the unknown diffusion coefficient $D(u)$ (or $D(x)$) from (26) or (27), by adding to these problems the condition $\varphi_L(t) := -(Du_x)_{x=0}$, has no sense.

6 Conclusions

Optimal control and inverse problems related to determination of unknown diffusion or/and convection coefficients for linear and nonlinear ion transport models are studied. For all considered models relationships between the flux and the current response/scaled total charge are established. For simplest linear models explicit analytical formulas between inputs (diffusion or/and convection coefficients) and output (measured flux data) are derived. The presented analytical solutions and other qualitative results can be used as an important tool in numerical simulations. Numerical approach for coefficient inverse problems, in the case of variable coefficients, will be presented in next study.

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