



- (51) **International Patent Classification:**
H01M 4/66 (2006.01) *H01M 10/0585* (2010.01)
H01M 10/04 (2006.01)
- (21) **International Application Number:**
PCT/EP2015/052860
- (22) **International Filing Date:**
11 February 2015 (11.02.2015)
- (25) **Filing Language:** English
- (26) **Publication Language:** English
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furt am Main (DE).
- (81) **Designated States** (*unless otherwise indicated, for every
kind of national protection available*): AE, AG, AL, AM,
AO, AT, AU, AZ, BA, BB, BG, BH, BN, BR, BW, BY,

BZ, CA, CH, CL, CN, CO, CR, CU, CZ, DE, DK, DM,
DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT,
HN, HR, HU, ID, IL, IN, IR, IS, JP, KE, KG, KN, KP, KR,
KZ, LA, LC, LK, LR, LS, LU, LY, MA, MD, ME, MG,
MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM,
PA, PE, PG, PH, PL, PT, QA, RO, RS, RU, RW, SA, SC,
SD, SE, SG, SK, SL, SM, ST, SV, SY, TH, TJ, TM, TN,
TR, TT, TZ, UA, UG, US, UZ, VC, VN, ZA, ZM, ZW.

- (84) **Designated States** (*unless otherwise indicated, for every
kind of regional protection available*): ARIPO (BW, GH,
GM, KE, LR, LS, MW, MZ, NA, RW, SD, SL, ST, SZ,
TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, RU,
TJ, TM), European (AL, AT, BE, BG, CH, CY, CZ, DE,
DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU,
LV, MC, MK, MT, NL, NO, PL, PT, RO, RS, SE, SI, SK,
SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ,
GW, KM, ML, MR, NE, SN, TD, TG).

Declarations under Rule 4.17:

— *of inventorship (Rule 4.17(iv))*

Published:

— *with international search report (Art. 21(3))*

(54) **Title:** BIPOLAR ELECTROCHEMICAL SYSTEM

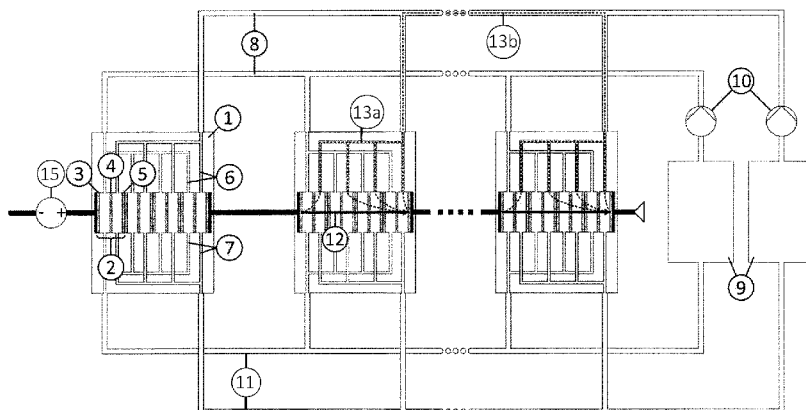


FIG. 1 (PRIOR ART)

(57) **Abstract:** A bipolar electrochemical system comprises at least one bipolar stack (1) consisting of a plurality of cells (2) each having an anode (3), a separator (4), a cathode (5), an electrolyte inlet (6) and an electrolyte outlet (7); at least one electrolyte supply circuit (8) and a shunt current interrupter (16). The shunt current interrupter (16) comprises a perforated plate (17) having boreholes (20) penetrating the perforated plate (17), wherein electrolyte is guided to an upper surface (24) of the perforated plate (17) to flow through the boreholes (20). On a lower surface (25) of the perforated plate (17) facing a collecting section (19) grooves (21) are provided between adjacent boreholes (21).



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BIPOLAR ELECTROCHEMICAL SYSTEM

5 The present invention is directed to a bipolar electrochemical system according to the preamble of claim 1 comprising at least one bipolar stack consisting of a plurality of cells each having an anode, a separator, a cathode, an electrically conductive liquid as the electrolyte, an electrolyte inlet and an electrolyte outlet; at least one electrolyte supply circuit and at least one shunt current interrupter.

10

Background of the Invention

15 Bipolar electrochemical systems like redox flow batteries or alkaline electrolyzers usually comprise at least one bipolar stack with multiple single cells. As schematically shown in Fig. 1, a bipolar stack 1 consists of multiple single cells 2 with anode 3, separator 4, cathode 5, electrolyte inlet 6 and outlet 7 for each cell 2. A liquid electrolyte is supplied to the at least one bipolar stack through at least one electrolyte supply circuit 8, including pipe connections, storage tanks 9 and active or passive fluid conveying systems 10 such as natural circulation or a pump. The electrolyte is withdrawn from the cells through an electrolyte withdrawal circuit (lines) 11 and recirculated to the storage tanks 9. In Fig. 1 a Vanadium Redox Flow Battery using two different liquid electrolytes is shown as an example, but the invention applies to all bipolar electrochemical systems which

20

25 use at least one liquid electrolyte

State of the art concerning the electrical connection of the bipolar stacks is a serial arrangement. The coupling of the regarded system to an electric energy system or to the grid can be realized by a transformer and a rectifier, a converter

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or an inverter and a transformer, depending on the direction of the flow of the electric energy.

The differences of potential lead to a current flow. Thereby the main current flow direction 12 within a bipolar electrochemical system is orthogonally to the single cells through the stack. The voltage of a bipolar stack is the sum of all cell voltages. The electrolyte feed to every single cell is realized by a stack internal distribution system whereby an electrical connection of each cell of the stack across the electrolyte is given. The same is true for the electrolyte withdrawal from the cells. If at least two stacks are used, there exist electrical connections between the stacks via the electrolyte supply circuit and via the electrolyte withdrawal circuit as well. Result is the formation of shunt currents 13a between the single cells of each stack across the electrolyte and shunt currents 13b between the stacks of the bipolar electrical system. Shunt currents in electrochemical systems reduce the efficiency factor significantly and thus its economic feasibility. Preconditions for the formation of shunt currents are differences of the electric potential and electrically conductive fluid phases (electrolyte solutions). Both preconditions are given in a typical bipolar electrochemical system using liquid electrolytes.

The different stack voltage levels cause shunt currents 13b across the electrolyte lines between at least two stacks via the electrolyte supply circuit. Fig. 1 shows the state of the art configuration of a Vanadium Redox Flow Battery, which uses two different liquid electrolytes, without any measures to reduce shunt currents as well as a schematic description of arising shunt currents within the system. To facilitate the understanding, only some exemplary shunt currents are shown in the figure. Other bipolar electrochemical systems need one liquid electrolyte only, but the principle of shunt current remains the same.

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The magnitude of the shunt currents depends on the differences of the electric potential as well as on conductivity properties of the electrolyte and on the geometrical dimensions of the electrolyte lines. Characteristic is the ohmic resistance that is dependent on cross section and length of the electrolyte channels.

The minimization or avoidance of shunt currents is essential for the efficiency increase of the system. Higher ohmic resistances within the electrolyte channels reduce the shunt currents and therefore increase the electrochemical efficiency. On the other hand, smaller cross-sections and longer lengths of the electrolyte lines also increase the pressure drop in the electrolyte system and hence the necessary pump power, which in turn decreases the overall energy efficiency of the bipolar system.

Two main approaches are known to minimize or to avoid shunt currents: Manipulation of the electrolyte flow through the electrochemical system to increase the ohmic resistances and reducing the differences of potential by choice of an alternative electrical connection. The reduction of shunt currents within a stack 13a and between several stacks 13b must be considered separately.

An effective possibility to reduce the shunt currents within a stack 13a is both reducing the cross section and increasing the length of the electrolyte lines at the same time. Fig. 2 shows the state of the art arrangement of such a system, including measures to reduce shunt currents within a stack but without any measures to reduce shunt currents between several stacks. As shown in Fig. 2 each electrolyte channel has a meander structure 14.

Concerning the reduction of shunt currents between two or more stacks 13b, two approaches have been pursued. A simple but expensive way to reduce shunt currents between two stacks is the choice of a parallel electrical connec-

– 4 –

tion of each stack to avoid the high potential differences generated by an electric series connection of the stacks. Figure 3 shows the principle of this alternative electrical connection arrangement, which requires the usage of a transformer and rectifier, a converter or an inverter and transformer 15 for each stack to couple the bipolar electrochemical system to the electrical energy system or to the grid. It sets all stacks of the system to the same defined electrical potential level. This arrangement decreases shunt currents significantly due to a lack of potential difference.

10 A second approach manipulates the electrolyte flow. A simple way to increase the electric resistance is to extend the length of the ducts or to reduce the duct diameter. These approaches are inefficient and lead to high material and pumping costs.

15 The solutions described for example in DE 31 40 347 A1 or DE 699 16 869 T2 show adapted and shunt current reducing ducts integrated in the stack design. A local interruption that means creating a discontinuous flow of the electrolyte to extinct shunt currents is also shown in the documents US 679,050, CH 206 960, US 2,673,232 or JP 62160664 A. All of these solutions are based on the same principle shown in Fig. 4, where 16 depicts the fluid based shunt current inter-
20 rupters.

Both fundamental principles need additional equipment, e.g. shunt current inter-
rupters within the electrolyte channels or several transformers/ converters/
25 inverters to couple the bipolar electrochemical system to the electric energy system or to the grid. Additional equipment implies additional costs (investment as well as operational costs). It also is an additional source of error during operation.

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By generating a mathematical simulation model of such a bipolar electrochemical system, the advantages of using fluid interrupters (Fig. 4) or an electrical parallel arrangement of the stacks (Fig. 3) have been shown. As results, the calculated cumulative shunt current losses for the different arrangements as
5 function of the stack number are shown in Fig. 5.

If no shunt current reducing measures are taken, an exponential increase of shunt current losses as a function of stack number can be demonstrated with the simulation model. If a measure to reduce shunt currents is applied, the
10 losses are decreased. If the shunt currents 13b between the stacks can be decreased to zero by interruption of the shunt currents, the losses become independent of the number of stacks.

Both methods have disadvantages that have to be considered: To reduce shunt
15 currents sustainably by using electrolyte flow interrupters within the bipolar electrochemical system as shown in Fig. 4, every stack needs 4 interruption units. In other bipolar electrochemical systems which need one electrolyte only, every stack needs 2 shunt current interruption units. The known interruption units require a high amount of maintenance. An electrical parallel arrangement
20 of stacks needs expensive additional installation costs of transformers/ converters for every stack.

Summary of the Invention

25 It is the object of the present invention to reliably realize a minimization or even avoidance of shunt currents in bipolar electrochemical systems based on an efficient and cost effective concept.

The problem is solved by a bipolar electrochemical system comprising the fea-
30 tures of claim 1, wherein according to the invention the shunt current interrupter

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comprises a perforated plate having boreholes penetrating the perforated plate, wherein electrolyte is guided to an upper surface of the perforated plate to flow through the boreholes, and wherein on a lower surface of the perforated plate, facing a collecting section, grooves are provided between adjacent boreholes.

5 There is a drop height h_d between the lower surface of the perforated plate and the electrolyte level within the collecting section.

Thereby, droplets are formed directly on the lower surface of the perforated plate wherein the grooves prevent the formation of a fluid film on the lower plate surface and a liquid flow down the wall of the cell. The individual droplets fall from the lower surface of the perforated plate to the electrolyte level in the collecting section of the apparatus. The invention shows a very simple shunt current interrupter apparatus that is able to reliably create discontinuous electrolyte flows. The shunt current across the apparatus is totally avoided, because the space between the droplets is filled with a non-conductive gas which does not allow the shunt current to flow.

According to a preferred embodiment of the invention, the cross section of said grooves is rectangular, triangular, trapezoidal or circular. This design has proven to be particularly effective in forming droplets and preventing the formation of a fluid film.

Preferably, the angle between the inner edge of the groove and the lower surface of the perforated plate is between 30° and 120°, more preferably between 45° and 90° and most preferably between 45° and 75°.

Particularly good results are achieved if a width of a wall between the borehole and the adjacent groove fulfills the following equation

$$d_h + 2 \cdot w_w < d_d$$

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where

d_h = diameter of the borehole, see Fig. 7 and Fig. 9

w_w = wall width, see Fig. 7 and Fig. 9

5 d_d = maximum droplet diameter.

In a specific embodiment, the width w_w of the wall between the borehole and the adjacent groove is between 0.0 and 3.0 mm, preferably between 0.1 and 1.0 mm.

10

In order to avoid the formation of a fluid film flowing down the wall of the cell the invention preferably provides a circumventing groove between the outermost boreholes and the outer circumference of the perforated plate.

15 The perforated plate may have a circular or rectangular shape when viewed from above.

If below the perforated plate a collecting section is formed for collecting the electrolyte liquid flowing through the perforated plate, then forming droplets and then dropping onto the fluid level F, it is preferred within the invention that the drop height h_d between the lower surface of the perforated plate and the fluid level F within the collecting section is between 5 and 400 mm, more preferably between 12 and 200 mm.

20
25 The invention also is directed to a method for operating a bipolar electrochemical system as described above, wherein the following relation is fulfilled

$$0.001 \leq We \leq 2.0,$$

30 where

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$$We = \frac{\rho_g \cdot u_f^2 \cdot d_h}{\sigma_f}$$

We = Weber number,

ρ_g = gas phase (around the droplets) density (kg/m³)

u_f = fluid velocity in the borehole (m/s)

σ_f = fluidal surface tension (N/m)

5 d_h = borehole diameter (m)

The Weber number (We) is a dimensionless number in fluid mechanics that describes the ratio between inertia and surface forces.

10 Fig. 4 shows the arrangement of the shunt current interrupters 16 in the electrolyte supply lines 8 and the electrolyte withdrawal lines 11 to and from each stack of a Vanadium Redox Flow Battery as an example of a bipolar electrochemical system, whereby the shunt currents 13b between stacks are avoided.

15 It is within this invention to also apply the shunt current interrupters within a stack. In this case the shunt current interrupters are applied in the electrolyte inlets 6 and electrolyte outlets 7 to of each cell and replace the meander structures 14. This avoids the shunt currents 13a within the stack.

20 By arranging the shunt current interrupters both in the electrolyte supply and withdrawal circuits to the stacks, 8 and 11, and in the electrolyte inlets and outlets of the cells, 6 and 7, both kinds of shunt currents, 13a and 13b, are avoided.

25 The invention will now be explained in detail with reference to preferred embodiments and the drawings. All features described and/or illustrated form the subject-matter of the invention per se or in any combination, independent of their inclusion in the claims or their back-reference.

Brief Description of the Drawings

- 5 Fig. 1 schematically depicts a bipolar electrochemical system wherein shunt currents are formed,
- Fig. 2 shows a state of the art arrangement of the bipolar electrochemical system,
- 10 Fig. 3 schematically shows the reduction of shunt currents by choice of an alternative electrical connection,
- Fig. 4 schematically shows shunt current reduction by fluid interruption,
- 15 Fig. 5 shows cumulative shunt currents as function of stack number,
- Fig. 6 schematically depicts a shunt current interrupter according to the present invention,
- 20 Fig. 7 shows alternative embodiments of the cross section of the grooves in the shunt current interrupter according to the present invention,
- Fig. 8 shows a shunt current interrupter plate from below, and
- 25 Fig. 9 shows a cross section of the shunt current interrupter plate along line A-A in Fig. 8.

Detailed description of the preferred embodiments

The bipolar electrochemical system according to the present invention includes a shunt current interrupter 16 as schematically shown in Fig. 6. The shunt current interrupter 16 consists of a perforated plate 17, a drum section 18 and a collecting section 19. The system interrupts the continuous electrolyte flow by creating droplets with the maximum droplet diameter d_d under the perforated plate 17. This causes a significant reduction of the shunt currents. If the drop height h_d between the lower surface of the perforated plate and the liquid level F in the collecting section is sufficiently high, the shunt current across the shunt current interrupter is decreased to zero and thereby completely avoided.

Centerpiece of the invented shunt current interrupter 16 is the perforated plate 17. It can have either an angled or a circular base. As shown in Fig. 6 to 9, the plate 17 contains penetrating boreholes 20 with the diameter d_h and grooves or slots 21 with the groove or slot width w_s . As shown in Fig. 8, an additional circumventing groove 23 is provided between the outermost boreholes 20 and the outer circumference of the perforated plate 17. The grooves 21, 23 are provided on the lower surface 25 of the perforated plate 17. The cross section of the grooves 21, 23 can be rectangular as well as triangular, trapezoidal or circular as shown in Fig. 7. It is within the invention to provide other suitable shapes, such as polygonal forms. The angle α_s between the inner edge 21i of groove 21, 23 and the lower surface 25 of the perforated plate 17 is between 30° and 120°, preferably between 45° and 90° and most preferably between 45° and 75°.

A further characteristic of the perforated shunt current interrupter plate 17 is the web width w_w between the boreholes 20 and the grooves 21 corresponding to the width of the walls 22 between the boreholes 20 and the grooves 21. The web works as tearing edge for the droplets and avoids the formation of a current

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conductive fluid coat due to agglomerating droplets as well as the formation of a fluid jet. If the equation

$$d_h + 2 \cdot w_w < d_d \quad (0.1)$$

5

is fulfilled, the web width between the borehole 20 and the groove 21 is primarily decisive for the droplet size and not the physical properties of the fluid. Furthermore, the drop height of the droplets h_d is decisive for the shunt current interrupting properties. As shown as an example in Fig 8, the perforated shunt current interrupter plate 17 has a circular base. Other shapes, such as a rectangular or polygonal base are also possible within the invention.

10

In operation and similar to the arrangement shown in Fig. 4, electrolyte will be introduced via the electrolyte supply circuit 8 into the shunt current interrupter 16 and is collected above the upper surface 24 of the perforated plate 17 forming an upper fluid level F_u above said plate 17 (see Fig. 6). The electrolyte then flows through the boreholes 20 wherein on the lower surface 25 of the plate 17 droplets 26 are generated and drip into the collecting section 19 without the formation of a fluid film on the lower surface 25 of the plate 17. Thereby electrical shunt currents are reliably avoided.

15

20

The dimensioning of such a droplet based shunt current interrupter is strongly dependent on the physical properties of the electrolyte. The Weber number

$$We = \frac{\rho_g \cdot u_f^2 \cdot d_h}{\sigma_f} \quad (0.2)$$

25

as a function of gas phase (around the droplets!) density ρ_g , fluid velocity in the borehole u_f , fluidal surface tension σ_f and borehole diameter d_h , describes the ratio of inertial and surface forces. The Weber number for the operation range of

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the shunt current interrupter according to the invention preferably is between 0.001 and 2.0, more preferably between 0.002 and 1.0.

In a preferred embodiment of the invention, the minimal web width w_w between the boreholes 20 and the grooves 21 is between 0.0 mm and 3.0 mm, preferably between 0.0 mm and 1.0 mm. The drop height of the droplets h_d between the lower surface 25 of the interrupter plate 17 and the fluid level F_l within the collecting section 19 is between 5 mm and 400 mm, preferably between 12 mm and 200 mm.

According to the invention the local interruption of the electrolyte flow preferably is controlled and adjusted to the working flow rate. Additionally, the invention leads to minor efficiency losses so that the electrochemical system can run within the designed and defined working range. This ensures the efficient minimization of the shunt currents at the design point as well as in defined part-load or overload operation points.

The present invention ensures a controlled and defined fluid flow interruption. It also enables tailor made droplet generation adapted to the regarded electrochemical system and working parameters. This ensures a firm and wideband shunt current interruption in a given and defined operating range. This operation range is dependent on the borehole diameter d_h , the web width w_w between the boreholes 20 and the grooves 21 and the angle α_s between the groove edge 21i and the plate 17. It can be predefined and adapted to the requirements of the system. The defined web width w_w between the boreholes 20 and the grooves 21 provides an immediate droplet break-off and consequently ensures a minimum size of the drop height of the droplets h_d and consequently of the shunt current interruption apparatus.

List of reference numbers

	1	bipolar stack
	2	cell
5	3	anode
	4	separator
	5	cathode
	6	electrolyte inlet
	7	electrolyte outlet
10	8	electrolyte supply circuit
	9	storage tank
	10	fluid conveying system
	11	electrolyte withdrawal circuit
	12	main current flow direction
15	13	shunt current
	13a	shunt current between cells
	13b	shunt current between stacks
	14	meander structure
	15	converter
20	16	shunt current interrupter
	17	perforated plate
	18	drum section
	19	collecting section
	20	borehole
25	21	groove
	21i	inner edge of groove 21
	22	wall
	23	circumventing groove
	24	upper surface of plate 17
30	25	lower surface of plate 17

– 14 –

26 droplet

 F_u, F_l upper/ lower fluid level

5

 α_s angle between the groove edge and the plate d_h borehole diameter h_d drop height of the droplets w_s groove or slot width

10

 w_w web width ρ_g gas phase density (around the droplets) u_f fluid velocity in the borehole σ_f fluidal surface tension

15

Claims:

1. Bipolar electrochemical system comprising

5 at least one bipolar stack (1) consisting of a plurality of cells (2) each having an anode (3), a separator (4), a cathode (5), an electrolyte inlet (6) and an electrolyte outlet (7);

at least one electrolyte supply circuit (8) and a shunt current interrupter (16),

10

characterized in that the shunt current interrupter (16) comprises a perforated plate (17) having boreholes (20) penetrating the perforated plate (17), wherein electrolyte is guided to an upper surface (24) of the perforated plate (17) to flow through the boreholes (20), and

15

that on a lower surface (25) of the perforated plate (17) facing a collecting section (19) grooves (21) are provided between adjacent boreholes (21).

20

2. Bipolar electrochemical system according to claim 1, **characterized in** that the cross section of said grooves (21) is rectangular, triangular, trapezoidal or circular.

25

3. Bipolar electrochemical system according to claim 1 or 2, **characterized in** that an angle (α_s) between an inner edge (21i) of the groove (21) and the lower surface (25) of the perforated plate (17) is between 30° and 120°, preferably between 45° and 90° and most preferably between 45° and 75°.

30

4. Bipolar electrochemical system according to any of the preceding claims, **characterized in** that a width (w_w) of a wall (22) between the borehole (20) and the adjacent groove (21) fulfills the following equation

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$$d_h + 2 \cdot w_w < d_d$$

wherein

- 5 d_h = diameter of the borehole (20)
 w_w = width of wall (22)
 d_d = maximum droplet diameter.

- 10 5. Bipolar electrochemical system according to any of the preceding claims,
 characterized in that the width (w_w) of a wall (22) between the borehole (20)
 and the adjacent groove (21) is between 0.0 mm and 3.0 mm, preferably be-
 tween 0.1 and 1.0 mm.
- 15 6. Bipolar electrochemical system according to any of the preceding claims,
 characterized in that between the outermost boreholes (20) and the outer
 circumference of the perforated plate (17) a circumventing groove (23) is
 provided.
- 20 7. Bipolar electrochemical system according to any of the preceding claims,
 characterized in that the perforated plate (17) has a circular or rectangular
 shape when viewed from above.
- 25 8. Bipolar electrochemical system according to any of the preceding claims,
 characterized in that below the perforated plate (17) a collecting section
 (19) is formed for collecting the electrolyte liquid flowing through the perfo-
 rated plate (17) and then dripping from the lower surface (25) of the perforat-
 ed plate (17) and that the drop height (h_d) between the lower surface (25) of
 the perforated plate (17) and the fluid level (F_l) within the collecting section
 (19) is between 5 and 400 mm, preferably between 12 and 200 mm.

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9. Method for operating a bipolar electrochemical system according to any of the preceding claims, **characterized in** that the following relation is fulfilled

$$0.001 \leq We \leq 2.0,$$

5

wherein

We = Weber number

ρ_g = gas phase (around the droplets) density

u_f = fluid velocity in the borehole

10 σ_f = fluidal surface tension

d_h = borehole diameter

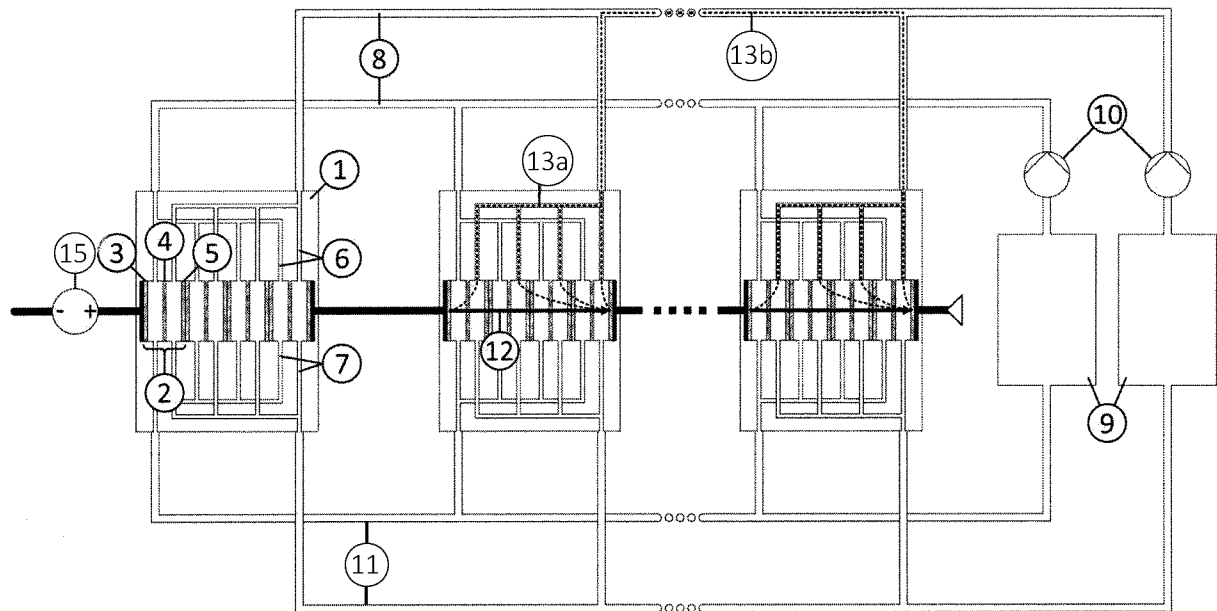


FIG. 1 (PRIOR ART)

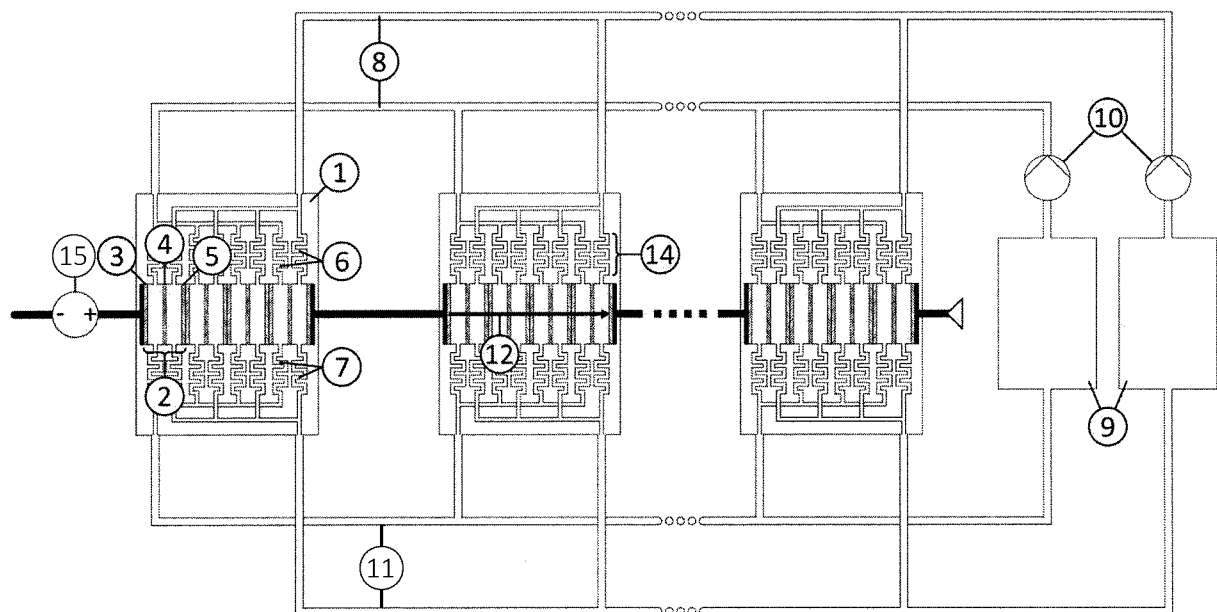


FIG. 2 (PRIOR ART)

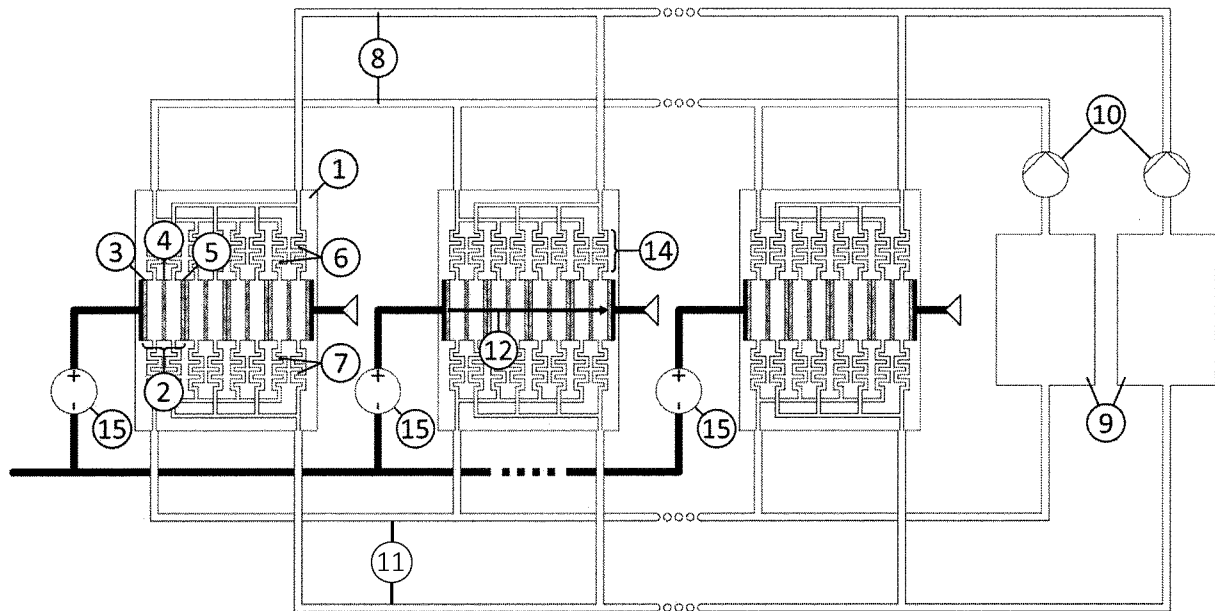


FIG. 3 (PRIOR ART)

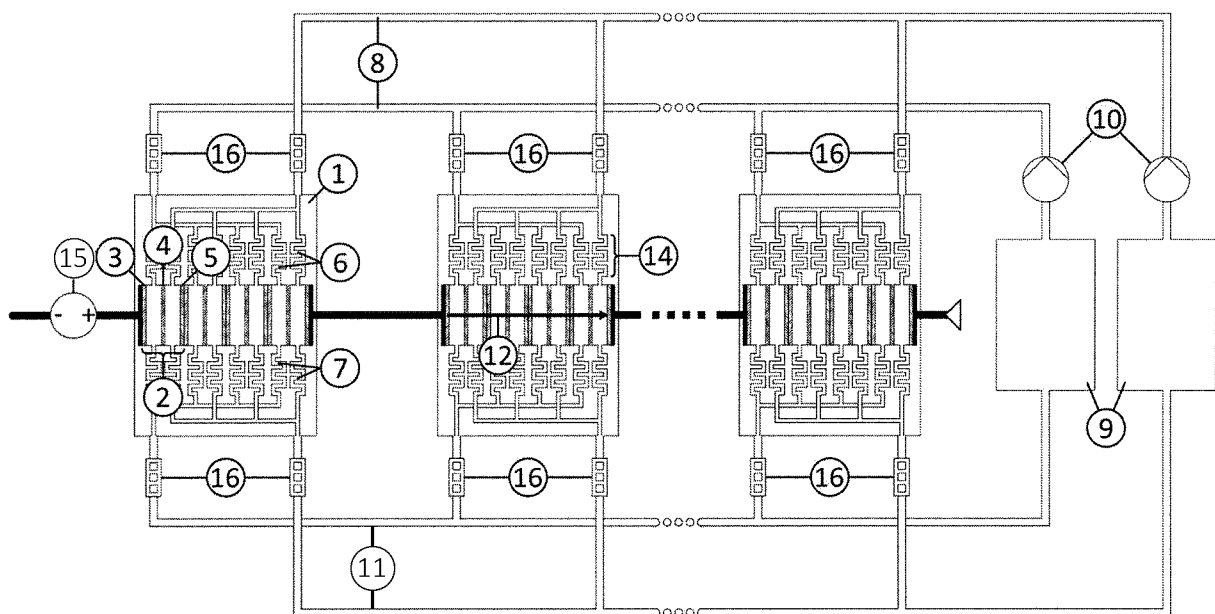


FIG. 4 (PRIOR ART)

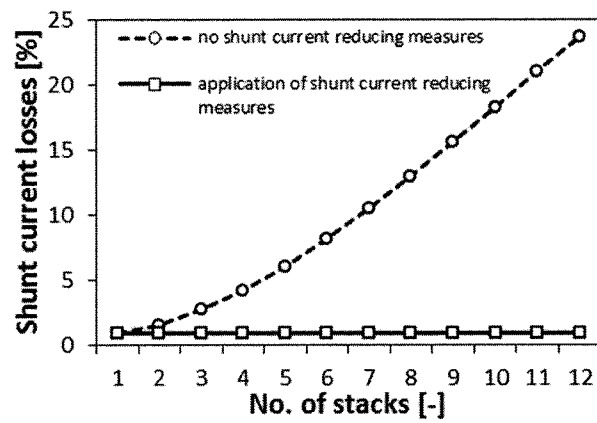


FIG. 5

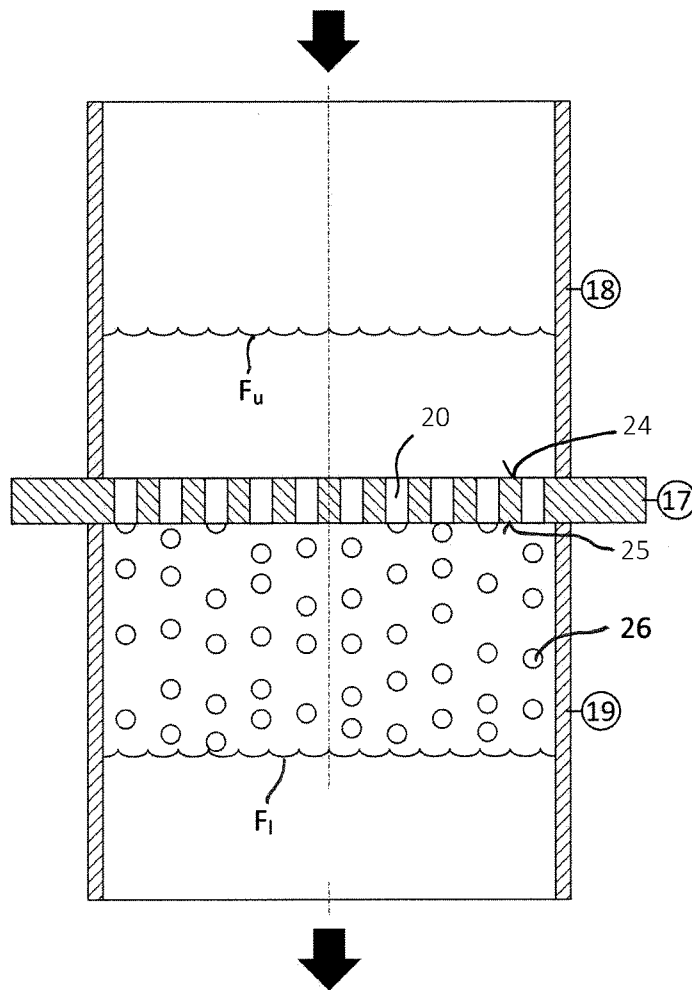
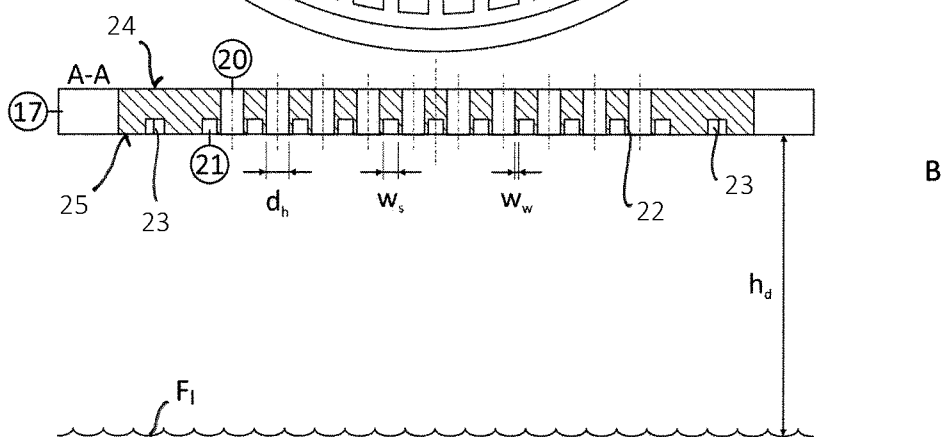
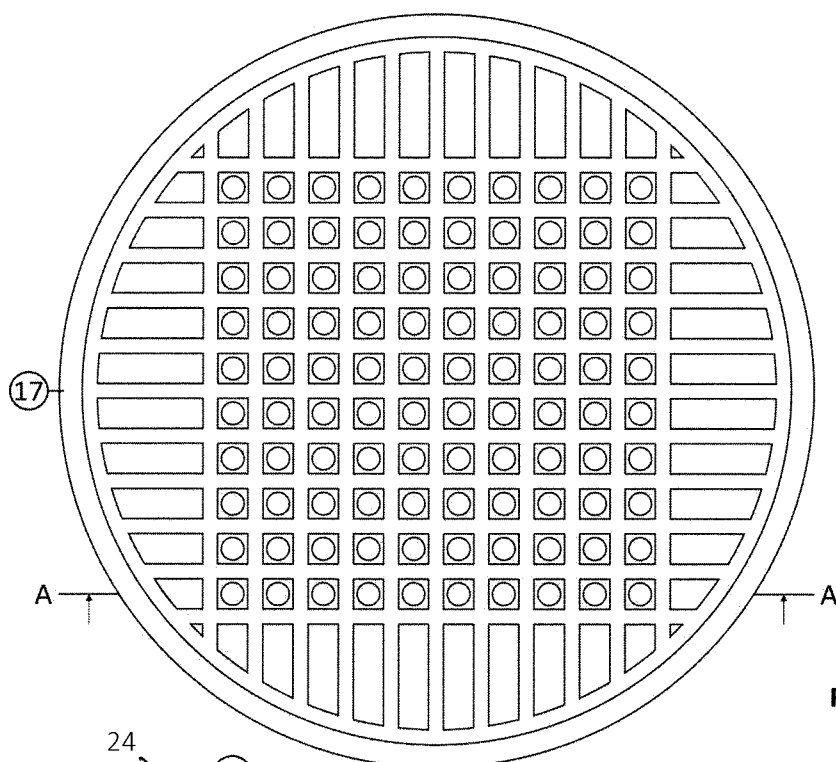
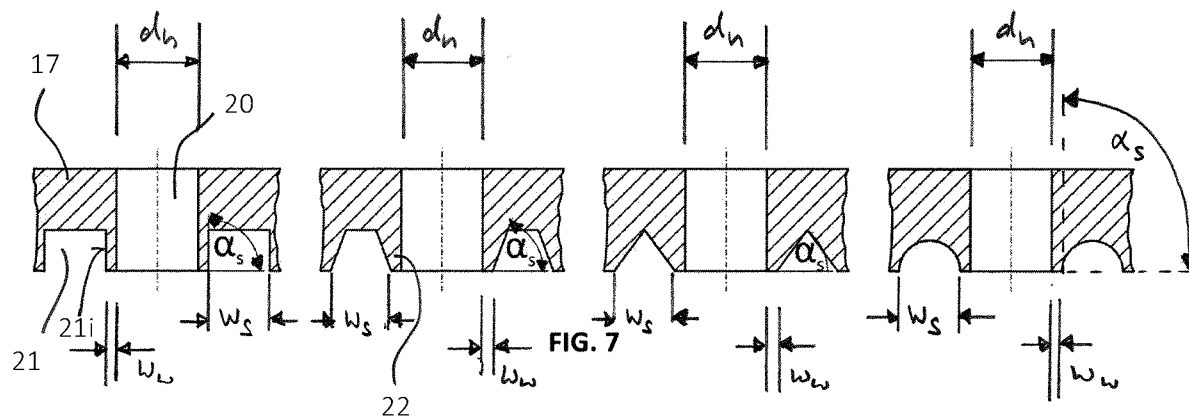


FIG. 6



INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2015/052860

A. CLASSIFICATION OF SUBJECT MATTER

INV. H01M4/66 H01M10/04 H01M10/0585
ADD.

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
H01M

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

EPO-Internal, COMPENDEX, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 2014/065460 A1 (EVANS CRAIG [US] ET AL) 6 March 2014 (2014-03-06)	1-3,7
Y	paragraph [0080] - paragraph [0097] paragraph [0030] - paragraph [0032] -----	1-8
X	US 4 533 455 A (BALKO EDWARD N [US] ET AL) 6 August 1985 (1985-08-06)	1,2,7
Y	column 1, line 35 - column 2, line 17 column 5, line 20 - column 6, line 33 -----	1-8



Further documents are listed in the continuation of Box C.



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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/EP2015/052860

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