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Variations of cell resistance and current distribution with current feeder configurations

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Two models of current feeder configurations for resistive electrodes are presented, one in which the feeders were connected at the same end of a pair of electrodes of a unit cell and one in which connection was made at opposite ends. Expressions for the cell resistance and the current distribution were derived for these two current feeder configurations on the assumption of a linear type of overpotential. The cell resistance in the 'opposite ends' configuration was larger than that for the 'same ends' arrangement. Conversely, the current distribution in the former was more uniform than that in the latter. The relation between the total cell resistance and the number of current feeders, n, was obtained. An increase in n led to a decrease in the resistive loss of the electrodes by an amount corresponding to $1/n^2$, irrespective of current feeder configuration, when the resistance of the electrode was not so great as that of the solution.

Nomenclature

a	linear overpotential coefficient, defined by Equation 10
b	linear overpotential coefficient, defined by Equation 10
C	constant
d	interelectrode distance
d_{al}	d_{cl}, d_m thickness of the anolyte, catholyte or membrane
H	height of the cell
h	half distance between two adjacent current feeders
I	current fed to the anode per the unit cell
i	current density flowing in the solution
i_{av}	average current density
i_{max}	maximum current density
n	number of current feeders at the anode, defined by $H/(2h)$
p	ratio of the cathode to anode resistance, defined by Equation 24
r	cell resistance defined by Equation 15
r'	equivalent cell resistance calculated from the electric power on the assumption of uniform current distribution
r_t	total cell resistance
t_a, t_c	thickness of the anode or cathode
V	cell voltage
V_{eq}	open circuit inner potential difference between the anode and cathode
W	electric power
w	width of the electrode
x	axis normal to electrode surface
y	axis in the direction of electrode height
η	overpotential
θ	dimensionless parameter defined by Equation 14, which roughly expresses the ratio of electrode resistance to solution resistance
ϱ	average resistivity
ϕ	inner potential of the solution
ψ	inner potential of the electrode

Subscripts

a	anode
al	anolyte

c cathode
cl catholyte
m membrane

Superscript

T terminal

1. Introduction

In industrial electrolysis, an ohmic drop in the electrodes frequently lowers the electrolysis efficiency. Such a problem may be appreciable when the electrodes are made of poorly conducting materials, when they are thin and long, or when they contain voids for releasing gas bubbles. The resistive loss of the electrodes causes non-uniform current distribution and hence may be detrimental to product quality. In order to minimize the resistive loss of the electrodes it is necessry to design a proper size and position of the bus bar and current feeder. Several investigations have been directed towards the effects of electrode resistance, solution resistance and overpotential on the current distribution. Tobias and Wijsman [1] evaluated the effet of electrode resistance on current distribution in parallel plane electrode systems when the current was fed to the same ends of the electrodes. Ishizaka *et al.* [2] calculated the current distribution in coaxially arranged cylindrical electrode systems and verified it experimentally. Robertson [3] obtained the optimum current feeder arrangement for various combinations of anode, cathode and solution resistance. Scott [4] compared utilization factors for disc and square electrodes for various feeder arrangements. Vaaler [5] approximated cell and electrode resistance as a three-dimensional grid network and calculated the current distribution in the cell. Other reports on the resistive electrode problem have been summarized by Ibl [6].

Most of this work has been concentrated on the current distribution which is relevant to product quality. From an economic viewpoint, the cell resistance between two current feeders has to be taken into account for a cell design. No attention has been paid to the cell resistance in the resistive electrode problem. The relationship between the current distribution and the cell resistance is discussed in this paper. Two models of current feeder configurations are proposed, along with the Robertson model [3]. The cell resistances at the two models are calculated and compared.

2. Current feeder models

The cell treated here consists of two parallel plate electrodes and a separator. For a large scale electrolysis cell, current is fed to electrodes with the aid of conductive bus bars. There may be two

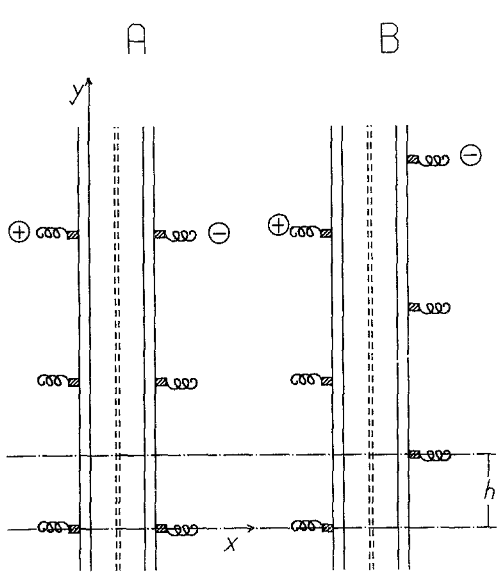

Fig. 1. Schematic diagram of electrolysis cells with (A) 'same ends' configuration and (B) 'opposite ends' configuration. The dashed lines denote the membrane. A part of the cell held between the two lines (–·–·–) is a unit cell.

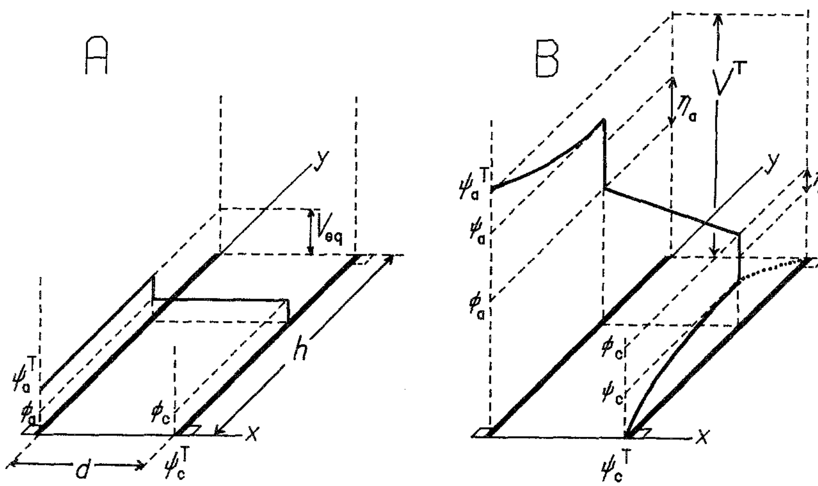

Fig. 2. Schematic diagram of potential distribution in a cell without flowing current (A) and with current (B) for the 'same ends' configuration. The dotted curve show potential distribution for the 'opposite ends' case.

types of current feeder configurations, as shown in Fig. 1A and 1B. The current feeders in Fig. 1A face each other while those in Fig. 1B are located alternately. The model cells are composed of combinations of unit cells with a half distance between two adjacent current feeders (Fig. 1). The unit cell in Fig. 1A has current feeders at the same end while that in Fig. 1B has current feeders at opposite ends [3].

The following assumptions are made.

(a) The electrodes, solutions and the separator have uniform resistivities.

(b) The electrodes are so thin in comparison with the length that current flows only in the direction of the y-axis within the electrode.

(c) The current in the solution flows perpendicularly to the electrodes. This assumption is valid when the interelectrode distance is smaller than the height of the cell [1, 7].

(d) Overpotential of a linear type is present at the interface between the solution and the electrodes.

(e) The bus bars are in equipotential and their thickness in the y-direction is negligibly small.

These assumptions are reasonable for common industrial electrolysis cells. In Fig. 2, a schematic diagram of the potential variation in the cell is illustrated three-dimensionally.

3. Derivation of the cell resistance

3.1. *Same ends*

When current flows from the anode to the cathode, the relationship between the current density and the inner potentials of the solution is given by

$$i\varrho d = \phi_a - \phi_c \tag{1}$$

with

$$\varrho d = \varrho_{al} d_{al} + \varrho_{cl} d_{cl} + \varrho_m d_m \tag{2}$$

Taking into account the current balance in the anode and cathode, gives, respectively

$$I = -(wt_a/\varrho_a)(d\psi_a/dy) + w\int_0^y i\,dy \tag{3}$$

$$I = (wt_c/\varrho_c)(d\psi_c/dy) + w\int_0^y i\,dy \tag{4}$$

The boundary conditions are given by

$$\psi_a = \psi_a^T \tag{5a}$$

$$\psi_c = \psi_c^T \tag{5b}$$

when $y = 0$, or

$$d\psi_a/dy = 0 \tag{6a}$$

$$d\psi_c/dy = 0 \tag{6b}$$

when $y = h$.
The terminal cell voltage, V^T, is expressed by

$$V^T = \psi_a^T - \psi_c^T \tag{7}$$

Subtracting Equation 3 from Equation 4 and integrating the resulting equation by use of Equation 5 gives

$$(t_c/\varrho_c)(\psi_c - \psi_c^T) + (t_a/\varrho_a)(\psi_a - \psi_a^T) = 0 \tag{8}$$

By referring to the schematic of the potential variation in Fig. 2, the relationship between the difference in the inner potentials of the electrodes and the difference in those of the solution is given by

$$\psi_a - \psi_c = \eta + V_{eq} + (\phi_a - \phi_c) \tag{9}$$

where η is overpotential of a linear type

$$\eta = \eta_a + \eta_c = a + (b_a + b_c)i \tag{10}$$

Inserting Equations 1 and 10 into Equation 9 yields

$$\psi_a - \psi_c = V_{eq} + a + i(\varrho d + b_a + b_c) \tag{11}$$

Differentiating Equation 3 with respect to y, eliminating i from Equation 11 and further eliminating ψ_c and ψ_c^T by use of Equations 7 and 8 gives

$$(\varrho d + b_a + b_c)\,d^2\psi_a/dy^2 = (\varrho_a/t_a + \varrho_c/t_c)(\psi_a - \psi_a^T) + (\varrho_a/t_a)(V^T - V_{eq} - a) \tag{12}$$

Solving Equation 12 and combining Equations 5 and 6 yields

$$\psi_a = \psi_a^T + [t_c\varrho_a/(t_c\varrho_a + t_a\varrho_c)](V^T - V_{eq} - a)[e^{\theta y/h}/(1 + e^{2\theta}) + e^{-\theta y/h}/(1 + e^{-2\theta}) - 1] \tag{13}$$

where

$$\theta = [(\varrho_a/t_a + \varrho_c/t_c)/(\varrho d + b_a + b_c)]^{1/2}h \tag{14}$$

Equation 13 represents the potential distribution at the anode [3].
The cell resistance is defined by

$$r = (V^T - V_{eq} - a)/I \tag{15}$$

Inserting Equation 3 for $y = 0$ into Equation 15, replacing ψ_a by Equation 13 and rearranging the resulting equation yields

$$r/[(\varrho d + b_a + b_c)/(hw)] = \theta/\tanh(\theta) \tag{16}$$

The current density on the anode flowing into the solution can be obtained by differentiating Equation 13 twice with respect to y. This gives

$$i/[I/(hw)] = \theta \cosh[\theta(1 - y/h)]/\sinh(\theta) \tag{17}$$

On the basis of assumption (c) from Section 2, this current distribution is the same as that on the cathode.

3.2. *Opposite ends*

When the anode feeder is located at $y = 0$ and the cathode feeder is at $y = h$, the current balance at the anode is given by Equation 3 and that at the cathode is expressed by

$$I = -(wt_c/\varrho_c)(d\psi_c/dy) + w\int_y^h i\,dy \tag{18}$$

The boundary conditions are given by

$$\psi_a = \psi_a^T \tag{19a}$$

$$d\psi_c/dy = 0 \tag{19b}$$

when $y = 0$, or

$$d\psi_a/dy = 0 \tag{20a}$$

$$\psi_c = \psi_c^T \tag{20b}$$

when $y = h$.

Adding Equation 3 to Equation 18 and integrating the resulting equation yields

$$(wt_a/\varrho_a)\psi_a + (wt_c/\varrho_c)\psi_c = -Iy + C_1 \tag{21}$$

where C_1 is a constant to be determined. Following the same procedure as the derivation of Equation 12 gives

$$(\varrho d + b_a + b_c)\,d^2\psi_a/dy^2 = (\varrho_a/t_a + \varrho_c/t_c)\psi_a + [\varrho_a\varrho_c/(t_a t_c)](Iy/w + C_1) - (V_{eq} + a)\varrho_a/t_a \tag{22}$$

The solution of Equation 22 is

$$\psi_a = C_2 \exp(-\theta y/h) + C_3 \exp(\theta y/h) - (Iy/w + C_1 - V_{eq} - a)/(1 + p) \tag{23}$$

with

$$p = \varrho_c t_a/\varrho_a t_c \tag{24}$$

where C_2 and C_3 are constants. C_2 and C_3 can be determined by use of Equations 19, 20 and 21, and C_2 is given by

$$C_2 = \{e^\theta(V^T - V_{eq} - a) - \{p\varrho_a Ih/[t_a(1 + p)w]\}[e^\theta + (1 + pe^\theta)/\theta]\}/\{2[p + \cos h(\theta)]\} \tag{25}$$

C_3 is expressed by Equation 25 in which θ is replaced by $-\theta$. C_1 can be determined from the following equation:

$$C_1 = (t_a/\varrho_a + t_c/\varrho_c)[C_2 + C_3 + (V_{eq} + a)/(1 + p) - \psi_a^T] \tag{26}$$

The expression for the cell resistance can be obtained by combination of Equation 3 (for $y = 0$), Equations 15 and 23. Then

$$r/[(\varrho d + b_a + b_c)/(hw)] = [\theta/(p + p^{-1} + 2)][\theta + 2/\sinh(\theta) + (p + p^{-1})/\tanh(\theta)] \tag{27}$$

When $p = 1$, Equation 27 is reduced to

$$r/[(\varrho d + b_a + b_c)/(hw)] = \theta^2/4 + (\theta/2)/\tanh(\theta/2) \tag{28}$$

The current density on the anode flowing into the solution is given by

$$i/[I/(hw)] = \{p\theta/[(p + 1)\sinh(\theta)]\}\{\cosh(\theta y/h) + (1/p)\cosh[\theta(y/h - 1)]\} \tag{29}$$

For $p = 1$, Equation 29 is reduced to

$$i/[I/(hw)] = (\theta/2)\cosh[\theta(y/h - 1/2)]\sinh(\theta/2) \tag{30}$$

4. Discussion

4.1. *Cell resistance*

In Fig. 3, variations of cell resistance with θ are plotted for several values of $p + p^{-1}$ for the case of the 'opposite ends' configuration (curves a–e) and for the case of the 'same ends' configuration (curve f). Curve a corresponds to the cell with 'opposite ends' connection in which the resistance of the anode is the same as that of the cathode. With increase in $p + p^{-1}$, r decreases. Equation 27 tends to Equation 16 as $p + p^{-1} \to \infty$, $p \to 0$ or $p \to \infty$. Hence the cell resistance with 'opposite ends' connection for $p \to 0$ or ∞ is the same as that with the 'same ends' connection. This fact indicates that the cell resistance is independent of the configuration of the current feeders when the resistance of one electrode is quite different from that of the other. From the comparison of these curves it is obvious that the resistive loss of the electrodes for the 'same ends' configuration is lower than that for the 'opposite ends' arrangement.

As values of θ tend to 0 for the 'same ends' configuration, Equations 16 and 27 approach unity, i.e. r approaches $(\varrho_a d_a + \varrho_c d_c + \varrho_m d_m)/(hw)$, which represents the sum of the solution resistance, membrane resistance and the resistance due to the overpotential. For large values of θ, Equation 16 is reduced to

$$r/[(\varrho d + b_a + b_c)/(hw)] = \theta \tag{31a}$$

or

$$r = [(\varrho_a/t_a + \varrho_c/t_c)(\varrho d + b)]^{1/2}/w \tag{31b}$$

This indicates that the cell resistance is expressed by the geometric mean of the electrode resistance and the solution. For extremely large values of θ, assumption (c) from Section 2 breaks down. However, assumption (c) and hence Equation 31b are valid for values of θ that satisfy the condition

$$\theta^4 < [(\varrho_a/t_a + \varrho_c/t_c)\, h/\varrho](h/d)^2,$$

according to [7].

It is interesting to estimate roughly the resistive loss of the electrodes by taking into account the electric power consumed in the anode, given by

$$W_a = \int_0^h [(wt_a/\varrho_a)(\mathrm{d}\psi_a/\mathrm{d}y)]^2[\varrho_a/(wt_a)]\,\mathrm{d}y \tag{32}$$

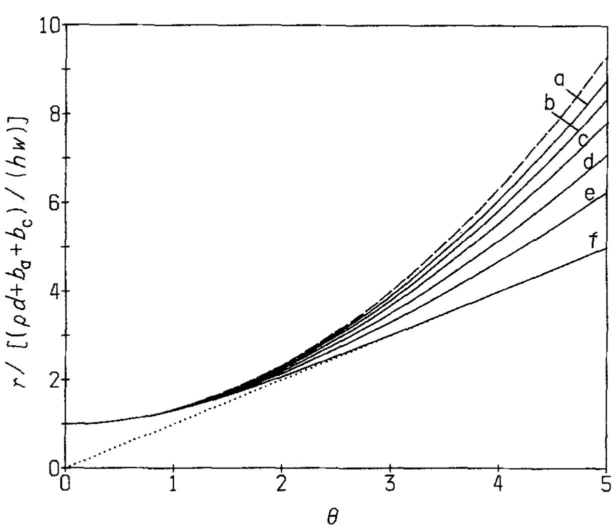

Fig. 3. Dependence of the dimensionless unit cell resistance on θ for $p + p^{-1}$ equal to: (a) 2 ($p = 1$); (b) 2.5 ($p = 2$ or 0.5); (c) 3.33 ($p = 3$ or 0.33); (d) 5.2 ($p = 5$ or 0.2); (e) 10.1 ($p = 10$ or 0.1); (f) ∞ ($p \to 0$ or $p \to \infty$).

Here, the term $(wt_a/\varrho_a)(\mathrm{d}\psi_a/\mathrm{d}y)$ is the current density flowing in the anode. It is assumed that the current density flowing out of the electrode to the solution is uniformly distributed. Inserting Equation 3 with a constant i into Equation 32 and carrying out the integration, yields

$$W_a = I^2(\varrho_a h/wt_a)/3 \tag{33}$$

W_a is equal to $I^2 r'_a$ if the current were to flow through the resistance r'_a. Then, r'_a is given by

$$r'_a = (\varrho_a h/wt_a)/3 \tag{34}$$

Following the same discussion as above, the electric power consumed in the cathode, r'_c, is given by

$$r'_c = (\varrho_c h/wt_c)/3 \tag{35}$$

Series connection of r'_a, r'_c and $(\varrho d + b_a + b_c)/(hw)$ yields

$$r' = r'_a + r'_c + (\varrho d + b_a + b_c)/(hw) = [(\varrho d + b_a + b_c)/(hw)](1 + \theta^2/3) \tag{36}$$

This is the cell resistance averaged through the electric power on the assumption of uniform current distribution. The dotted curve in Fig. 3 was calculated from Equation 36. It is very near to curve a, indicating that this approximation is good except for large values of θ. Indeed, the first two terms of the Taylor expansion of Equations 16 and 27 about $\theta = 0$ is identical to Equation 36. This approximate method may be applied to complicated electrode geometries. Since the current distribution has been assumed to be uniform in the derivation of Equation 36, the electric power given by Equation 33 is not minimum, and hence it follows that $r' > r$, as shown in Fig. 3.

The total cell resistances, r_t, can be given by a parallel combination of $2n$ unit cell resistances. The total cell resistances for the 'same ends' and the 'opposite ends' configurations are expressed respectively by

$$r_t = r/(2n) = [(\varrho d + b_a + b_c)/(Hw)][\theta/\tanh(\theta)] \tag{37}$$

$$r_t = [(\varrho d + b_a + b_c)/(Hw)][\theta/(p + p^{-1} + 2)][\theta + 2/\sinh(\theta) + (p + p^{-1})/\tanh(\theta)] \tag{38}$$

where θ is given by

$$\theta = [(\varrho_a/t_a + \varrho_c/t_c)/(\varrho d + b_a + b_c)]^{1/2} H/(2n) \tag{39}$$

The relationship between r_t and θ is the same as in Fig. 3. When $\theta < 1$, an increase in n leads to a decrease in the resistive loss of the electrodes by an amount corresponding to $1/n^2$, irrespectively of current feeder configuration, according to Equation 36. This relation holds well for $\theta > 1$ in the 'opposite ends' configuration when p is near unity. When $\theta > 2$ for the 'same ends' configuration, the cell voltage is inversely proportional to n.

4.2. *Relationships between the cell resistance and current uniformity*

One measure of the non-uniformity of the current distribution is the ratio of the maximum to the average current density, i.e. i_{max}/i_{av}. The inverse of this ratio has been termed an electrode utilization factor [3]. The average current is equal to $I/(hw)$. It is obvious that there is a maximum current density at $y = 0$ for the 'same ends' configuration. Then i_{max}/i_{av} is given by

$$i_{max}/i_{av} = \theta/\tanh(\theta) \tag{40}$$

The term on the right hand side is the same as the dimensionless cell resistance given by Equation 16. On the other hand, the maximum of the current density for the 'opposite ends' configurations occurs at $y = h$ when $p \geqq 1$. Replacing y by h in Equation 29 yields

$$i_{max}/i_{av} = p\theta[\cosh(\theta) + p^{-1}]/[(p + 1)\sinh(\theta)] \tag{41}$$

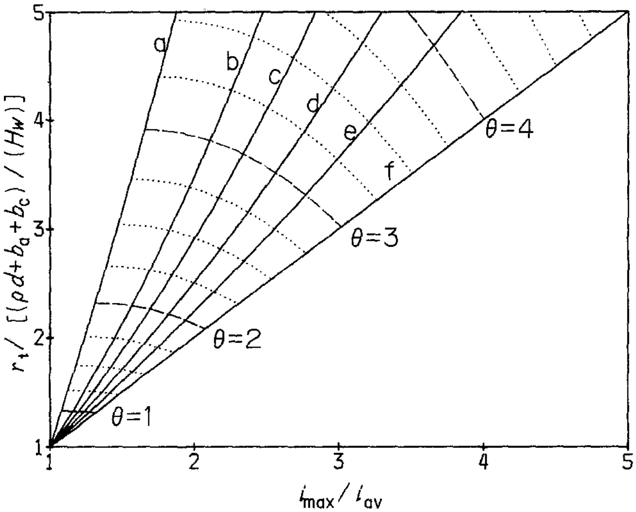

Fig. 4. Plots of the dimensionless total cell resistance against i_{max}/i_{av} for $p + p^{-1}$ equal to: (a) 2 ($p = 1$); (b) 2.5 ($p = 2$ or 0.5); (c) 3.33 ($p = 3$ or 0.33); (d) 5.2 ($p = 5$ or 0.2); (e) 10.1 ($p = 10$ or 0.1); (f) ∞ ($p \to 0$ or $p \to \infty$). The dashed and the dotted curves are contour lines for θ.

When values of $p + p^{-1}$ tends to infinity, Equation 41 approaches Equation 40, as for the case of the cell resistance.

In Fig. 4, values of the dimensionless total cell resistance expressed by Equations 37 and 38 are plotted against i_{max}/i_{av} for several values of p and θ. The curves in Fig. 4 show that the non-uniformity increases the cell resistance. At a given value of the dimensionless cell resistance, the non-uniformity increases with an increase in $p + p^{-1}$. At a given value of θ, the 'opposite ends' configuration with $p = 1$ has about one-third of the measure of the uniformity of the current distribution for the 'same ends' configuration. With an increase in n, or with a decrease in the sum of the resistances of the anode and the cathode, both the total cell resistance and the uniformity of the current distribution are improved. It is also possible to improve the current uniformity by taking p to be near unity. This diagram may be helpful for designing production-type electrolysis cells.

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