

Estimation of current bypass in a bipolar electrode stack from current-potential curves

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Received 24 July 1990; revised 3 September 1990

A simple method is proposed for the estimation of the current bypass from experimental current-potential ($i-U$) curves measured for a "bipolar reactor" and with a one-element cell of similar geometry. The model is valid only in the region where a linear $i-U$ relation is obtained.

Notation

F Faraday constant ($C\ mol^{-1}$)
 i_0 electrical feed current density ($A\ m^{-2}$)
 i_j current density in cell j ($A\ m^{-2}$)
 I_0 current (A)
 N number of cells
 P pressure ($N\ m^{-2}$)
 R gas constant ($J\ mol^{-1}\ K^{-1}$)
 R_e slope of the linear part of the $i-U$ relation for one element cell ($\Omega\ m^2$)

T temperature (K)
 U_0 intercept of the linear $i-U$ relation with U axis for one element cell (V)
 U_1 potential difference for one element cell (V)
 U_N potential difference for a bipolar electrode stack with N cells (V)
 U_j potential difference for cell j in the stack (V)
 \dot{V} experimental gas flow rate ($m^3\ s^{-1}$)
 \dot{V}_0 theoretical gas flow rate given by Relation (7) ($m^3\ s^{-1}$)
 ψ current bypass

1. Introduction

Direct and indirect electroorganic syntheses have been widely studied on the laboratory scale but only few processes have been applied on an industrial scale [1]. The main reason is the complex nature of the electrochemical reactor compared with the chemical reactor, which is usually a simple stirred tank.

A new industrial undivided "bipolar electrochemical reactor" of channel type with flat parallel electrodes has been developed in our laboratory and is now commercialized. Its design is very simple, it consists of a bipolar stack of 20-30 metallic sheets (electrodes) separated from each other by insulating plastic spacers at a fixed distance (2-8 mm). The electrode stack is introduced in the reactor body (made of polymer-coated steel) and the electrolyte is fed through the inlet cone and comes out through the outlet (Fig. 1). The main advantages of this electrochemical reactor are: simple and cheap construction, compact design with high specific electrode area, and possibility of using multiphase electrolytes (solid-liquid).

The main disadvantage of the reactor is the presence of parasitic electrical currents, or current bypass, at the lower and upper parts of the electrode stack. This results in an electrical efficiency loss, a non-uniform potential and current distribution, and consequently,

a poor selectivity, in addition to an increase in corrosion rates of electrodes.

Many investigators have treated the subject of "bipolar electrochemical reactors" using an electrical analogy [2-5], or by solving directly the Laplace equation with the finite element method or the finite difference method [6, 7]. Yet, in spite of the large number of papers relative to this field, the experimental verification of the proposed models is relatively limited and the studied cases contain relatively few experimental results.

In this paper a simple method is proposed for the estimation of the current bypass from experimental current-potential curves measured for a bipolar reactor and with a one element cell of similar geometry.

2. Theoretical treatment

For a bipolar electrode stack with N cells the applied potential difference between the two electrode feeders (U_N) is equal to the algebraic sum of the individual potential difference (U_j) for each cell in the stack.

$$U_N = \sum_{j=1}^{j=N} U_j \quad (1)$$

Supposing a linear current-potential relation for each cell in the stack we obtain

$$U_N = \sum_{j=1}^{j=N} (U_0 + R_e i_j) \quad (2)$$

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As U_0 and R_e are constant for a given electrode reaction and electrolyte conductivity

$$U_N = NU_0 + R_e \sum_{j=1}^{j=N} i_j \quad (3)$$

This relation is compared with that theoretically obtained when the current bypass is almost zero ($\psi = 0$)

$$NU_1 = NU_0 + NR_e i_0 \quad (4)$$

From Equations 3 and 4 and the definition of the current bypass (Equation 5).

$$\psi = \frac{Ni_0 - \sum_{j=1}^{j=N} i_j}{Ni_0} \quad (5)$$

We obtain the relation (Equation 6) which allows the estimation of current bypass from the current-potential curve.

$$\psi = \frac{NU_1 - U_N}{NU_1 - NU_0} \quad (6)$$

The graphical determination of current bypass is illustrated in Fig. 2.

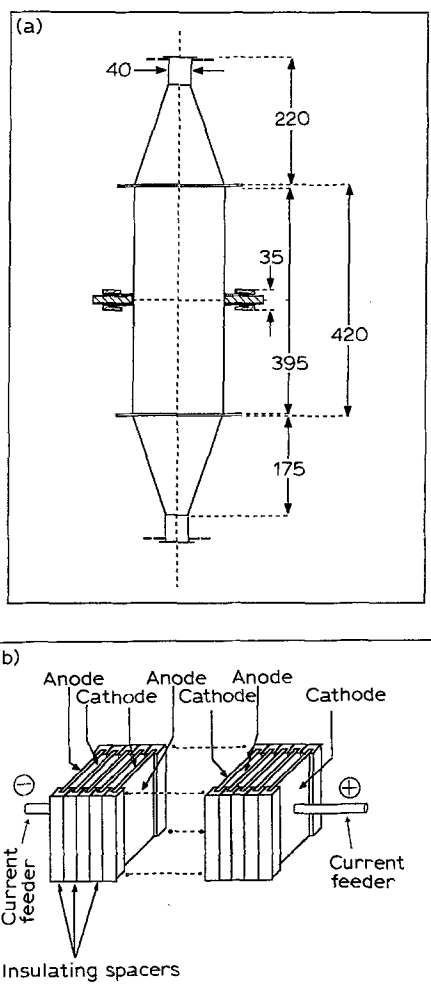


Fig. 1. Schematic representation of (a) the electrochemical reactor and (b) the bipolar electrode stack.

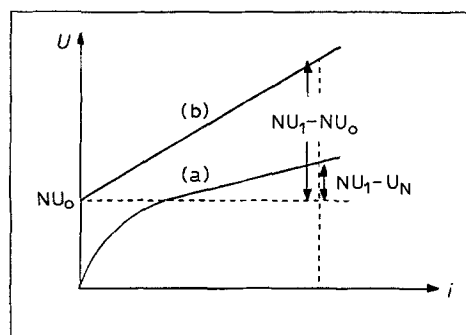


Fig. 2. Graphical determination of current bypass from current-potential curves: (a) for the bipolar stack with N cells, and (b) $[(i-U)$ for one element cell] $\times N$.

3. Experimental details

3.1. Equipment

The electrode stack (Fig. 1) contains 14 bipolar nickel electrodes ($396 \text{ mm} \times 17 \text{ mm} \times 2 \text{ mm}$) and two nickel current feeders; the electrodes are separated from each other by insulating spacers at a fixed distance (3.3 mm) giving a total anode (cathode) area of 0.1 m^2 for 15 cells stack.

Figure 3 is a schematic representation of the equipment used. The electrolyte was pumped through the electrode stack with an average velocity of 0.5 m s^{-1} . Its temperature was kept constant (50°C) using a thermostat (Lauda LTH 100) connected to a glass heat exchanger (Sovirel, 1 m^2). The concentration of electrolyte was analysed during the experiments by potentiometric titration.

3.2. Experimental determination of bypass current

The electrochemical reaction used for the determination of current bypass of the "bipolar reactor" is water

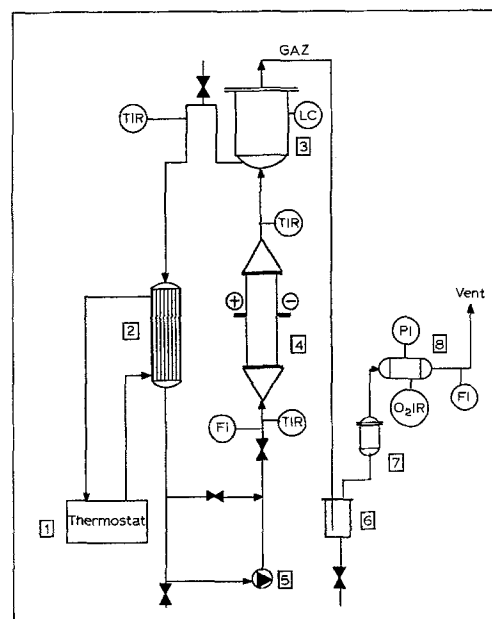


Fig. 3. Schematic presentation of the equipment used. Key: (1) thermostat, (2) heat exchanger, (3) gas-liquid separator, (4) electrochemical reactor, (5) circulation pump, (6) liquid separator, (7) alumina bed, and (8) gas analysis.

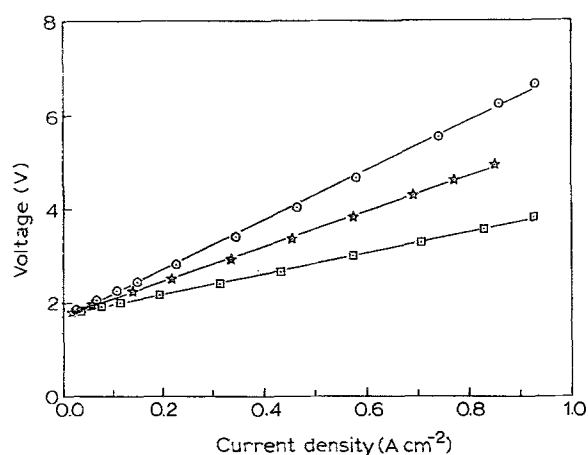
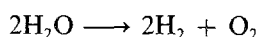


Fig. 4. Current-potential curves for one element cell at 50°C obtained at different KOH concentrations: (□) 1.0, (☆) 0.5 and (○) 0.325 mol dm⁻³.

electrolysis in alkaline medium (0.325, 0.50 and 1.0 M KOH).



Electrolysis was effected at constant current density and temperature, the evolved gas (H₂ + O₂) was dried on an alumina bed, its flow rate (\dot{V}) was measured using a bubble flowmeter. The current bypass (ψ) was calculated using the relation

$$\psi = \frac{\dot{V}_0 - \dot{V}}{\dot{V}_0}$$

where \dot{V}_0 is the theoretical volumetric flow rate of gas in absence of current bypass ($\psi = 0$) and is given by

$$\dot{V}_0 = \left[0.75 \frac{RT}{FP} I_0 \right] N \quad (7)$$

3.3. Determination of i - U curves

The i - U curves were obtained by sweeping the potential (1 mV s⁻¹) and recording the resulting current. An electrical rectifier (Electronic Measurements Inc., Type SCR 100 A/100 V) commanded by a computer was used to scan the potential. An X - Y recorder (Gould 6000) recorded the i - U curves.

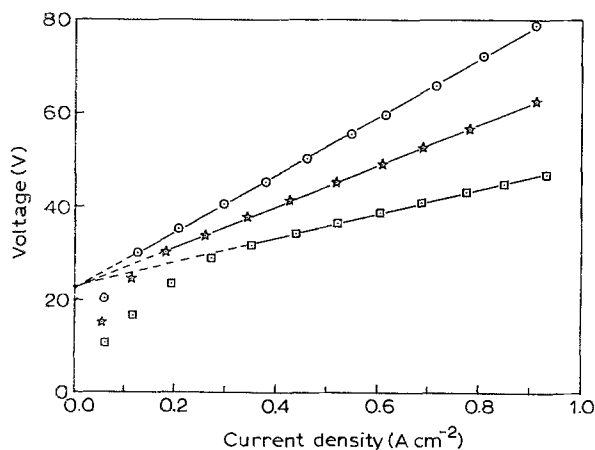


Fig. 5. Current-potential curves for a bipolar stack with 15 cells obtained at different KOH concentrations: (□) 1.000, (☆) 0.5000 and (○) 0.325 M.

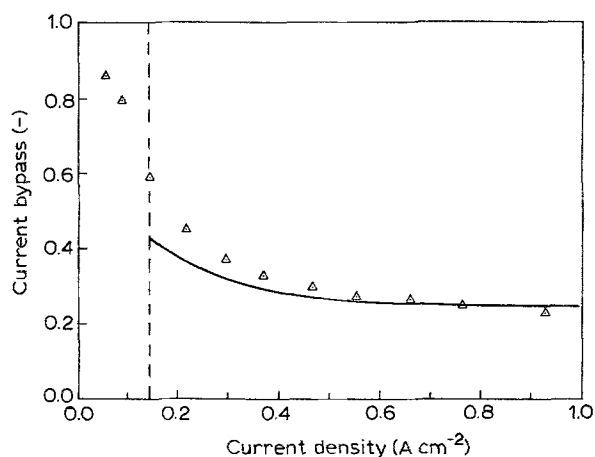


Fig. 6. Current bypass as a function of current density with electrolyte: 0.325 mol dm⁻³ KOH at 50°C. (Δ) experimental, (—) estimated from i - U curves (for the linear i - U region only).

4. Results and discussion

Figure 4 presents the effect of KOH concentration on the current-potential curves of one element cell. For the three KOH concentrations studied the current density can be related to the potential by a linear relationship of the type

$$U_1 = U_0 + R_e i$$

where U_0 is a constant independent of KOH concentration ($U_0 = 1.7$ volt) and R_e decreases with increasing KOH concentration.

The current-potential curves obtained for a "bipolar reactor" with 15 cells are given in Fig. 5 for three KOH concentrations. Transition from non-linear to linear i - U relation is observed; the critical current density at which transition occurs depends on the conductivity of the electrolyte (KOH concentration). The non-linear i - U behaviour observed at low current densities and high KOH concentration is due to the fact that under these conditions the electric field inside the reactor is not sufficiently high to polarize all the electrodes so some of them remain inactive.

As the proposed model supposes that all cells of the bipolar reactor are active; the linear i - U region is only considered.

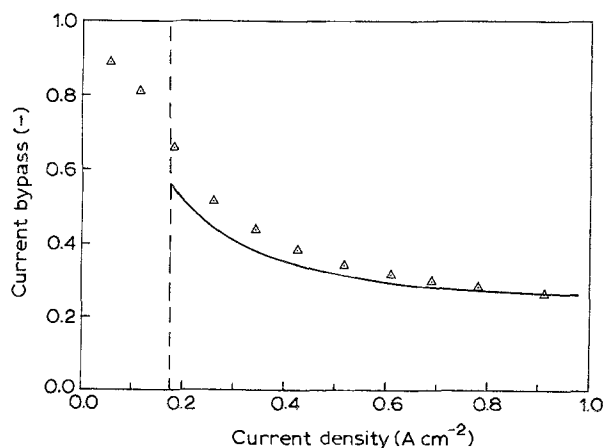


Fig. 7. As for Fig. 6 with electrolyte: 0.50 mol dm⁻³ KOH at 50°C.

The theoretical current bypass calculated from Figs 4 and 5 using Equation 6 and those determined experimentally are given in Figs 6 and 7 for two different KOH concentrations. A good agreement between experimental and estimated bypass current is observed especially at high current density.

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