

DI-ELECTRODE OF TRANSFER AFFECTED BY DIFFUSION AT ITIES. EXPRESSIONS FOR g_{ITIES}^m AND i^m IN POTENTIAL-DEPENDENT KINETIC CONSTANTS. I. TRANSFER OF TWO IONS OF SAME CHARGE ACROSS ITIES

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abstract: This paper deals with two-ion (of same charge) transfer across the ITIES under the “mixed potential” (i.e., in open circuit) condition. A transfer affected by diffusion is considered and the expressions for g_{ITIES}^m (the “mixed potential” of the ITIES) and for i^m (the individual current density at “mixed potential”) are deduced as functions of potential-dependent kinetic constants. As concerns the relative magnitude of the exchange current densities of the two transferred ions, three limiting cases are discussed.

Introduction

Transfer of only one ionic species across ITIES has been modelled from an electrodic viewpoint [1-8] as a charge transfer reaction across the electrochemical double layer existing in the interphase region.

ITIES together with the transfer of only one ionic species, well-known in classical electrode kinetics as single-electrode, was denoted [8] as single-electrode of transfer. Of course, if two ionic species pass across the ITIES a di-electrode of transfer appears [9-14]. The electrode kinetics of the single-electrode of transfer [8] and of the di-electrode of transfer [9-14] existing at ITIES, in steady-state conditions, under a combined overpotential of charge transfer and diffusion [8] or under a pure charge transfer overpotential [9, 11, 13] or under a pure diffusion overpotential [10, 12, 14] was theoretically studied, both for two ions of the opposite charge [9, 10, 13, 14] and for two ions of the same charge [11, 12].

At the same time, the expressions for the “mixed potential”, g_{ITIES}^m , and for the “exchange mixed” current density at “mixed potential”, i^m , were deduced using both the traditional quantities of electrode kinetics [8-12] and the traditional quantities of chemical kinetics.

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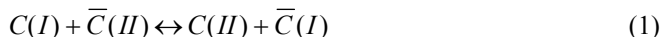
In the present paper we discuss the limiting case of a di-electrode of transfer affected by diffusion (under diffusion control) for two ions of same charge which are transferable across the ITIES.

Theoretical

Transfer affected by diffusion

In a previous paper [14] it has been analysed the case of a di-electrode of transfer unaffected by diffusion (in fact, under charge transfer control). This paper deals with the case of a di-electrode affected by diffusion (controlled by diffusion). At least one of two diffusion processes is here rate-determining step.

For two cations C (initially in phase I only) and \bar{C} (initially in phase II only) the di-electrode of transfer can be modelled by the following equivalent reaction:



which occurs under the thermodynamic condition $\Delta G_{tr,C}^{o,I \rightarrow II} < 0$, $\Delta G_{tr,\bar{C}}^{o,I \rightarrow II} > 0$ leading to the following condition concerning the individual reversible electrode potentials:

$$g_{ITIES,C}^{rev}(t) \leq g_{ITIES}^m(t) \leq g_{ITIES,\bar{C}}^{rev}(t) \quad (2)$$

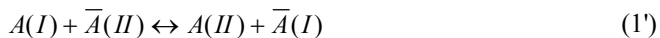
Under closed circuit condition (i.e., under a nonzero polarisation, $P \neq 0$ or $g \neq g_{ITIES}^m(t)$) the current density is given by:

$$i = \frac{\bar{k}_{C,a} a_C^I - \bar{k}_{C,c} a_C^{II}}{1 + \frac{\bar{k}_{C,a}}{i_{C,l}^I} a_C^I - \frac{\bar{k}_{C,c}}{i_{C,l}^{II}} a_C^{II}} + \frac{\bar{k}_{\bar{C},a} a_{\bar{C}}^I - \bar{k}_{\bar{C},c} a_{\bar{C}}^{II}}{1 + \frac{\bar{k}_{\bar{C},a}}{i_{\bar{C},l}^I} a_{\bar{C}}^I - \frac{\bar{k}_{\bar{C},c}}{i_{\bar{C},l}^{II}} a_{\bar{C}}^{II}} \quad (3)$$

and under open circuit condition (i.e., under a zero polarisation, $P = 0$ or $g = g_{ITIES}^m(t)$) when the net (total) current density is zero but each individual current density (each electrode reaction component of the di-electrode of transfer has associated its own individual current density), at "mixed potential", are equal with each other and with the i^m quantity, designed here as "exchange mixed" current density:

$$i^m = i_C^m = -i_{\bar{C}}^m = \frac{k_{C,a} a_C^I - k_{C,c} a_C^{II}}{1 + \frac{k_{C,a}}{i_{C,l}^I} a_C^I - \frac{k_{C,c}}{i_{C,l}^{II}} a_C^{II}} = - \frac{k_{\bar{C},a} a_{\bar{C}}^I - k_{\bar{C},c} a_{\bar{C}}^{II}}{1 + \frac{k_{\bar{C},a}}{i_{\bar{C},l}^I} a_{\bar{C}}^I - \frac{k_{\bar{C},c}}{i_{\bar{C},l}^{II}} a_{\bar{C}}^{II}} \quad (4)$$

For two anions A (initially in phase I only) and \bar{A} (initially in phase II only) the di-electrode of transfer can be modelled by the following equivalent reaction:



which occurs under the thermodynamic condition $\Delta G_{tr,A}^{o,I \rightarrow II} < 0$, $\Delta G_{tr,A}^{o,I \rightarrow II} > 0$ leading to the following condition concerning the individual reversible electrode potentials:

$$g_{ITIES,\bar{A}}^{rev}(t) \leq g_{ITIES}^m(t) \leq g_{ITIES,A}^{rev}(t) \quad (2)$$

Under closed circuit condition (i.e., under a nonzero polarisation, $P = 0$ or $g = g_{ITIES}^m(t)$) the current density is given by:

$$i = \frac{\bar{k}_{A,a} a_A^{II} - \bar{k}_{A,c} a_A^I}{1 + \frac{\bar{k}_{A,a}}{i_{A,l}^{II}} a_A^{II} - \frac{\bar{k}_{A,c}}{i_{A,l}^I} a_A^I} + \frac{\bar{k}_{\bar{A},a} a_A^{II} - \bar{k}_{\bar{A},c} a_A^I}{1 + \frac{\bar{k}_{\bar{A},a}}{i_{A,l}^{II}} a_A^{II} - \frac{\bar{k}_{\bar{A},c}}{i_{A,l}^I} a_A^I} \quad (3)$$

and under open circuit condition (i.e., under a zero polarisation, $P \neq 0$ or $g \neq g_{ITIES}^m(t)$) when the net (total) current density is zero but each individual current density (each electrode reaction component of the di-electrode of transfer has associated its own individual current density), at "mixed potential", are equal with each other and with the i^m quantity, designed here as "exchange mixed" current density:

$$i^m = i_A^m = -i_{\bar{A}}^m = \frac{k_{A,a} a_A^{II} - k_{A,c} a_A^I}{1 + \frac{k_{A,a}}{i_{A,l}^{II}} a_A^{II} - \frac{k_{A,c}}{i_{A,l}^I} a_A^I} = - \frac{k_{\bar{A},a} a_A^{II} - k_{\bar{A},c} a_A^I}{1 + \frac{k_{\bar{A},a}}{i_{A,l}^{II}} a_A^{II} - \frac{k_{\bar{A},c}}{i_{A,l}^I} a_A^I}$$

where the potential-dependent constants are defined in the following manner:

$$\begin{aligned} \bar{k}_{C,a} &= k'_{C,a} e^{\alpha_C z_C f g} & k'_{C,a} &= \frac{i_C^o}{a_C} e^{-\alpha_C z_C f g_{ITIES,C}^{rev}} \\ \bar{k}_{C,c} &= k'_{C,c} e^{-(1-\alpha_C) z_C f g} & k'_{C,c} &= \frac{i_C^o}{a_C} e^{(1-\alpha_C) z_C f g_{ITIES,C}^{rev}} \\ \bar{k}_{A,a} &= k'_{A,a} e^{-(1-\alpha_A) z_A f g} & k'_{A,a} &= \frac{i_A^o}{a_A} e^{(1-\alpha_A) z_A f g_{ITIES,A}^{rev}} \\ \bar{k}_{A,c} &= k'_{A,c} e^{\alpha_A z_A f g} & k'_{A,c} &= \frac{i_A^o}{a_A} e^{-\alpha_A z_A f g_{ITIES,A}^{rev}} \\ k_{C,a} &= k'_{C,a} e^{\alpha_C z_C f g^m} & k_{C,c} &= k'_{C,c} e^{-(1-\alpha_C) z_C f g^m} \\ k_{A,a} &= k'_{A,a} e^{-(1-\alpha_A) z_A f g^m} & k_{A,c} &= k'_{A,c} e^{\alpha_A z_A f g^m} \end{aligned}$$

and in analogue manner for the cation \bar{C} and for anion \bar{A} .

Case of a pair of transferable cations

Case a

If the two electrode reactions, component of the transfer di-electrode, modelling the cations transfer of C and \bar{C} , occur under the following conditions:

- for C : fast charge transfer, slow diffusion:

$$\eta_C > 0; \quad k_{C,a} a_C^I \gg i_{C,l}^I; \quad i_{C,l}^{II} \ll i_{C,l}^I e^{-z_C f \eta_C} \quad (5a)$$

- for \bar{C} : fast charge transfer, slow diffusion:

$$\eta_{\bar{C}} < 0; \quad k_{\bar{C},c} a_{\bar{C}}^{II} \gg i_{\bar{C},l}^{II}; \quad i_{\bar{C},l}^{II} \gg i_{\bar{C},l}^I e^{-z_{\bar{C}} f \eta_{\bar{C}}} \quad (5b)$$

then, taking account of conditions (5a) and (5b) as well as the expressions of kinetic constants, the equations (3) and (4) become:

$$i = i_{C,l}^I \left(1 - \frac{\bar{k}_{C,c} a_{\bar{C}}^{II}}{\bar{k}_{C,a} a_C^I} \right) - i_{\bar{C},l}^{II} \left(\frac{\bar{k}_{\bar{C},a} a_{\bar{C}}^I}{\bar{k}_{\bar{C},c} a_{\bar{C}}^{II}} - 1 \right) \quad (7a)$$

$$i^m = i_C^m = -i_{\bar{C}}^m = i_{C,l}^I \left(1 - \frac{k_{C,c} a_{\bar{C}}^{II}}{k_{C,a} a_C^I} \right) = i_{\bar{C},l}^{II} \left(\frac{k_{\bar{C},a} a_{\bar{C}}^I}{k_{\bar{C},c} a_{\bar{C}}^{II}} - 1 \right) \quad (8a)$$

To obtain the expression for the “mixed potential” one can use the second equality (8a) in the conditions $\eta_C \rightarrow 0$ and $\eta_{\bar{C}} \rightarrow 0$. Taking also into account the expressions of potential-dependent kinetic constants, after several mathematical calculations, one gets the expression:

$$g_{ITIES}^m = \frac{z_C i_{C,l}^I g_{ITIES,C}^{rev} - z_{\bar{C}} i_{\bar{C},l}^{II} g_{ITIES,\bar{C}}^{rev}}{z_C i_{C,l}^I - z_{\bar{C}} i_{\bar{C},l}^{II}} \quad (9a)$$

g_{ITIES}^m being somewhere between the two individual reversible electrodes potential at enough distance from each of them.

Using any of the expressions of i^m , given in (8a), and the “mixed potential” expression (9a), it results the individual expression of the current density at the “mixed potential”:

$$i^m = -z_C z_{\bar{C}} f i_{C,l}^I i_{\bar{C},l}^{II} \frac{g_{ITIES,\bar{C}}^{rev} - g_{ITIES,C}^{rev}}{z_C i_{C,l}^I - z_{\bar{C}} i_{\bar{C},l}^{II}} \quad (10a)$$

where i^m is, obviously, a positive quantity.

Case a'

If initially \bar{C} is only in phase I, (being transferred under a positive individual overpotential $\eta_{\bar{C}} > 0$) and C is only in phase II (being transferred under a negative individual

overtension $\eta_C < 0$), then the equations (5a)-(10a) are still valid but it is necessary to interchange C and \bar{C} . Obviously the transfer of \bar{C} takes place under condition (5a), with \bar{C} in place of C and the transfer of C takes place under condition (6a), with C in place of \bar{C} . The conditions $\eta_C \rightarrow 0$ and $\eta_{\bar{C}} \rightarrow 0$ continue keeping their role.

Case b

If the two electrode reactions, component of the transfer di-electrode, modelling the cations transfer of C and \bar{C} , occur under the following conditions

- for C : fast charge transfer, slow diffusion:

$$\eta_C \geq 0; \quad k_{C,a} a_C^I \gg i_{C,l}^I; \quad i_{C,l}^{II} \ll i_{C,l}^I e^{-z_C f \eta_C} \quad (5b)$$

- for \bar{C} : slow charge transfer, fast diffusion:

$$\eta_{\bar{C}} < 0; \quad k_{\bar{C},c} a_{\bar{C}}^{II} \ll i_{\bar{C},l}^{II}; \quad k_{\bar{C},a} a_{\bar{C}}^I \ll i_{\bar{C},l}^I; \quad k_{\bar{C},a} < k_{\bar{C},c} \quad (6b)$$

then the equations (3) and (4) become:

$$i = i_{C,l}^I \left(1 - \frac{\bar{k}_{C,c} a_{\bar{C}}^{II}}{\bar{k}_{C,a} a_C^I} \right) - \bar{k}_{\bar{C},c} a_{\bar{C}}^{II} \quad (7b)$$

$$i^m = i_C^m = -i_{\bar{C}}^m = i_{C,l}^I \left(1 - \frac{k_{C,c} a_{\bar{C}}^{II}}{k_{C,a} a_C^I} \right) = k_{\bar{C},c} a_{\bar{C}}^{II} \quad (8b)$$

From the first equality (8b) of i^m and the condition $\frac{i^m}{i_{C,l}^I} \rightarrow 0$ one gets the "mixed potential" expression; this g_{ITIES}^m will follow the behaviour of the individual reversible electrode potential of the cation C , $g_{ITIES,C}^{rev}$, therefore of the ion which is faster transferable in the charge transfer step:

$$g_{ITIES}^m \cong g_{ITIES,C}^{rev} \quad (9b)$$

To obtain the expression for the individual current density from the "mixed potential" one can use the second expression of i^m under the assistance of the individual reversible electrode potential (due to the action of the equation (9b)); one gets:

$$i^m = k'_{\bar{C},c} \left(\frac{k'_{C,c}}{k'_{C,a}} \right)^{\sigma_{\bar{C}}} \left(\frac{a_{\bar{C}}^{II}}{a_C^I} \right)^{\sigma_{\bar{C}}} a_{\bar{C}}^I \quad (10b)$$

where:

$$\sigma_{\bar{C}} = \frac{-(1 - \alpha_{\bar{C}}) z_{\bar{C}}}{z_C} \quad (11b)$$

Case b'

If initially \bar{C} is only in phase I, (being transferred under a positive individual overtension $\eta_{\bar{C}} > 0$) and C is only in phase II (being transferred under a negative individual overtension $\eta_C < 0$), then the equations (5b)-(10b) are still valid but it is necessary to interchange C and \bar{C} . Obviously the transfer of \bar{C} takes place under condition (5b), with \bar{C} in place of C and the transfer of C takes place under condition (6b), with C in place of \bar{C} . The "mixed potential" will follow closely the behaviour of the individual reversible potential of \bar{C} , because its charge transfer step is fast and its diffusion step is slow.

Case c

If the two electrode reactions, component of the transfer di-electrode, modelling the cations transfer of C and \bar{C} , occur under the following conditions

- for C : slow charge transfer, fast diffusion:

$$\eta_C > 0; \quad k_{C,a} a_C^I \ll i_{C,l}^I; \quad k_{C,e} a_C^{II} \ll i_{C,l}^{II}; \quad k_{C,a} > k_{C,e} \quad (5c)$$

- for \bar{C} : fast charge transfer, slow diffusion:

$$\eta_{\bar{C}} \leq 0; \quad k_{\bar{C},e} a_{\bar{C}}^{II} \gg i_{\bar{C},l}^{II}; \quad i_{\bar{C},l}^{II} \gg i_{\bar{C},l}^I e^{-z_{\bar{C}} f \eta_{\bar{C}}} \quad (6c)$$

then the equations (3) and (4) become:

$$i = \bar{k}_{C,a} a_C^I - i_{\bar{C},l}^{II} \left(\frac{\bar{k}_{\bar{C},a} a_{\bar{C}}^I}{\bar{k}_{\bar{C},e} a_{\bar{C}}^{II}} - 1 \right) \quad (7c)$$

$$i^m = i_C^m = -i_{\bar{C}}^m = k_{C,a} a_C^I = i_{\bar{C},l}^{II} \left(\frac{k_{\bar{C},a} a_{\bar{C}}^I}{k_{\bar{C},e} a_{\bar{C}}^{II}} - 1 \right) \quad (8c)$$

From the second equality (8c) of i^m and the condition $\frac{i^m}{i_{\bar{C},l}^{II}} \rightarrow 0$ one gets the "mixed potential" expression; this g_{ITIES}^m will follow the behaviour of the individual reversible electrode potential of the cation \bar{C} , $g_{ITIES,\bar{C}}^{rev}$, therefore of the ion which is faster transferable in the charge transfer step:

$$g_{ITIES}^m \cong g_{ITIES,\bar{C}}^{rev} \quad (9c)$$

To obtain the expression for the individual current density from the "mixed potential" one can use the first expression of i^m under the assistance of the individual reversible electrode potential (due to the action of the equation (9c)); one gets:

$$i^m = k'_{C,a} \left(\frac{k'_{\bar{C},a}}{k'_{\bar{C},c}} \right)^{\sigma_C} \left(\frac{a_{\bar{C}}^{II}}{a_{\bar{C}}^I} \right)^{\sigma_C} a_C^I \quad (10c)$$

where:

$$\sigma_C = \frac{\alpha_C z_C}{z_{\bar{C}}} \quad (11c)$$

Case c'

If initially \bar{C} is only in phase I, (being transferred under a positive individual overpotential $\eta_{\bar{C}} > 0$) and C is only in phase II (being transferred under a negative individual overpotential $\eta_C < 0$), then the equations (5c)-(10c) are still valid but it is necessary to interchange C and \bar{C} . Obviously the transfer of \bar{C} takes place under condition (5c), with \bar{C} in place of C and the transfer of C takes place under condition (6c), with C in place of \bar{C} . The "mixed potential" will follow closely the behaviour of the individual reversible potential of C , because its charge transfer step is fast and its diffusion step is slow.

Case of a pair of transferable anions

Case a

The anion A from phase I is transferred under $\eta_A < 0$ and the anion \bar{A} from phase II under the condition $\eta_{\bar{A}} > 0$. Both anions pass the ITIES practically out of the equilibrium state, the charge transfer step being fast while the diffusion step is slow (the "mixed potential" is somewhere between the two individual reversible electrode potentials):

$$g_{ITIES}^m = \frac{z_{\bar{A}} i_{\bar{A},I}^{II} g_{ITIES,\bar{A}}^{rev} + z_A i_{A,I}^{II} g_{ITIES,A}^{rev}}{z_{\bar{A}} i_{\bar{A},I}^{II} + z_A i_{A,I}^{II}} \quad (12a)$$

$$i^m = -z_{\bar{A}} z_A f_{\bar{A},I}^{II} i_{A,I}^{II} \frac{g_{ITIES,A}^{rev} - g_{ITIES,\bar{A}}^{rev}}{z_{\bar{A}} i_{\bar{A},I}^{II} + z_A i_{A,I}^{II}} \quad (13a)$$

Case a'

If initially \bar{A} is only in phase I, (being transferred under a positive individual overpotential $\eta_{\bar{A}} < 0$) and A is only in phase II (being transferred under a negative individual overpotential $\eta_A > 0$), then the equations (12a) and (13a) are still valid but it is necessary to interchange A with \bar{A} . Each of two anions will pass the ITIES out of its own individual reversible electrode potential, enough far from it.

Case b

If initially A is only in phase I, (being transferred under a positive individual overpotential $\eta_A \geq 0$) and \bar{A} is only in phase II (being transferred under a negative individual

overtension $\eta_{\bar{A}} < 0$), then A is transferable practically at its own equilibrium state (charge transfer step being fast while diffusion step is slow) and \bar{A} out of its own equilibrium electrode potential, enough far from it, under a very negative overtension (charge transfer step being slow while diffusion step is fast):

$$\mathcal{G}_{ITIES}^m \cong \mathcal{G}_{ITIES,A}^{rev} \quad (12b)$$

$$i^m = k'_{\bar{A},a} \left(\frac{k'_{A,a}}{k'_{A,c}} \right)^{\sigma_{\bar{A}}} \left(\frac{a_A^{II}}{a_A^I} \right)^{\sigma_{\bar{A}}} a_A^{II} \quad (13b)$$

Case b'

If initially \bar{A} is only in phase I, (being transferred under a positive individual overtension $\eta_{\bar{A}} \leq 0$) and A is only in phase II (being transferred under a negative individual overtension $\eta_A > 0$), then \bar{A} passes the ITIES practically at its own equilibrium state (charge transfer step being fast while diffusion step is slow) and A out of its own equilibrium electrode potential, enough far from it, under a very positive overtension (charge transfer step being slow while diffusion step is fast).

Case c

If initially A is only in phase I, (being transferred under a positive individual overtension $\eta_A < 0$) and \bar{A} is only in phase II (being transferred under a negative individual overtension $\eta_{\bar{A}} \geq 0$), then \bar{A} is transferable practically at its own equilibrium state (charge transfer step being fast while diffusion step is slow) and A out of its own equilibrium electrode potential, enough far from it, under a very negative overtension (charge transfer step being slow while diffusion step is fast):

$$\mathcal{G}_{ITIES}^m \cong \mathcal{G}_{ITIES,\bar{A}}^{rev} \quad (12c)$$

$$i^m = k'_{A,c} \left(\frac{k'_{\bar{A},a}}{k'_{\bar{A},c}} \right)^{\sigma_A} \left(\frac{a_A^{II}}{a_A^I} \right)^{\sigma_A} a_A^I \quad (13c)$$

Case c'

If initially \bar{A} is only in phase I, (being transferred under a negative individual overtension $\eta_{\bar{A}} < 0$) and A is only in phase II (being transferred under a positive individual overtension $\eta_A \geq 0$), then A passes the ITIES practically at its own equilibrium state (charge transfer step being fast while diffusion step is slow) and \bar{A} out of its own equilibrium electrode potential, enough far from it, under a very positive overtension (charge transfer step being slow while diffusion step is fast).

Conclusions

In this theoretical study the di-electrode of transfer affected by diffusion is analysed for two ionic species of same charge crossing the ITIES.

The equations describing the "mixed potential" and the individual current densities from the "mixed potential" show which are the quantities which can control or determine them in the open circuit condition, $i = 0$ and $g_{ITIES} = g_{ITIES}^m$.

The "mixed potential" will follow the better the individual reversible electrode potential of an ion which transfers faster in the charge transfer step the greater its exchange current density is in comparison with the other transferable ion.

Equation (3), in its specialised forms, could be used to obtain the individual current densities from the "mixed potential" either from the Tafel analyse of the polarisation curve or from the polarisation resistance determination. That is the situation for the *case a* and *case a'* when $P \rightarrow 0$ it results the expression for i^m . For the *case b* and *case b'*, as well as for *case c* and *case c'*, one can obtain the corresponding i^o quantity.

From the expression of g_{ITIES}^m one could notice that is the ion controlling its value and from the expression of i^m which are the kinetic parameters influencing its value.

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