PATENT SPECIFICATION

(54) ELECTROCHEMICAL REACTOR AND PROCESS

(71) We, DECHEMA, Deutsche Gesellschaft für chemisches Apparatewesen e.V., a body corporate organised according to the laws of the Federal Republic of Germany, of 6000 Frankfurt -am-Main 97, Federal Republic of Germany, do hereby declare the invention for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:-

The present invention relates to an electrochemical reactor suitable for the electrodeposition of metals from dilute solutions when high depletion rates are required.

Previously proposed electrolytic cells with plate or mesh electrodes (see, for example, C.L. Mantell, Electrochemical Engineering, MrGraw-Hill Inc., New York, 1960) give such low space-time yields, when working with solutions having a concentration of the metal to be deposited of less than 1 to 5 g/l, that they are not used industrially for the electrolysis of such dilute solutions.

Considerably better space-time yields may be achieved by means of so-called three-dimensional electrodes, consisting of an open three-dimensional matrix of electrode material into which the electrolyte can penetrate; such electrodes have a very large surface area per unit volume and are therefore especially useful for processes that can only be carried out economically at low current intensities. The matrix may be a fixed entity, as in a porous electrode, or may consist of a large number of discrete particles, as in a packed-bed or fluidised-bed electrode. In the latter type of electrode the bed of particles is fluidised by upward flow of electrolyte. The superiority of three-dimensional electrodes when working with dilute solutions may be illustrated by the fact that the electrolysis of a solution containing 10 p.p.m. of silver in a conventional silver-refining cell was found to give a space-time yield of 0.0017 g/lh, whereas the electrolysis of the same solution in a packed-bed cell gave a space-time yield of 1.8 g/lh.

Further information on three-dimensional electrodes may, for example, be found in the following literature references:

D.N. Bennion and J. Newman, J. Appl. Electrochem. 2 (1972), 113; A.K.P. Chu, M. Fleischmann and G.J. Hills, J. Appl. Electrochem 4 (1974), 323; M. Fleischmann, J.W. Oldfield and L. Tennakoon, J. Appl. Electrochem. 1 (1971), 103; British Patent Specification No. 1 194 181; F.J. Wilkinson and K.P. Haines, Trans. Inst. Mining Met. (Section C) 81 (1972), 157; and D.S. Flett, Chem. and Ind. (1971), 300.

That dimension of a three-dimensional electrode that is parallel to the direction of flow of electric current is called the geometrical bed depth. That dimension is, however, limited by what is known as the effective bed depth, since electrochemical conversion takes place only within this depth. Hitherto, this limitation has been considered to be important since it has been assumed that the effective bed depth is generally only in the order of magnitude of 1 cm (see M. Fleischmann and J.W. Oldfield, J. Electroanal. Chem. 29 (1971), 211).

All previously proposed electrochemical cells with three-dimensional electrodes have a constant geometrical bed depth. With view to reducing outlay costs, i.e. of reducing the quantity of electrode and membrane materials to be used, it is desirable to oppose a given counter-electrode area or diaphragm area by the largest possible volume of three-dimensional electrode. This volume is, however, limited by the fact that the space-time yield is reduced near the effective bed depth.

An object of the present invention is to provide a electrochemical cell or series of cells capable of giving a greater space-time yield than previously proposed electrochemical cells.

The present invention provides an electrochemical reactor containing a three-dimensional electrode or a series of three-dimensional electrodes through which, in operation, electrolyte flows in a direction transverse to the direction of flow of electric current, wherein the geometrical bed depth (as hereinbefore defined) increases in the direction of flow of the electrolyte.

In this specification, the geometrical bed depth is as defined above. In practice, this dimension normally corresponds to the distance from the feeder electrode, which supplies current to the three-dimensional electrode, and the diaphragm, which separates the electrode from its corresponding counter-electrode. The dimension of the three-dimensional electrode parallel to the direction of flow of the electrolyte is referred to as the electrode length, the third dimension of the electrode being referred to as the width.

If the material of the electrode itself does not form a permanent part of the reactor, as is the case with a packed-bed or fluidised-bed electrode, the reactor without the electrode material is included within the scope of the invention. Accordingly, references in what follows to the goemetry of the electrodes are to be construed as applying also to chambers for containing such electrodes.

The increase in geometrical bed depth along the flow path of the electrolyte may be continuous or discontinuous.

The reactor may consist of a single cell, and in this case either or both of the electrodes may be three-dimensional and of increasing geometrical bed depth. A cell having two three-dimensional electrodes, only one of which has increasing geometrical bed depth, may be especially advantageous under certain circumstances.

In the case of a single cell, a continuous increase in the geometrical bed depth from the inlet to the outlet is especially preferred.

The reactor of the invention may, on the other hand, consist of a series of cells. In one possible arrangement, the individual cells are of different sizes and each has a three-dimensional electrode of substantially constant geometrical bed depth, and a stepwise overall increase in geometrical bed depth is achieved by arranging the cells in series in order of increasing geometrical bed depth.

In investigations and calculations relating to the effective bed depth that led to the invention, it was found, notwithstanding the previous assumption mentioned above, that the effective bed depth throughout the ppm concentration range can reach values in the order of magnitude of 10 cm. It was found that optimum performance of a three-dimensional electrode is achieved if the geometrical bed depth along the length of the electrode is increased to correspond with the decrease of concentration within the cell. The reactor of the invention is based upon reducing this principle to a practical construction, whereby it is possible to effect a considerable improvement in the space-time yield over that of previously proposed cells having three-dimensional electrodes, and at the same time to reduce the amount of material used.

As previously mentioned, in the reactor of the invention the geometrical bed depth of the three-dimensional electrode increases continuously or stepwise along the length of the electrode from the inlet to the outlet. This increase in the geometrical bed depth can also be achieved by means of a series of cells of increasing geometrical bed depth through which the electrolyte flows in turn. The width of the electrode can be increased or decreased along its length or it may remain constant. In a preferred arrangement, the width of the electrode is decreased along its length to such an extent that the flow cross-section has the same value at the inlet and outlet, or, more preferably, remains constant from the inlet to the outlet. This enables the mass transfer coefficient to be kept substantially constant over the entire zone of the electrode.

The reactor of the invention offers various advantages over previously proposed arrangements in which the geometrical bed depth is constant. Since, for the same performance of the electrode bed and with a given distance between the diaphragm and the counter-electrode the reactor of the invention has a smaller volume in the space for the counter-electrode, a higher space-time yield is achieved with the construction described. Moreover the diaphragm area required for a given volume of bed is considerably smaller in a cell in accordance with the invention than in previously proposed constructions, so that the amount of material required is reduced. Thirdly, the reduction in concentration along the length of the electrode results in a reduction in the local current density. Since, on the other hand, the cell voltage is constant along the length of the electrode, this results in an increase in the kinetic overvoltages and therefore possibly in a drastic reduction in the current efficiency. If, in a prior art cell of constant geometrical bed depth, the current density decreases by the factor f at a given decrease of concentration, than for the same decrease of concentration, the current density in a cell in accordance with the invention is reduced only by the factor $\sqrt{f}$. This means that considerably better current efficiencies can be obtained with a cell in accordance with the invention.

The three-dimensional electrode or electrodes used in the reactor of the invention may, for example, be of the porous solid type, the packed-bed type or the fluidised-bed type. Packed-bed electrodes are especially preferred. The electrode material may be conductive, for example, metal, graphite or another form of carbon, or may be semi-conductive; a non-conductor coated with a conductive or semi-conductive substance may be used. Where the electrode consists of particulate material, the geometry of the particles is not critical; spherical particles, granulates or chip materials may, for example, be used.

As previously mentioned, a diaphragm is normally used to separate anode and cathode compartments, and, in the case of a particulate electrode, to support and contain the electrode material. The diaphragm may consist, for example, of an ion-exchanger membrane, or, alternatively, a grid or mesh, for example, of plastics material, through which the electrolyte as a whole can pass.

The invention further provides a process for carrying out an electrochemical reaction, wherein an electrolyte is passed through a three-dimensional electrode so associated with a counter-electrode that electric current flows in a direction transverse to the direction of flow of the electrolyte, or through a series of three-dimensional electrodes each thus associated with a counter-electrode, wherein the geometrical bed depth (as hereinbefore defined) increases in the direction of flow of the electrolyte.

The process is especially favourable if the concentration of the electrolyte is low, that is to say, in the p.p.m. range, and is particularly suitable for the electrodeposition of metals from dilute solution. In this case, the electrolyte may comprise an inorganic and/or organic solution containing ions of the metal to be deposited. When working with concentrations of the reacting species that lie in the p.p.m. range, the conductivity of the electrolyte preferably does not fall below 1 to 5 mS/cm, in order to obtain effective bed depths of a few centimetres.

The electrolyte passing through the cathode chamber may be either the same as, or different from, the electrolyte passing through the anode chamber, and either a parallel-flow arrangement or a counter-flow arrangement may be used.

The reactor and process of the invention may, for example, be used for substantially eliminating and/or recovering dissolved metals from waste liquors, or for electrowinning metals from ore liquors having a low metal content. Examples of suitable metals include silver, copper, lead, mercury, gold and platinum. The metals that become concentrated in the three-dimensional electrode after the treatment can be obtained in the form of concentrated solutions by dissolving chemically or electrochemically in suitable electro-lytes. If the electrode itself consists at least partially of the metal to be deposited, further processing may be carried out by metallurgical methods.

The reactor and process of the invention will now be described in greater detail, by way of example only, with reference to the accompanying drawings, in which;

Figure 1 consists of two graphs showing the theoretical relationship between the various electrode dimensions in a cell constructed according to the invention;

Figure 2 is a diagrammatic perspective view of a packed-bed cell constructed according to the invention; and

Figure 3 is a longitudinal section through the cell shown in Figure 2.

Referring now to Figure 1 of the accompanying drawings, the first graph shows the relationship between the electrode length l and the geometrical bed depth h, and the second graph shows the relationship between l and the electrode width b. The theoretical relationships are indicated in each graph by broken lines, the continuous lines showing the dimensions that would in practice be adopted for reasons of construction. These profiles will be discussed in more detail below.

Referring now to Figure 2 and 3 of the accompanying drawings, a cell having a packed-bed cathode and a plate anode is illustrated. Electrolyte flows through an inlet opening 1 into an inlet chamber 7 separated from an outlet chamber 12 by a partition 15. The electrolyte then passes through a holding mesh 13 into an inlet zone 8 which serves to homogenise the flow and is filled with particles, for example, glass spheres having a diameter of 1 mm. From the inlet zone 8 the electrolyte flows through a separating mesh 14 into a chamber 2 containing a packed-bed cathode consisting, for example, of graphite particles having a diameter of 1.25 mm. Electric current is supplied to the packed-bed cathode via a feeder electrode 9 in the form of a graphite plate mounted flush in the cell wall. The electrolyte flows out of the cathode chamber through an upper limiting mesh 10 and through an overflow duct 3 into an anode chamber 16. The anode chamber 16 is separated from the cathode chamber 2 by a cation-exchange membrane secured by means of a frame 11. The membrane 4 also serves to contain the particles of the cathode. The anode chamber 16 contains an anode 5 which is a graphite plate mounted flush in the cell wall. Any gas formed in the anode chamber can escape through a riser tube 17. The electrolyte passes out of the anode chamber 16 into the outlet chamber 12 and leaves the

cell through an outlet opening 6.

As mentioned above, the effective bed depth represents the maximum bed depth within which electrochemical conversion takes place, and is a function both of concentration and of the length of the electrode. The effective bed depth, and the reduction in concentration along the electrode length, can be calculated with the aid of the system of differential equations given below. In the equations, x represents the co-ordinate parallel to the direction of flow of the electric current, the direction from the diaphragm to the feeder electrode being defined as positive, and y represents the co-ordinate parallel to the direction of flow of the electrolyte, the direction from the inlet to the outlet being defined as positive; x and y correspond respectively to h and l mentioned above with reference to, and shown in, Figure 1.

$$\eta(x,y) = \emptyset_p(x,y) - \emptyset_s(x,y) \tag{1}$$

$$\frac{\delta^2\emptyset_s(x,y)}{\delta x^2} = - \frac{A}{x_s v} i\,[\eta(x,y),\,c(x,y)] \tag{2}$$

$$\frac{\delta^2\emptyset_p(x,y)}{\delta x^2} = \frac{A}{x_p (1-v)} i\,[\,(x,y),\,c(x,y)] \tag{3}$$

$$\frac{\delta i_b(x,y)}{\delta x} = A\, i[\eta(x,y),\,c(x,y)] \tag{4}$$

$$\frac{\delta c(x,y)}{\delta y} = - A\frac{i[\eta(x,y),\,c(x,y)]}{uzF} \tag{5}$$

A - specific electrode area (cm^2/cm^3)
c - concentration $(mol\ cm^3)$
i_b - bed current density (A/cm^2)
l - electrode length (cm) (y-coordinate)
h - bed depth (cm) (x-coordinate)
v - voidage
β - current efficiency
η - overvoltage
x_p - effective particle conductivity (S cm)
x_s - electrolyte conductivity (S cm)
$\emptyset_p$ - particle potential (V)
$\emptyset_s$ - electrolyte potential (V)

By using known difference procedures and iterative methods the differential equation system (1) to (5) can be solved numerically with the aid of a computer for each microkinetic rate equation i $(\eta,\,c)$.

Calculation example

The following complete equations for calculating the cell dimensions were obtained as a solution of the differential equation system (1) to (5) for the special case, often occurring in the electrodeposition of metals, of a diffusion - controlled first-order reaction, in a packed bed cell having a constant flow cross-section and with the geometrical bed depth so chosen that the full limiting current density obtains at all points of the electrode:

$$l_l = \frac{u}{k.A}\log\frac{c_o}{c_l}. \tag{6}$$

$$h(l) = \left\{\frac{V.x_s.d_p^{(\eta-0.12)}}{3(1-v)k.z.F.c_o}\right\}^{1/2}\ \exp\left\{\frac{k.A.l}{2u}\right\} \tag{7}$$

$$b(l) = \frac{V_d}{u.\bar{h}(l)}. \tag{8}$$

A	-	specific electrode area (cm^2/cm^3)
$b(l)$	-	width of electrode at l (cm)
c_o	-	initial concentration (mol/cm^3)
c_1	-	final concentration (mol/cm^3)
d_p	-	particle diameter (cm)
F	-	Faraday number (As/val)
$h(l)$	-	bed depth at l (cm)
k	-	mass transfer coefficient (cm/s)
l	-	length co-ordinate of the electrode (cm)
l_t	-	electrode length (cm)
u	-	flow velocity (cm/s)
v	-	voidage
v_d	-	flow rate (cm^3/s)
z	-	charge number of reaction (val/mol)
β	-	current efficiency
η	-	overvoltage at the bed electrode (V)
x_s	-	electrolyte conductivity (S/cm)

A waste water (x_s — 0.019 S/cm), containing 100 p.p.m. of silver, was to be depleted to 1 p.p.m. electrolytically in a packed-bed cell using a throughput of 50 l/h.
Other predetermined values were as follows:

A	-	21.12 cm^2/cm^3
d_p	-	0.125 cm
v	-	0.56
u	-	0.5 cm/s
k	-	1.624×10^{-3} cm/s
β	-	0.60
η	-	0.4 V.

Using the equations (6) to (8), the following dimensions for the electrode bed were obtained in the case of the packed-bed cell illustrated diagrammatically in Figure 2:

Electrode length	-	67.15 cm
Geometrical bed depth		
at the electrode inlet	-	1.08 cm
at the electrode outlet	-	10.80 cm
Electrode width		
at the electrode inlet	-	25.72 cm
at the electrode outlet	-	2.57 cm

The theoretical dependence of the geometrical bed depth h and the width of the electrode b upon the electrode length l, and the to-scale dimensions of the parameters h, b and l are shown in broken lines in Figure 1.
If a value of 1 cm is selected for the distance between the ion-exchange membrane and the anode, then the described cell has a space-time yield of 1.78 g/lh. A cell having the same conversion capacity and a constant geometrical bed depth of 1 cm would on the other hand have a space-time yield of only 1.3 g/lh and would require approximately twice as much anode and membrane material.
In a cell having a constant bed depth of 1 cm, the local current density would decrease at the current outlet to 1/100 of its value at the current inlet, whereas in the described cell it would be reduced only to 1/10 of its initial value. For this reason, the cell to which the calculation in this Example relates and which has an increasing geometrical bed depth can provide a considerably better current efficiency. If, in the cell in accordance with the invention a current yield of 60% is obtained, this figure decreases to approximately 13% in a cell having a constant geometrical bed depth.

Constructional example
For the purpose of constructing an experimental cell, and in order to simplify construction, the theoretical profiles, shown in broken lines in Figure 1, were linearised (solid lines in Figure 1).
Linearisation of the profiles illustrated in Figure 1 resulted in over-dimensioning of the cell, illustrated in Figure 2, for dealing with the problem of reducing a silver concentration from 100 to 1 p.p.m. as described in the Calculation Example. If the actual geometry of the cell, illustrated in Figure 2, is used as a basis while using the profiles shown in solid lines in

Figure 1, and if it is borne in mind that the flow cross-section in fact has the same value at the electrode inlet and the electrode outlet but passes through a maximum between these, then the final concentration that can be theoretically expected can be calculated afresh. By means of the details given in the Calculation Example and of the equations

$$k \quad = \quad 3.827 \times 10^{-3} \sqrt{u} \qquad (9)$$

$$h(l) \quad = \quad 1.08 + 0.1448\ l \qquad (10)$$

$$b(l) \quad = \quad 25.72 - 0.3448\ l \qquad (11)$$

which result from the geometry of the electrode bed of the cell illustrated in Figure 2, a theoretical final concentration of 0.2 p.p.m. is obtained by corresponding use of the differential equations (1) to (5). For this depletion a cell current of 2.07 A is necessary when a current efficiency of 60% is assumed.

In the electrolysis of a 0.2 M $NaNO_3$-solution ($x_s = 0.019$ S/cm), containing 100 p.p.m. of silver ions and with a throughput of 50 l/h in the cell illustrated in Figures 2 and 3, the following experimental results were obtained:

Cell current	—	2.17 A
Cell voltage	—	2.5 V
Final concentration	—	0.13 p.p.m.
Current efficiency	—	0.57

These results are very similar to the required performance of the cell which was designed for the electrodeposition of silver.

A further experiment relating to the electrolysis of a 0.15 M Na_2SO_4 solution ($x_s = 0.021$ S/cm) containing 30 p.p.m. of copper ions, using a throughput of 50 l/h, in an identical cell, gave the following results:

Cell current	—	2.16 A
Cell voltage	—	2.8 V
Final concentration	—	0.01 p.p.m.
Current efficiency	—	0.59

WHAT WE CLAIM IS:

1. An electrochemical reactor containing a three-dimensional electrode or a series of three-dimensional electrodes through which, in operation, electrolyte flows in a direction transverse to the direction of flow of electric current, wherein the geometrical bed depth (as hereinbefore defined) increases in the direction of flow of the electrolyte.

2. An electrochemical reactor as claimed in claim 1, wherein the geometrical bed depth increases continuously.

3. An electrochemical reactor as claimed in claim 1 or claim 2, which is a single cell having a three-dimensional cathode and/or a three-dimensional anode.

4. An electrochemical reactor as claimed in claim 1, wherein the geometrical bed depth increases discontinuously.

5. An electrochemical reactor as claimed in claim 4, which includes a series of three-dimensional electrodes each having a substantially constant geometrical bed depth, the geometrical bed depth increasing from one electrode to the next.

6. An electrochemical reactor as claimed in any one of claims 1 to 5, wherein the electrode width (as hereinbefore defined) decreases in the direction of flow of the electrolyte.

7. An electrochemical reactor as claimed in claim 6, wherein the electrolyte flows, in operation, from an inlet to an outlet and the decrease in electrode width is such that, in operation, the flow cross-section is substantially identical at the inlet and at the outlet.

8. An electrochemical reactor as claimed in claim 7, wherein the decrease in electrode

width is such that, in operation, the flow cross-section is substantially constant along the flow path of the electrolyte through the or each electrode.

9. An electrochemical reactor as claimed in any one of claims 1 to 5, wherein the electrode width is substantially constant along the flow path of the electrolyte.

10. An electrochemical reactor as claimed in any one of claims 1 to 5, wherein the electrode width increases in the direction of flow of the electrolyte.

11. An electrochemical reactor as claimed in any one of claims 1 to 10, wherein the or at least one three-dimensional electrode is a packed-bed electrode.

12. An electrochemical reactor as claimed in any one of claims 1 to 10, wherein the or at least one three-dimensional electrode is a fluidised-bed electrode.

13. An electrochemical reactor as claimed in any one of claims 1 to 10, wherein the or at least one three-dimensional electrode is a porous solid body.

14. An electrochemical reactor as claimed in any one of claims 11 to 13, wherein the or at least one three-dimensional electrode comprises metal, graphite or another form of carbon.

15. An electrochemical reactor as claimed in any one of claims 11 to 13, wherein the or at least one three-dimensional electrode comprises a semi-conductor.

16. An electrochemical reactor as claimed in any one of claims 11 to 13, wherein the or at least one three-dimensional electrode comprises a non-conductor coated with a conductive or semi-conductive substance.

17. An electrochemical reactor as claimed in claim 11 or claim 12, wherein the or at least one three-dimensional electrode is supported and separated from its counter-electrode by a diaphragm permeable or impermeable to electrolyte.

18. An electrochemical reactor substantially as hereinbefore described with reference to, and as shown in, any one of Figures 1 to 3 of the accompanying drawings.

19. An electrochemical reactor as claimed in any one of claims 1 to 18 wherein the or each electrode contains an electrolyte, the geometrical bed-depth and electrode length being in accordance with the system of differential equations (1) to (5) hereinbefore given.

20. An electrochemical reactor having a chamber for containing a three-dimensional electrode or a series of chambers for containing three-dimensional electrodes, through which chamber or chambers, in operation, electrolyte flows in a direction transverse to the direction of flow of electric current, wherein the geometrical bed-depth (as hereinbefore defined) increases in the direction of flow of the electrolyte.

21. An electrochemical reactor as claimed in claim 20, wherein the geometrical bed-depth increases continuously.

22. An electrochemical reactor as claimed in claim 20 or claim 21, which is a single cell having a chamber for containing a three-dimensional cathode and/or a chamber for containing a three-dimensional anode.

23. An electrochemical reactor as claimed in claim 20, wherein the geometrical bed-depth increases discontinuously.

24. An electrochemical reactor as claimed in claim 23, which includes a series of chambers for containing three-dimensional electrodes, each chamber having a substantially constant geometrical bed depth, the geometrical bed depth increasing from one electrode chamber to the next.

25. An electrochemical reactor as claimed in any one of claims 20 to 24, wherein the electrode chamber width (as hereinbefore defined) decreases in the direction of flow of the electrolyte.

26. An electrochemical reactor as claimed in claim 25, wherein the electrolyte flows, in operation, from an inlet to an outlet and the decrease in electrode chamber width is such that, in operation, the flow cross-section is substantially identical at the inlet and at the outlet.

27. An electrochemical reactor as claimed in claim 26, wherein the decrease in electrode chamber width is such that, in operation, the flow cross-section is substantially constant along the flow path of the electrolyte through the or each electrode chamber.

28. An electrochemical reactor as claimed in any one of claims 20 to 24, wherein the electrode chamber width is substantially constant along the flow path of the electrolyte.

29. An electrochemical reactor as claimed in any one of claims 20 to 24, wherein the electrode chamber width increases in the direction of flow of the electrolyte.

30. An electrochemical reactor as claimed in any one of claims 20 to 28, wherein the or at least one electrode chamber is separated from the corresponding counter-electrode or counter-electrode chamber by a diaphragm permeable or impermeable to electrolyte.

31. A process for carrying out an electrochemical reaction, wherein an electrolyte is passed through a three-dimensional electrode so associated with a counter-electrode that electric current flows in a direction transverse to the direction of flow of the electrolyte, or through a series of three-dimensional electrodes each thus associated with a counter-

electrode, wherein the geometrical bed depth (as hereinbefore defined) increases in the direction of flow of the electrolyte.

32. A process as claimed in claim 31, wherein the electrolyte is passed through an electrochemical reactor as claimed in any one of claims 1 to 18.

33. A process as claimed in claim 31 or claim 32, wherein the electrolyte comprises an inorganic or organic solution containing ions of a metal.

34. A process as claimed in claim 33, wherein the metal is selected from silver, copper, lead, mercury, gold and platinum.

35. A process as claimed in claim 33 or claim 34, wherein, the or at least one three-dimensional electrode consists at least partially of the metal the ions of which are present in the electrolyte.

36. A process as claimed in any one of claims 31 to 35, wherein the conductivity of the electrolyte is at least 1 to 5 mS/cm.

37. A process as claimed in any one of claims 31 to 36, for carrying out a diffusion-controlled first order reaction in a packed-bed cell having a constant flow cross-section, wherein the dimensions of the or at least one three-dimensional electrode are in accordance with equations (6) to (8) hereinbefore given.
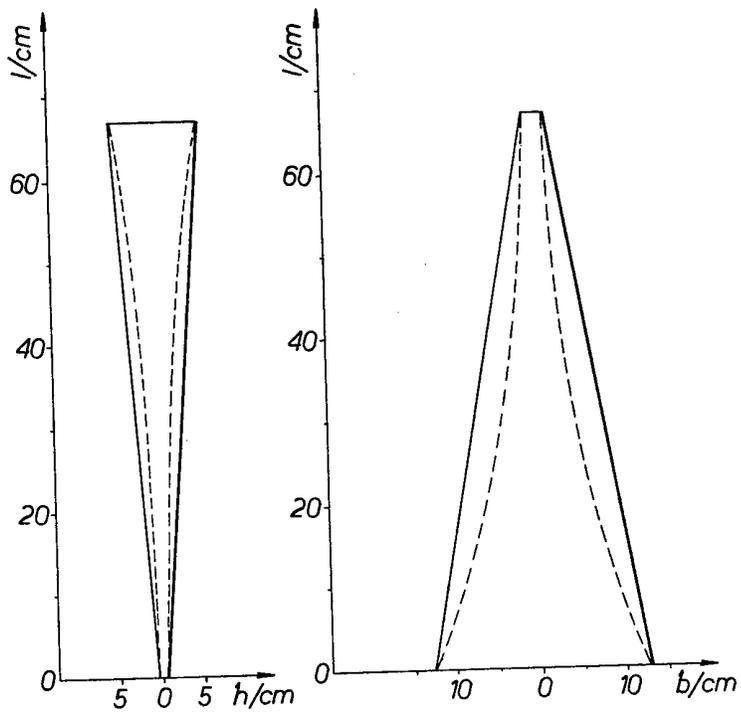
38. An electrochemical reaction carried out substantially as hereinbefore described in either of the Examples.

39. A product of an electrochemical reaction, whenever obtained by a process as claimed in any one of claims 31 to 38.

40. A product as claimed in claim 39, which is a metal.

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FIG./.

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2 SHEETS   This drawing is a reproduction of
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Sheet 2

FIG.2.

FIG.3.