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

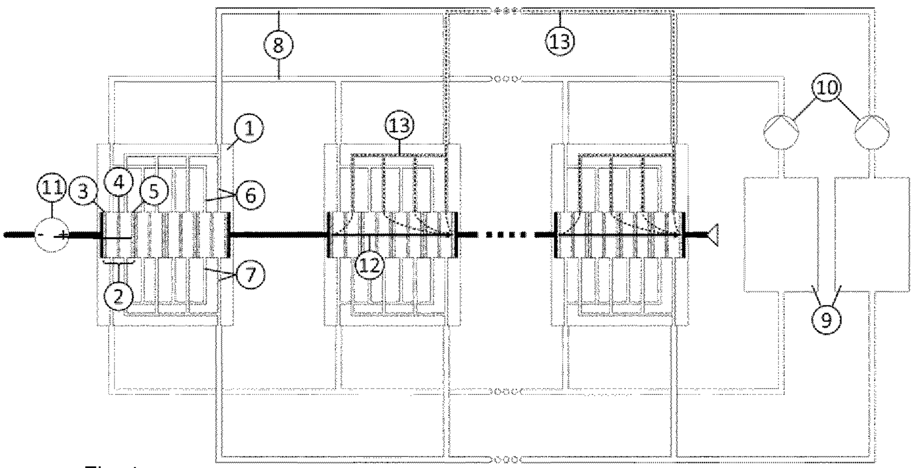

Fig. 1

(57) **Abstract:** The bipolar electrochemical system comprises at least one module (M_1 - M_n) with at least two bipolar stacks (1). Each stack (1) consists of at least two cells (2) each having an anode (3), a separator (4), a cathode (5), at least one electrolyte inlet (6) and at least one electrolyte outlet (7) opposed to the respective electrolyte inlet (6). There is a serial electrical connection (X) between the stacks (1) in the respective module (M_1 - M_n) with a fluidic connection (Y) directed from one electrolyte inlet (6) to the respective electrolyte outlet (7) for the electrolyte in at least one stack (1) of at least one module (M_1 - M_n). The serial electrical connection (X) is perpendicularly to the fluidic connection (X).

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BIPOLAR ELECTROCHEMICAL SYSTEM

The invention relates to a bipolar electrochemical system comprising at least one module with at least two bipolar stacks wherein each stack consists of at least two cells each having an anode, a separator, a cathode, at least one electrolyte inlet and at least one electrolyte outlet opposed to the respective electrolyte inlet, wherein there is a serial electrical connection between the stacks in the respective module, and wherein there is a fluidic connection directed from one electrolyte inlet to the respective electrolyte outlet for the electrolyte in at least one stack of at least one module.

Background of the Invention

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. An example is a Vanadium Redox Flow Battery using two different liquid electrolytes, but the invention applies to all bipolar electrochemical systems which 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 or an inverter and a transformer, depending on the direction of the flow of the electric energy.

Potential differences result in 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. This results in the formation of shunt currents 13a between the single cells of each stack across the electrolyte and also shunt currents between the stacks of the bipolar electrical system. Shunt currents in electrochemical systems leads to high current losses and thus lower their 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 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.

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 connection 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.

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.

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 number 16 depicts the fluid based shunt current interrupters.

Both fundamental principles need additional equipment, e.g. shunt current interrupters within the electrolyte channels or several transformers/ converters/ 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.

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 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 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 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 of stacks needs expensive additional installation costs of transformers/ converters for every stack.

Summary of the Invention

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 features of claim 1. Therein, a bipolar electrochemical system comprising at least one module with at least two bipolar stacks is claimed. Each stack consists of at least two cells. Each cell in turn features an anode, a separator, a cathode, at least one electrolyte inlet and at least electrolyte outlet opposed to the respective electrolyte inlet. Between all stacks of one module, a serial electrical connection is provided. Further a fluidic connection directed from one electrolyte inlet to the respective electrolyte outlet for the electrolyte is established in at least one stack of at least one module.

Preferably, this fluid connection is formed as a straight line between is the electrolyte inlet to the respective electrolyte outlet.

The essential feature of the invention is that the serial electrical connection is arranged perpendicular to the fluidic connection.

The fluidic connection and the electrical connection form two straight lines. Where these two lines intersect at a point, four angles are formed. Perpendicularly in the sense of the invention means, that two angles opposite each other are between 75 and 90 °, preferably 85 and 90 °, most preferably all four angles are 90 °.

Thereby, a fluid connection between stacks of extensive different electric potential level and as a result a shunt current extinction is prevented. Further, the bipolar electrochemical system according to the invention is very simple in its set-up and obviates the need to include additional, complicated and expensive equipment like shunt current interrupters or converters.

Preferably, there is a fluidic connection for all stacks in all modules that have an equal or very similar electric potential. This eliminates shunt currents due to a lack of potential difference between fluidically connected stacks. Especially, there is a separate fluidic connection for stacks having an electrical potential that varies from the electrical potential of other stacks by at least 10 V . This design prevents a fluid connection between stacks of extensively different electric potential levels, resulting in the avoidance of shunt currents since these require a fluidic connection between different stacks.

In an embodiment of the invention, the electrolyte inlets of two adjacent stacks in at least one module are fluidically connected in a parallel connection. Thereby, the number pumping system including an electrolyte storage tank, a pump and a tank for spent electrolyte is reduced.

A further embodiment of the invention there is a first electrolyte inlet (6) and a first electrolyte outlet opposed to the first electrolyte inlet for the anolyte and a second electrolyte inlet and a second electrolyte outlet opposed to the second electrolyte inlet for the catholyte. Thereby, the inventive system can be used for cells with different catholyte and anolyte.

Moreover, a first fluid connection between the first electrolyte inlet and the first electrolyte outlet and a second fluid connection between the second electrolyte inlet and the second electrolyte outlet have preferably the same flow direction to adjust the electrical connection.

In another embodiment of invention, the separator is a membrane. A membrane is a selective barrier, whereby ions are delivered selectively from the anodic part comprising the anode and a cathodic part comprising the cathode.

It is also possible that each cell in is divided liquid-impermeably into an anodic part comprising the anode and a cathodic part comprising the cathode.

Further, it is preferred that each module comprises 4 to 100, preferably 4 to 20 stacks and/or each stack comprises 10 to 200, preferably 20 to 100.

Moreover, a preferred system contains at least one electrolyte cycle comprising a reservoir of electrolyte, at least one pumping mechanism and a tank for spent electrolyte as well as corresponding fluid lines to ensure a steady electrolyte concentration.

Preferably, such a system contains one electrolyte cycle for the anolyte and one electrolyte cycle for the catholyte.

In another embodiment of the invention, the bipolar electrochemical system each module is connected to a DC/DC converter. A DC-to-DC converter is an electronic circuit which converts a source of direct current (DC) from one voltage level to another direct current (DC).

Linear DC/DC converter can only output at lower voltages from the input. Electronic switch-mode DC to DC converters convert one DC voltage level to another, by storing the input energy temporarily and then releasing that energy to the output at a different voltage. The storage may be in either magnetic field storage components (inductors, transformers) or electric field storage components (capacitors). This conversion method is more power efficient (often 75% to 98%) than linear voltage regulation.

In magnetic DC-to-DC converters, energy is periodically stored into and released from a magnetic field in an inductor or a transformer, typically in the range from 300 kHz to 10 MHz. By adjusting the duty cycle of the charging voltage (that is, the ratio of on/off time), the amount of power transferred can be controlled.

Switched capacitor converters rely on alternately connecting capacitors to the input and output in differing topologies. For example, a switched-capacitor reducing converter might charge two capacitors in series and then discharge them in parallel. This would produce an output voltage of half the input voltage, but at twice the current (minus various inefficiencies).

Most DC-to-DC converters are designed to move power in only one direction, from the input to the output. However, all switching regulator topologies can be made bi-directional by replacing all diodes with independently controlled active rectification. A bi-directional converter can move power in either direction, which is useful in applications requiring regenerative braking.

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

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,

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,

Fig. 5 schematically depicts a design of an electrochemical system according to the invention applied to one module,

Fig. 6 schematically depicts a design of an electrochemical system according to the invention applied to multiple modules,

Fig. 7 shows an alternative design of an electrochemical system according to the invention applied to multiple modules and

Fig. 8 shows cumulative shunt currents as function of stack number,

The design of the bipolar electrochemical system according to the invention can be applied to one module M_1 as shown in figure 5.

Each bipolar stack 1 comprises at least two cells. All bipolar stacks 1 are electrically connected which is depicted with line X.

The electrically connected stacks 1 form a module M_1whereby the stacks in the modules have an equal or very similar electric potential with a difference ox macimum 10 V. The electrical connection X is fed with the converter 15, preferably a DC/DC converter.

Further, the stack shows one electrolyte inlet 6 for the anolyte and one electrolyte inlet 6' for the catholyte as well as the corresponding electrolyte outlet 7 and 7'. Between the anolytic electrolyte inlet 6 and the anolytic electrolyte outlet 7 as well as between the catholytic electrolyte inlet 6' and the catholytic electrolyte outlet 7 a fluid connection Y is established.

The 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.

This fluid connection Y is positioned such that it is perpendicular to the electric connection X.

The invention also covers multiple modules as shown in figure 6. In the set-up according to figure 6, the electrical connection X is done perpendicularly in series according to the usual approach in the state of the art, while the respective fluidic stack connection Y is done perpendicularly, connecting stacks of different electrical strings. The perpendicular fluidic stack connection Y exclusively links stacks 1 which are of the same electric potential level, thereby eliminating shunt currents due to a lack of potential difference between fluidically connected stacks.

Furthermore, it is within the scope of the invention to fluidically connect two adjacent stacks 1 in each module M_{1-n} as shown in figure 7.

Figure 8 shows the maximum loss caused by shunt currents relating to the overall voltage of all stack. It is obviously, that with increasing stack number, plotted on the X-axis, the maximum loss increases nearly linear. For a system featuring 12 stacks, the overall loss is about 24 %.

In comparison, an inventive circuit only shows a loss of about 1 %, independent from the number of stacks.

List of reference numbers

1	bipolar stack
2	cell
3	anode
4	separator
5	cathode
6	electrolyte inlet
7	electrolyte outlet
8	electrolyte supply circuit
9	storage tank
10	fluid conveying system
11	electrolyte withdrawal circuit
12	main current flow direction
13	shunt current
14	meander structure
15	DC/DC converter
X	electrical connection
Y	fluid connection

Claims:

1. Bipolar electrochemical system comprising at least one module (M_1 - M_n) with at least two bipolar stacks (1) wherein each stack (1) consists of at least two cells (2) each having an anode (3), a separator (4), a cathode (5), at least one electrolyte inlet (6) and at least one electrolyte outlet (7) opposed to the respective electrolyte inlet (6), wherein there is a serial electrical connection (X) between the stacks (1) in the respective module (M_1 - M_n), and wherein there is a fluidic connection (Y) directed from one electrolyte inlet (6) to the respective electrolyte outlet (7) for the electrolyte in at least one stack (1) of at least one module (M_1 - M_n), **characterized in** that the serial electrical connection (X) is perpendicularly to the fluidic connection (X).

2. The bipolar electrochemical system according to claim 1, **characterized in** there is a fluidic connection (X) for all stacks (1) in all modules (M_1 - M_n) that have an equal or very similar electric potential.

3. The bipolar electrochemical system according to any of the preceding claims, **characterized in** that there is a separate fluidic connection (Y) for stacks (1) having an electrical potential that varies from the electrical potential of other stacks (1) by at least 10 V.

4. The bipolar electrochemical system according to any of the preceding claims, **characterized in** that a fluidic connection (Y) is provided for those stacks (1) only that differ in their electrical potential by at most 10 V.

5. The bipolar electrochemical system according to any of the preceding claims, **characterized in** that the electrolyte inlets (6) of two adjacent stacks (1) in at least one module (M_1 - M_n) are fluidically connected in a parallel connection.

6. The bipolar electrochemical system according to any of the preceding claims, **characterized in** that there is a first electrolyte inlet (6) and a first electrolyte outlet (7) for the anolyte and a second electrolyte inlet (6') and a second electrolyte outlet (7') for the catholyte.

7. The bipolar electrochemical system according to claim 6, **characterized in** that the fluid connection between the first electrolyte inlet (6) and the first electrolyte outlet (7) and the fluid connection between the second electrolyte inlet (6) and the second electrolyte outlet (7) have the same flow direction.

8. The bipolar electrochemical system according to any of the preceding claims, **characterized in** that the separator (4) is a membrane.

9. The bipolar electrochemical system according to any of claims 1 to 6, **characterized in** that each cell (2) is divided impermeably into an anodic part comprising the anode and a cathodic part comprising the cathode.

10. The bipolar electrochemical system according to any of the preceding claims, **characterized in** that each module comprises 40 to 100 stacks.

11. The bipolar electrochemical system according to any of the preceding claims, **characterized in** that each stack comprises 10 to 200 cells.

12. The bipolar electrochemical system according to any of the preceding claims, **characterized in** that the system contains at least one electrolyte cycle comprising a reservoir of electrolyte, at least one pumping mechanism and a tank for spent electrolyte.

13. The bipolar electrochemical system according to any of the preceding claims, **characterized in** that the system contains one electrolyte cycle for the anolyte and one electrolyte cycle for the catholyte.

14. The bipolar electrochemical system according to any of the preceding claims, **characterized in** that each module is connected to a DC/DC converter (15).

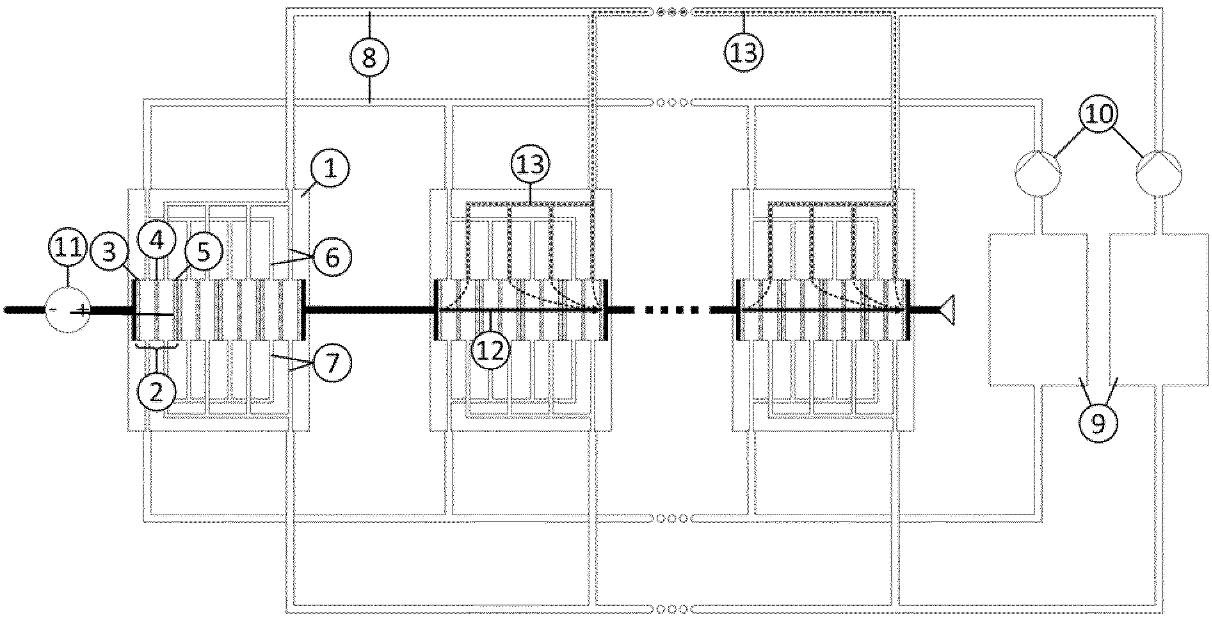

Fig. 1

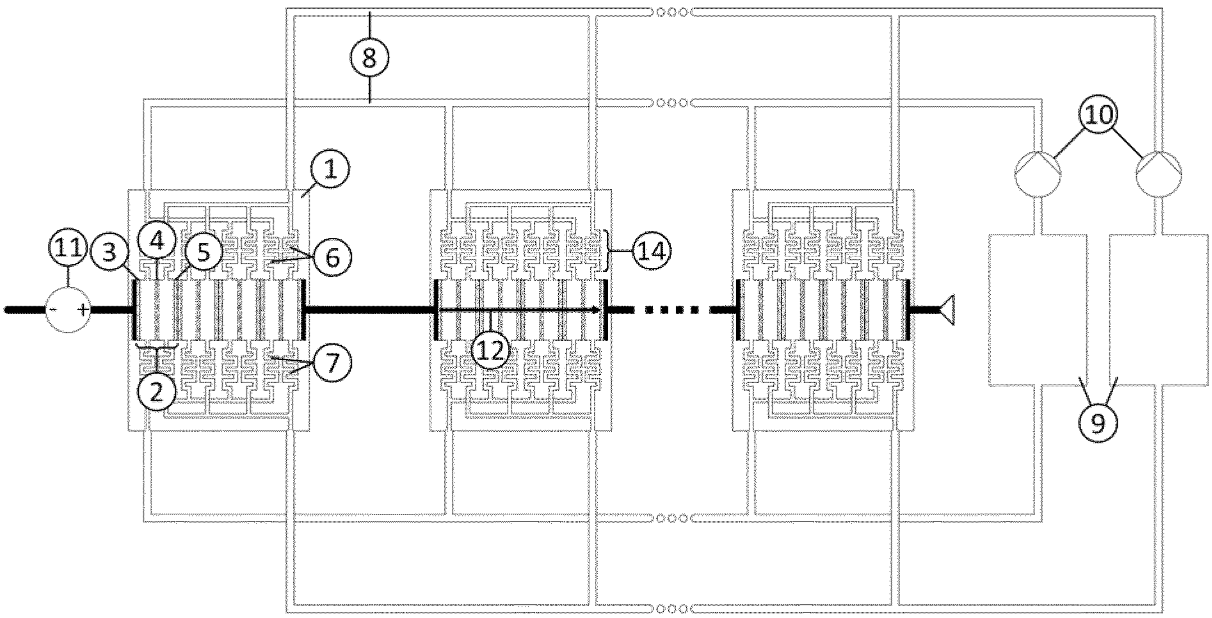

Fig. 2

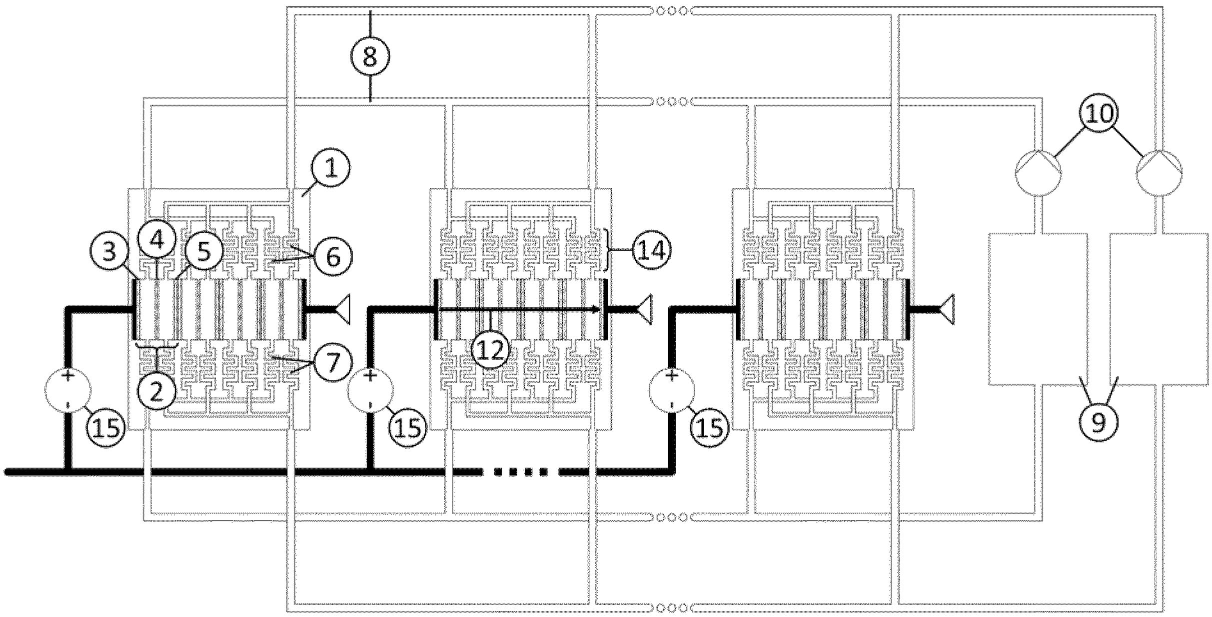
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Fig. 3

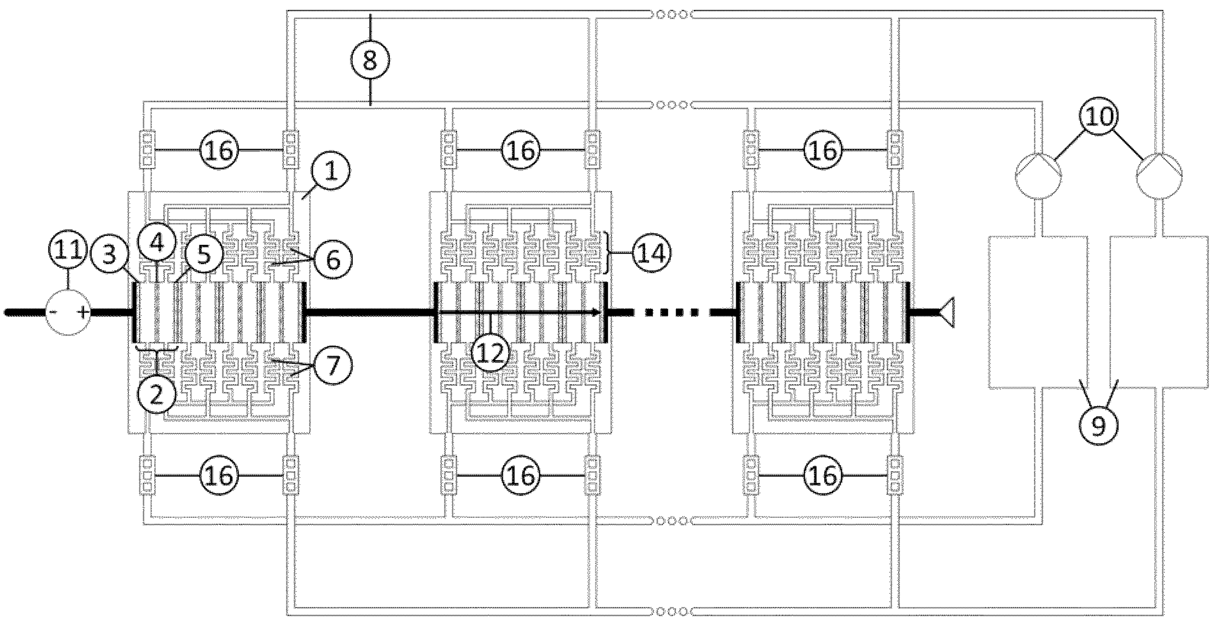
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Fig. 4

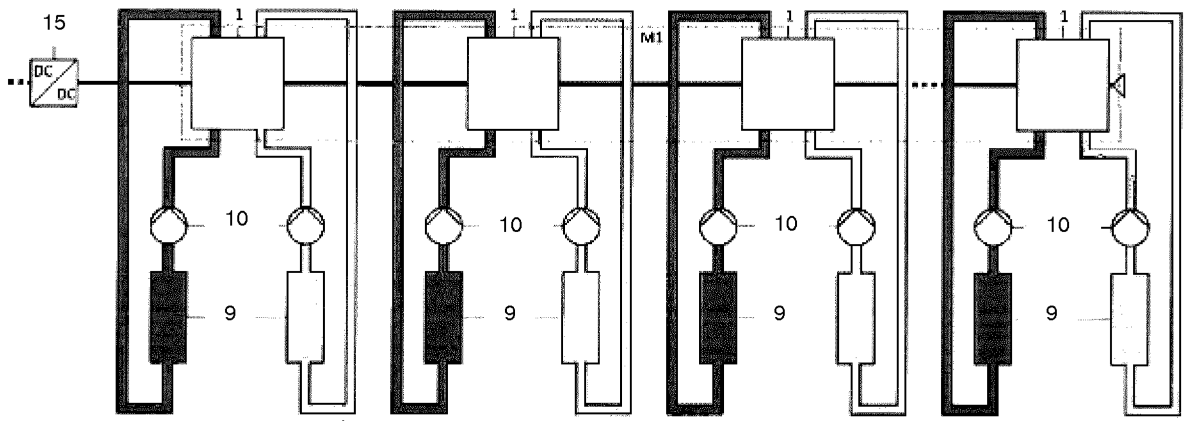


Fig. 5

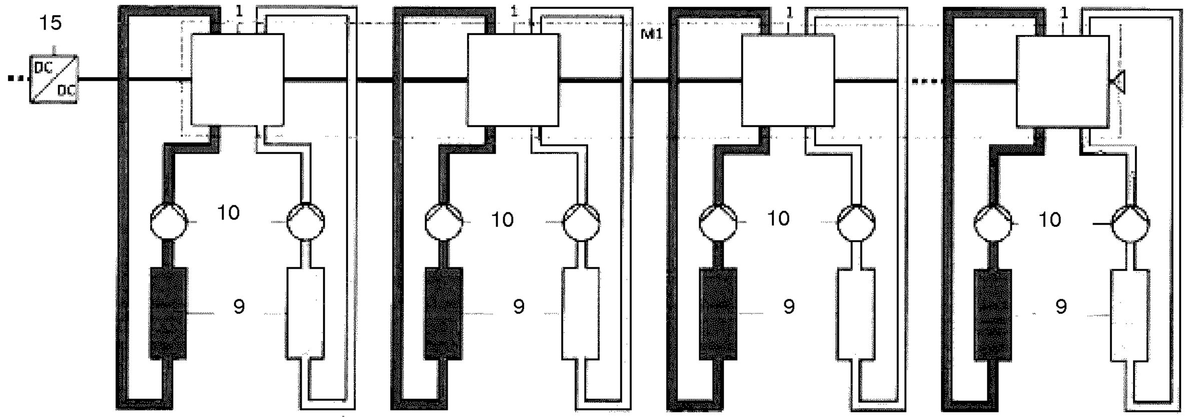


Fig 6

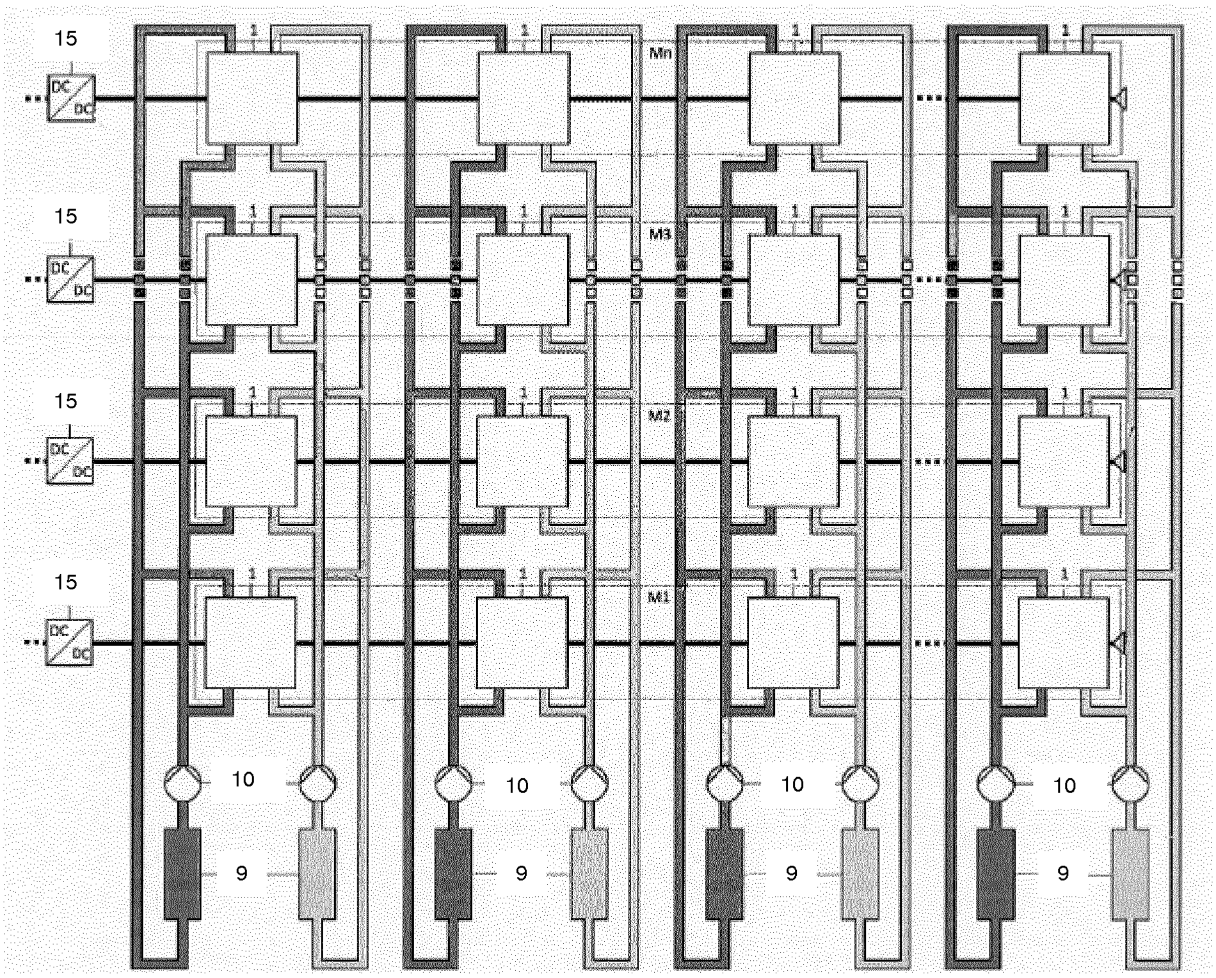
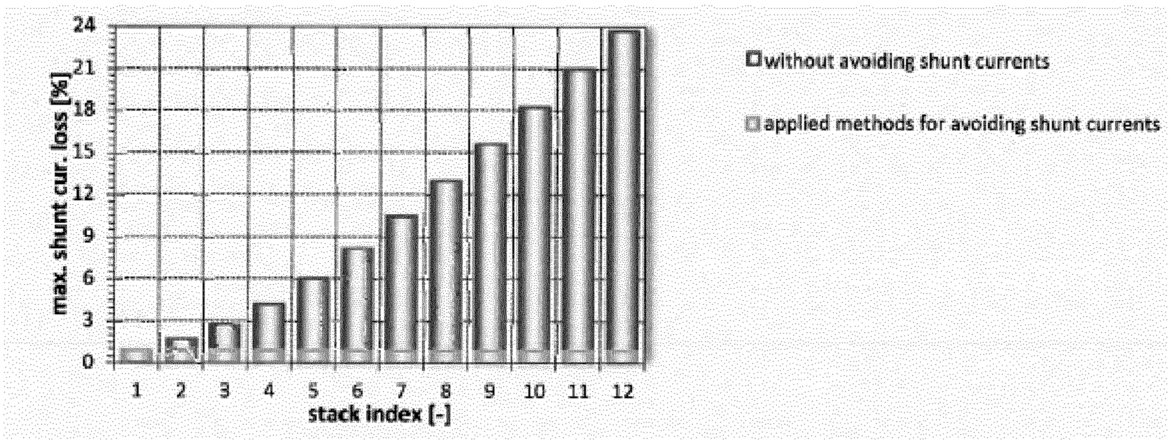


Fig. 7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15
12
9
6
3
0
max. shunt cur. loss [%]
1 2 3 4 5 6 7 8 9 10 11 12
stack index [-]
without avoiding shunt currents
applied methods for avoiding shunt currents

Fig. 8

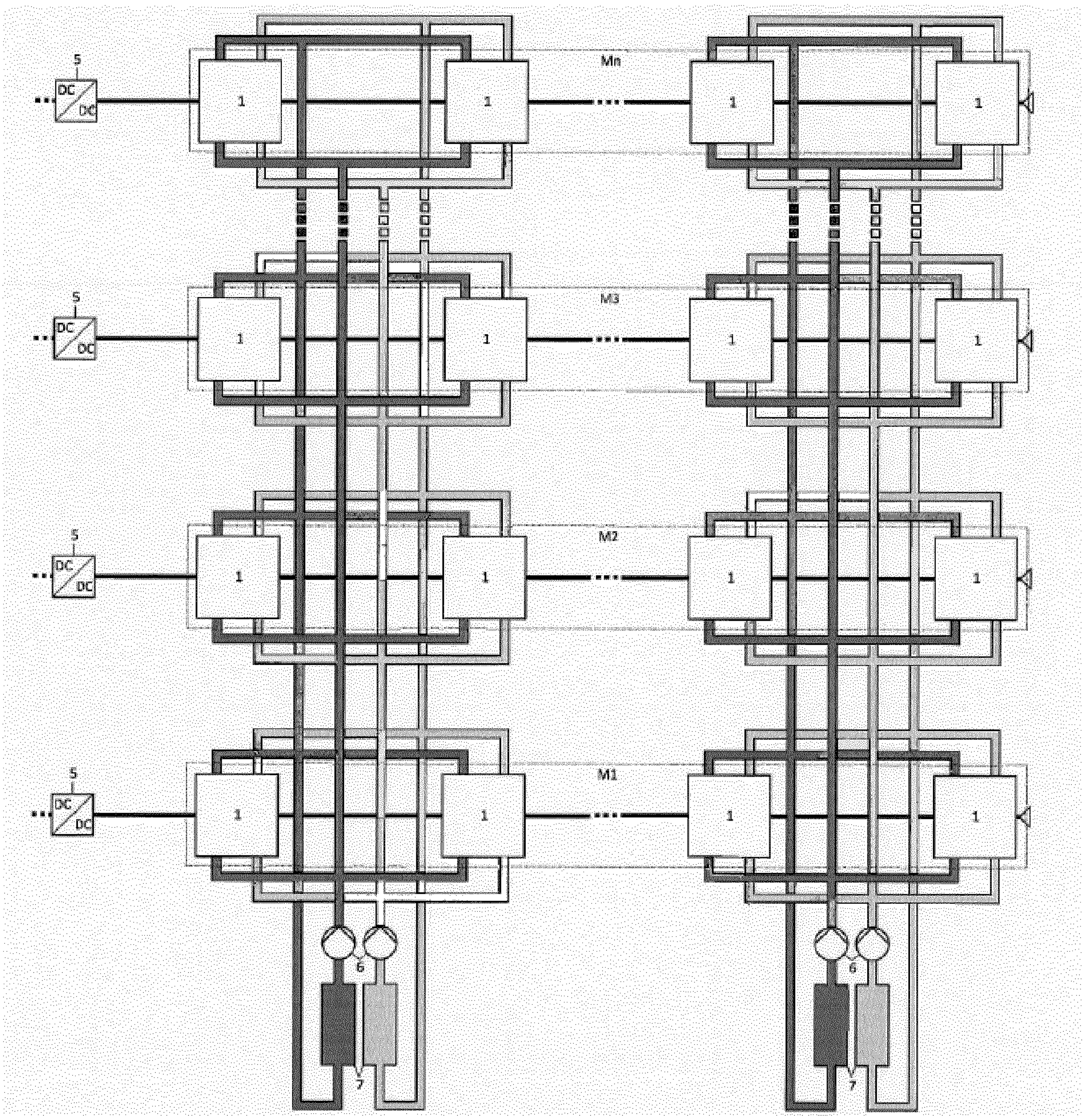

Fig. 9

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2016/077688

A. CLASSIFICATION OF SUBJECT MATTER
INV. H01M8/18 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, WPI Data, INSPEC

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X Y	WO 2014/145844 A1 (UNIENERGY TECHNOLOGIES LLC [US]) 18 September 2014 (2014-09-18) page 10, line 3 - page 11, line 30; figures 2, 13, 14	1,2,5-9, 12-14 10,11
X Y	WO 2016/128038 A1 (OUTOTEC OY [FI]) 18 August 2016 (2016-08-18) page 2, line 3 - page 5, line 21; figures 1-4	1,2,5-9, 12-14 10,11
X	US 2014/272484 A1 (LI LIYU [US] ET AL) 18 September 2014 (2014-09-18) paragraph [0063] - paragraph [0070]; figures 2, 4, 5	1,2,5-9, 12-14
X	WO 2007/131250 A1 (FUNKTIONSWERKSTOFFE FORSCHUNGS [AT]; WHITEHEAD ADAM HARDING [AT]; BALV) 22 November 2007 (2007-11-22) page 4, line 9 - page 6, line 34	1,2,5-9, 12-14

☐ Further documents are listed in the continuation of Box C.

☒ See patent family annex.

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"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
18 May 2017	30/05/2017

Name and mailing address of the ISA/	Authorized officer
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1

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/EP2016/077688

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US 2014272484	A1	18-09-2014	NONE		
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