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(54) **METHOD FOR BOOSTING A REDOX FLOW BATTERY, A MEMBRANE DEVICE, A MEMBRANE STACK, AND A SYSTEM TO PERFORM SAID METHOD**

(57) The invention relates to a method, device, membrane stack, and system for boosting a redox flow battery. The comprises the steps of:
- providing the redox flow battery with the membrane device comprising:
- *n* triplet of membranes, wherein *n* is an integer of 1 or more, and wherein each triplet of membranes consists of an anion exchange membrane, a bipolar membrane, and a cation exchange membrane; and
- 3*n* + 1 compartments, wherein one of the compartments comprises a cathode and wherein another of the compartments comprises an anode, and wherein the 3*n* + 1 compartments are at least partly delineated by one of the triplet of membranes;
- circulating a catholyte to the compartment with the cathode;
- circulating an anolyte to the compartment with the anode; and
- charging the redox flow battery by applying an electrical potential difference between the anode and the cathode and/or providing charged catholyte to the compartment with the cathode and anolyte to the compartment with the anode

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Dit octrooi is verleend ongeacht het bijgevoegde resultaat van het onderzoek naar de stand van de techniek en schriftelijke opinie. Het octrooischrift komt overeen met de oorspronkelijk ingediende stukken.

METHOD FOR BOOSTING A REDOX FLOW BATTERY, A MEMBRANE DEVICE, A MEMBRANE STACK, AND A SYSTEM TO PERFORM SAID METHOD

The present invention relates to a method for boosting a redox flow battery with a membrane device, a device, a membrane stack, and a system to perform said method.

The increasing energy demand and the impact of said energy demand on the environment are rising. Said issues pose a significant thread to the sustainable development of renewable energy sources. It is known that renewable energy sources such as wind and solar presents a barrier to their large-scale implementation. Furthermore, fluctuations in renewable energy production could endanger the power grid if directly integrated. A problem affiliated with the large-scale implementation is that efficient and effective means for electricity storage over periods of time are expensive and/or use a lot of space. The (temporarily) storage of a surplus of energy could be *via* electrochemistry, wherein the stored charge can be converted back to electrical power with high round-trip efficiency.

Alternatively, aqueous redox flow batteries may be used for storing and providing (stored) energy. To compete with alternative storage methods, improvements in energy and power density are needed.

An objective of the present invention is to provide a method for boosting a redox flow battery with a membrane device that obviates or at least reduces one or more of the aforementioned problems and/or is more effective as compared to conventional methods and systems.

This objective is achieved with a method for boosting a redox flow battery with a membrane device comprising the steps of:

- providing the redox flow battery with the membrane device comprising:
    - n triplet of membranes, wherein n may be an integer of 1 or more, and wherein each triplet of membranes consists of an anion exchange membrane, a bipolar membrane, and a cation exchange membrane; and
    - $3n + 1$ compartments, wherein one of the compartments comprises a cathode and wherein another of the compartments comprises an anode, and wherein the $3n + 1$ compartments are at least partly delineated by one of the triplet of membranes;
- circulating a catholyte to the compartment with the cathode;
- circulating an anolyte to the compartment with the anode; and
- charging the redox flow battery by applying an electrical potential difference between the anode and the cathode and/or providing charged catholyte to the compartment with the cathode and anolyte to the compartment with the anode.

The method according to the invention may start with the step of providing the redox flow battery with the membrane device. Said membrane device may comprise n triplet of membranes, wherein n is an integer of 1 or more, and wherein each triplet of membranes consists of an anion exchange membrane, a bipolar membrane, and a cation exchange membrane, and $3n + 1$ compartments, wherein one of the compartments comprises a cathode and wherein another of the compartments comprises an anode, and wherein the $3n + 1$ compartments are at least partly delineated by one of the triplet of membranes.

It is noted that n is an integer, thus, n is a number that is not a fraction but a whole number. Providing n triplet of membranes includes three membranes for each triplet. In other words, a triplet of membranes consists of three membranes.

The membrane device comprises n triplet of membranes and $3n + 1$ compartments. For example, when $n = 1$ the device has 4 compartments. In an embodiment wherein $n = 1$, the membranes and compartments are preferably configured such that the compartment comprising the cathode is at least partly delineated by the anion exchange membrane and is adjacent to the compartment which is at least partly delineated by the anion exchange membrane and the bipolar membrane. Adjacent to said compartment is a further compartment which is partly delineated by the bipolar membrane and the cation exchange membrane. Adjacent to this compartment, the compartment which is at least partly delineated by the cation exchange membrane and comprising the anode is configured.

It is noted that other configurations of the compartments are also possible, and that n may be another (integer) value.

The step of providing the redox flow battery with the membrane device may be followed by the step of circulating a catholyte to the compartment with the cathode and the step of circulating an anolyte to the compartment with the anode. Furthermore, said circulating steps may be followed by charging the redox flow battery by applying an electrical potential difference between the anode and the cathode and/or providing charged catholyte to the compartment with the cathode and anolyte to the compartment with the anode.

Circulating the catholyte to the compartment with the cathode and/or circulating the anolyte to the compartment with the anode may include providing the catholyte and/or anolyte from a catholyte storage and/or anolyte storage respectively.

Furthermore, charging the redox flow battery by applying an electrical potential difference between the anode and the cathode and/or providing charged catholyte to the compartment with the cathode and anolyte to the compartment with the anode includes that energy is provided to the catholyte and anolyte respectively. For example, the catholyte and/or anolyte may be charged separately from the redox flow battery and as a ‘ready to go’ solution (solution suitable to be discharged) provided to the compartment with the cathode or compartment with the anode respectively.

An advantage of the method according to the invention is that the gross output power of the redox flow battery at the same applied current may be boosted. As a result, the working current of the redox flow battery may be reduced without compromising the gross power output of the cell.

Therefore, the method according to the invention is suitable for us with low open circuit potentials and electrolyte solubility.

A further advantage of the method according to the invention is that energy may be stored and released efficiently and effectively. As a result, the method according to the invention can efficiently contribute to balance an open grid energy network. It was found that the method according to the invention can balance a wider area of the power grid.

Furthermore, the method according to the invention provides a higher battery potential compared to conventional redox flow batteries. As a result, the power density of the redox flow batteries is increased and a cost-effective energy storage is achieved.

In a preferred embodiment, the membrane device consists of *n* triplet of membranes. Said membrane device consists of a triplet of membranes multiplied by *n*.

In a preferred embodiment, the method according to the invention comprises charging the redox flow battery by applying an electrical potential difference between the anode and the cathode.

It was found that charging the redox flow battery by applying an electrical potential difference between the anode and the cathode provides an efficient and effective method for boosting a redox flow battery. Furthermore, applying an electrical potential difference between the anode and the cathode reduces the amount of (external) transport of the anolyte and/or catholyte. Furthermore, safety is increased as an operator performs less handlings and is less exposed to (potential) harmful/hazardous chemicals such as chemicals forming the anolyte and/or catholyte.

In a presently preferred embodiment according to the invention, the method further comprises the step of discharging the redox flow battery. It is noted that said step may also be referred to as discharging the anolyte and catholyte.

The step of discharging the anolyte and catholyte enables to provide energy, such as electricity, to an open or closed power grid, machine, and the like.

Furthermore, the step of discharging enables to form water at the bipolar membrane, and the step of charging enables to split water at the bipolar membrane. Said reactions may be referred to as:

$$\text{Discharging: } H^+ + OH^- \rightarrow H_2O$$

$$\text{Charging: } H_2O \rightarrow H^+ + OH^-$$

An advantage of the method according to the invention including the step of discharging is that energy can be stored and released on demand. As a result, less (renewable) energy is lost, and the potential of renewable energy sources can be increased.

It was found that the method according to the invention enhances the gross power of the redox flow battery by at least 40% at the source of the generation compared to conventional methods for boosting redox flow batteries.

In a further presently preferred embodiment according to the invention, the method further comprises the steps of providing freshwater including an acid to the compartment delineated by the anion membrane and bipolar membrane and providing freshwater including a base to the compartment delineated by the cation membrane and bipolar membrane.

It is noted that throughout this application freshwater is referred to naturally occurring water that is not salty, and is suitable for consumption if cleaned or processed.

It was found that providing freshwater including an acid to the compartment delineated by the anion membrane and bipolar membrane and providing freshwater including a base to the compartment delineated by the cation membrane and bipolar membrane improves the total voltage of the redox flow battery.

In a preferred embodiment, the steps of providing freshwater including an acid to the compartment delineated by the anion membrane and bipolar membrane and providing freshwater including a base to the compartment delineated by the cation membrane and bipolar membrane are at least partly performed simultaneously with the step of discharging the anolyte and catholyte.

It was found that at least partly performing the steps of discharging the anolyte and catholyte, providing freshwater including an acid to the compartment delineated by the anion membrane and bipolar membrane, and providing freshwater including a base to the compartment delineated by the cation membrane and bipolar membrane has a synergetic effect on the total voltage of the cell and enables an efficient and effective redox flow battery.

In a further presently preferred embodiment according to the invention, the acid may be one or more selected from the group of HCl, H_2SO_4, HI, and HF, and/or the base may be sodium hydroxide (NaOH) and/or potassium hydroxide (KOH).

In a preferred embodiment according to the invention, the acid may be HCl and/or H_2SO_4, preferably the acid may be HCl, and/or the base may be NaOH

It was found that an efficient method for boosting a redox flow battery was achieved using acid and base which enable an efficient displacement reaction. For example, HCl as acid and NaOH as base, or H_2SO_4 as acid and NaOH as base, or H_2SO_4 as acid and KOH as base, or HCl as acid and KOH as base.

In a further presently preferred embodiment according to the invention, the concentration of the acid and base may be independently selected from the range of 0.5 mol L^{-1} to 3 mol L^{-1}, preferably 0.5 mol L^{-1} to 2 mol L^{-1}, more preferably 1 mol L^{-1} to 1.5 mol L^{-1}.

It is noted that the concentration of the acid is independent from the concentration of the base. In other words, the concentration of the acid and the base are not correlated to each other.

It was found that providing the acid and base independently selected from said range, an efficient and effective discharge of the redox flow battery was achieved, wherein the method according to the invention provided a stable output of energy.

In a further presently preferred embodiment according to the invention, the method further comprises the steps of providing freshwater to the compartment delineated by the anion membrane and bipolar membrane and providing freshwater to the compartment delineated by the cation membrane and bipolar membrane.

It was found that providing freshwater to the compartment delineated by the anion membrane and bipolar membrane and providing freshwater to the compartment delineated by the cation membrane and bipolar membrane improves the charge capacity and total voltage of the redox flow battery.

In a preferred embodiment, the steps of providing freshwater to the compartment delineated by the anion membrane and bipolar membrane and providing freshwater to the compartment delineated by the cation membrane and bipolar membrane are at least partly performed simultaneously with the step of charging the anolyte and catholyte.

It was found that at least partly performing the steps of charging the anolyte and catholyte, providing freshwater to the compartment delineated by the anion membrane and bipolar membrane, and providing freshwater to the compartment delineated by the cation membrane and bipolar membrane has a synergetic effect on the total charge capacity and voltage of the cell and enables an efficient and effective redox flow battery.

In a further presently preferred embodiment according to the invention, n may be an integer of 2 or more, and wherein two triplets of membranes delineate an intermediate compartment.

An advantage of n is an integer of 2 or more is that the intermediate compartment enables to remove salt from the membrane device during discharge and provide ions to the membrane device during charge. As a result, a more efficient and effective redox flow battery is achieved.

It was found that adding and/or removing the ions enables to use multiple triplets of membranes and thus increasing the capacity of the membrane device. Furthermore, removing the ions during discharge and adding ions during charge increases the life-time of the membrane device by at least 25% compared to membrane devices without removing and/or adding said ions.

In a further presently preferred embodiment according to the invention, the method further comprises the step of providing freshwater or salt water to the intermediate compartment.

The step of providing freshwater or salt water to the intermediate compartment preferably includes the step of providing freshwater or salt water in an alternating manner. In other words, the intermediate compartment may be provided with salt water followed by providing the intermediate compartment with freshwater.

In a preferred embodiment, a preferred salt is dissolved in freshwater forming the desired salt water.

For example, during discharge, using HCl as acid and NaOH as base provides (dissolved) NaCl to the intermediate compartment, or H_2SO_4 as acid and NaOH as base provides (dissolved) Na_2SO_4 to the intermediate compartment, or H_2SO_4 as acid and KOH as base provides (dissolved) K_2SO_4 to the intermediate compartment, or HCl as acid and KOH as base provides (dissolved) KCl to the intermediate compartment.

In addition, during charge, salt water is provided to the intermediate compartment. For example, the salt water may be NaCl (aq), Na_2SO_4 (aq), K_2SO_4 (aq), or KCl (aq).

An advantage of selecting the salt water provided to the intermediate compartment is that fouling and/or blocking of the membrane device is prevented.

Thus, selecting the (dissolved) acid provided to the compartment delineated by the anion membrane and bipolar membrane and the (dissolved) base provided to the compartment delineated by the cation membrane and bipolar membrane during discharge, and the salt water provided to the intermediate compartment during charging such that said acid, base, and salt do not precipitate when combined is preferred. As a result, an efficient and effective redox flow battery is achieved.

In a further presently preferred embodiment according to the invention, the catholyte comprises one or more selected from the group of $Na_4\{FeCN\}_6$, $K_3\{FeCN\}_6$, Br_2/Br^-, VO^{2+}. Preferably, the anolyte comprises one or more selected from the group of $FeCl_2$, $FeCl_3$, triethanolamine, V^{2+}/V^{3+}, H_2/H^+, Zn/Zn^{2+}.

It was found that the catholyte selected from said group and/or the anolyte selected from said group provides an efficient and effective method for boosting a redox flow battery.

For example, VO^{2+} may have chloride ions as counterion. In other words, the catholyte may be $VO^{2+}/2\ Cl^-$.

In a further presently preferred embodiment according to the invention, the anolyte and/or catholyte comprises one or more bases independently selected from potassium hydroxide and sodium hydroxide. Preferably, the concentration of the base may be in the range of 1 mol L^{-1} to 5 mol L^{-1}, preferably 2 mol L^{-1} to 4 mol L^{-1}, more preferably 2.5 mol L^{-1} to 3.5 mol L^{-1}.

An advantage of adding a base to the anolyte and/or catholyte is that the lifespan of the anolyte and/or catholyte is extended. Furthermore, the efficiency of the anolyte and/or catholyte is increased.

In a further presently preferred embodiment according to the invention, the anolyte and/or catholyte may be heated to a temperature in the range of 25 °C to 80 °C, preferably in the range of 35 °C to 60 °C, more preferably in the range of 35 °C to 50 °C.

It was found that heating the anolyte and/or catholyte enables an efficient and effective charge and discharge of the redox flow battery. As a result, the redox flow battery may provide improved power grid stability.

In a preferred embodiment, the concentration of the anolyte and catholyte are independently selected from the range of 1 mol L^{-1} to 5 mol L^{-1}, preferably in the range of 1.5 mol L^{-1} to 4 mol L^{-1}, more preferably in the range of 2 mol L^{-1} to 3.5 mol L^{-1}. Preferably, the anolyte and/or catholyte provided in said concentrations are heated to a temperature in the range of 25 °C to 80 °C.

In a further presently preferred embodiment according to the invention, circulating the anolyte and circulating the catholyte is independently performed with a flushing rate in the range of 50 L min^{-1} per cubic decimetre of the compartment to 500 L min^{-1} per cubic decimetre of the compartment the anolyte or catholyte is circulated in, preferably in the range of 100 L min^{-1} per cubic decimetre of the compartment to 450 L min^{-1} per cubic decimetre of the compartment the anolyte or catholyte is circulated in, more preferably in the range of 250 L min^{-1} per cubic decimetre of the compartment to 450 L min^{-1} per cubic decimetre of the compartment the anolyte or catholyte is circulated in.

It is noted that circulating the anolyte and circulating the catholyte may be performed at different rates.

Flushing with the above mentioned rates, reduces the build up of undesired particles in the various compartments.

The invention also relates to a device for boosting a redox flow battery, configured to perform the method according to the invention, comprising:

- *n* triplet of membranes, wherein *n* may be an integer of 1 or more, and wherein each triplet of membranes consists of an anion exchange membrane, a bipolar membrane, and a cation exchange membrane;
- $3n + 1$ compartments, wherein one of the compartments comprises a cathode and wherein another of the compartments comprises an anode, and wherein the $3n + 1$ compartments are at least partly delineated by one of the triplet of membranes; and
- means for applying an electrical potential difference between the cathode and the anode,

wherein the compartment comprising the anode is configured to, during use, be provided with an anolyte and the compartment comprising the cathode is configured to, during use, be provided with a catholyte.

It is noted that the device comprises triplets of membranes. Preferably, the amount of membranes is a multiple of three.

The device for boosting a redox flow battery, configured to perform the method according to the invention provides the same effects and advantages as those described for the method for boosting a redox flow battery according to the invention.

An advantage of the device according to the invention is that efficient charging and discharging is enabled. Therefore, a robust device is achieved which has an extended lifespan compared to conventional redox flow batteries.

Furthermore, the device according to the invention may be easily coupled to a power grid, enabling to balance said power grid efficiently and effectively.

In a presently preferred embodiment according to the invention, *n* is an integer of 2 or more, and wherein two triplets of membranes delineate an intermediate compartment.

The device according to the invention wherein *n* is an integer of 2 or more enables a device comprising one or more intermediate compartments. Said configuration increases the applicability of the device according to the invention. For example, the (energy) capacity of the device increases. As a result, de device according to the invention may be included in means for stabilizing the power grid.

In a further presently preferred embodiment according to the invention, the intermediate compartment is configured to, during use in a discharge state, be provided with freshwater, and configured to, during use in a charge state, be provided with salt water.

The invention also relates to a membrane stack for boosting a redox flow battery, the stack comprising a number of cells which are configured to perform the method according to the invention.

The membrane stack for boosting a redox flow battery provides the same effects and advantages as those described for the method for boosting a redox flow battery according to the invention and the device for boosting a redox flow battery.

The invention also relates to a system for boosting a redox flow battery, comprising:

- a device according to the invention;
- a first pump configured for circulating an anolyte;
- a second pump configured for circulating a catholyte;
- an anolyte storage which is operatively coupled with an anolyte buffer of the device and the first pump; and
- a catholyte storage which is operatively coupled with a catholyte buffer of the device and the second pump.

The system for boosting a redox flow battery provides the same effects and advantages as those described for the method for boosting a redox flow battery according to the invention, the device for boosting a redox flow battery, and the membrane stack for boosting a redox flow battery.

An advantage of the system according to the invention is that a stable power grid is achieved.

It is noted that an anolyte buffer is a storage of charged or discharged anolyte, and that a catholyte buffer is a storage of charged or discharged catholyte.

In a presently preferred embodiment according to the invention, the means to provide an electrical potential difference is a windmill, a solar panel, and/or tidal power station.

Further advantages, features and details of the invention are elucidated on the basis of preferred embodiments thereof, wherein reference is made to the accompanying drawings, in which:

- Figure 1 shows a schematic overview of the method according to the invention;
- Figure 2 shows a schematic overview of the membrane device according to the invention comprising one triplet of membranes in the discharging state;
- Figure 3 shows a schematic overview of the membrane device according to the invention comprising two triplet of membranes in the discharging state;
- Figure 4 shows a schematic overview of the membrane device according to the invention comprising multiple triplet of membranes;
- Figure 5A shows a profile of a current sweep;
- Figure 5B shows the corresponding voltage profiles of the current sweep;
- Figure 6A shows a profile of a current sweep;
- Figure 6B shows the corresponding voltage profiles; and
- Figure 7 shows the heating of the flow streams influence on the voltage response.

Method 10 (Figure 1) for boosting a redox flow battery with a membrane device follows a sequence of steps.

In the illustrated embodiment method 10 may start with step 12 of providing the redox flow battery with the membrane device. In the discharging stater, step 12 may be followed by step 14 of circulating a catholyte to the compartment with the cathode and step 16 of circulating an anolyte to the compartment with the anode. Step 14 and step 16 may be followed by step 18 of providing freshwater including an acid to the compartment delineated by the anion membrane and bipolar membrane, and step 20 of providing freshwater including a base to the compartment delineated by the cation membrane and bipolar membrane. Furthermore, step 18 and step 20 may be followed by step 22 of providing freshwater or salt water to the intermediate compartment. Preferably, step 22 includes providing freshwater in the discharging state.

Step 22 may be followed by step 24 of discharging the redox flow battery. Said step may also referred to as discharging the anolyte and catholyte.

Preferably, steps 14, 16, 18, 20, 22, and 24 are performed simultaneously in the discharging state.

In the charging state, step 12 may be followed by step 14 of circulating a catholyte to the compartment with the cathode and step 16 of circulating an anolyte to the compartment with the anode. Step 12 and step 14 may be followed by step 26 of providing freshwater to the compartment delineated by the anion membrane and bipolar membrane, and step 28 of providing freshwater to the compartment delineated by the cation membrane and bipolar membrane. Furthermore, step 26 and step 28 may be followed by step 22 of providing freshwater or salt water to the intermediate compartment. Preferably, step 22 includes providing salt water in the discharging state.

Step 22 may be followed by step 30 charging the redox flow battery.

Preferably, steps 14, 16, 22, 26, 28, 30 are performed simultaneously in the charging state.

Membrane device 40 (Figure 2) comprises triplet of membranes 41. Triplet of membranes 41 comprises an anion exchange membrane 42, bipolar membrane 44, and cation exchange membrane 46. Furthermore, membrane device 40 comprises compartments 48, 50, 52, 54, wherein compartment 48 comprises electrode 56, and compartment 54 comprises electrode 58. Furthermore, compartment 48 is at least partly delineated by anion exchange membrane 42, and compartment 54 is at least partly delineated by cation exchange membrane 46.

Compartments 50 and 52 are at least partly delineated by bipolar membrane 44 and anion exchange membrane 42 and cation exchange membrane 46 respectively.

Compartment 48 is configured for holding/circulating electrolyte 60, and compartment 54 is configured for holding/circulating electrolyte 62. Electrolyte 60 may be provided from tank 64, wherein tank 64 is operatively coupled with compartment 48. Electrolyte 62 may be provided from tank 66, wherein tank 66 is operatively coupled with compartment 54.

Compartment 50 is configured to be provided with stream 68, and compartment 52 is configured to be provided with stream 70. Stream 68 and stream 70 are removed from compartment 68 and compartment 52 *via* outlet 72 and outlet 74 respectively.

Membrane device 40 further comprises means 76 for applying an electrical potential difference between the cathode and the anode.

In the charging state electrode 56 is an anode, and in the discharging state electrode 56 is a cathode. Furthermore, in the charging state electrode 58 is a cathode, and in the discharging state electrode 58 is an anode.

In the charging state electrolyte 60 is catholyte, and in the discharging state electrolyte 60 is an anolyte. Furthermore, in the charging state electrolyte 62 is catholyte, and in the discharging state electrolyte 62 is an anolyte.

In the charging state stream 68 and 70 are freshwater, in the discharging state stream 68 is acidic, and stream 70 is basic.

Membrane device 78 (Figure 3) comprises two triplet of membranes 41. Triplet of membranes 41 comprises anion exchange membrane 42, bipolar membrane 44, and cation exchange membrane 46. Furthermore, membrane device 78 comprises compartments 48, 50, 52, 54, wherein compartment 48 comprises electrode 56, and compartment 54 comprises electrode 58. Furthermore, compartment 48 is at least partly delineated by a first anion exchange membrane 42, and compartment 54 is at least partly delineated by a first cation exchange membrane 46.

Compartments 50 and 52 are at least partly delineated by bipolar membrane 44 and anion exchange membrane 42 and cation exchange membrane 46 respectively.

Compartment 48 is configured for holding/circulating electrolyte 60, and compartment 54 is configured for holding/circulating electrolyte 62. Electrolyte 60 may be provided from tank 64, wherein tank 64 is operatively coupled with compartment 48. Electrolyte 62 may be provided from tank 66, wherein tank 66 is operatively coupled with compartment 54.

Compartment 50 is configured to be provided with stream 68, and compartment 52 is configured to be provided with stream 70. Stream 68 and stream 70 are removed from compartment 68 and compartment 52 *via* outlet 72 and outlet 74 respectively.

Membrane device 78 further comprises intermediate compartment 80, wherein intermediate compartment 80 is at least partly delineated by cation exchange membrane 46 of a first triplet of membranes and at least partly delineated by anion exchange membrane 42 of a second triplet of membranes. Intermediate compartment 80 is configured to be provided with stream 82. Stream 82 is removed from intermediate compartment 82 *via* outlet 84.

Membrane device 78 further comprises means 76 for applying an electrical potential difference between the cathode and the anode.

In the charging state electrode 56 is an anode, and in the discharging state electrode 56 is a cathode. Furthermore, in the charging state electrode 58 is a cathode, and in the discharging state electrode 58 is an anode.

In the charging state electrolyte 60 is catholyte, and in the discharging state electrolyte 60 is an anolyte. Furthermore, in the charging state electrolyte 62 is catholyte, and in the discharging state electrolyte 62 is an anolyte.

In the charging state stream 68 and 70 are freshwater and stream 82 is salt water, in the discharging state stream 68 is acidic, stream 70 is basic, and stream 82 is freshwater.

Membrane device 86 (Figure 4) comprises multiple triplet of membranes 92. Triplet of membranes 92 comprises anion exchange membranes 94, bipolar membranes 96, and cation exchange membranes 98. Furthermore, membrane device 86 comprises intermediate compartments 100, which are at least partly delineated by cation exchange membrane 98 of a first triplet of membranes and anion exchange membrane 94 of a second triplet of membranes.

Membrane device 86 further comprises compartment 91 which is partly delineated by anion exchange membrane 94 of the first triplet of membranes 92 and compartment 89 which is partly delineated by cation exchange membrane 98 of the last triplet of membranes 92. Compartment 89 comprises electrode 88 and compartment 91 comprises electrode 90.

It is noted that the intermediate compartments are formed between two triplet of membranes, except from the compartments comprising an electrode which are at least partly delineated a membrane of the triplet of membranes.

In an experiment the method according to the invention was compared with a conventional redox flow battery.

The method according to the invention was deployed on a conventional all iron redox flow battery, wherein a complexed $FeCl_2$ and $FeCl_3$ as an anolyte and a K_3FeCN_6 + Na_4FeCN_6 catholyte, being pumped through two compartments of the cell, on either side of an anion exchange membrane.

To facilitate this, two redox-active electrolytes were prepared:

Anolyte - Iron(II) and Iron (III) complexed with Triethanolamine (TEA) and dissolved in aqueous NaOH:

- $FeCl_2$ (0.2 mol L^{-1})
- $FeCl_3$ (0.2 mol L^{-1})
- TEA (2 mol L^{-1})
- NaOH (3 mol L^{-1})
- Total volume is 200 mL

Catholyte - Iron ferri and ferrocyanide dissolved in aqueous NaOH:

- $Na_4[FeCN]_6$ (0.2 mol L^{-1})
- $K_3[FeCN]_6$ (0.2 mol L^{-1})
- NaOH (3 mol L^{-1})
- Total volume is 200 mL

The electrolytes (anolyte and catholyte) described above, when recirculated in a cell with two compartments separated by a cation-exchange membrane (CEM), form the conventional all-iron redox flow battery. The booster part requires two additional chemicals:

Anolyte side:

- 200 mL of 1 mol L^{-1} HCl solution

Catholyte side:

- 200 mL of 1 mol L^{-1} NaOH solution

The above description concludes the chemical aspect of the non-boosted (conventional) and boosted redox flow batter.

The device of a conventional redox flow battery comprises two compartments separated by a cation exchange membrane. The channels were made out of gaskets with integrated netting. The active area of the channels was 10 cm × 2 cm, while the thickness of the channels was 488 μm. The cation exchange membrane was sourced from Fumatech and a titanium mesh was used as electrode. The electrolytes were pumped through the cell in a closed loop with the reservoir using a peristaltic pump sourced from Cole Palmer Inc.

The device for boosting a redox flow battery according to the invention, is shown in Figure 2. The following components are included in said device according to the invention:

- bipolar membrane - sourced from Fumatech
- 1 mol L^{-1} HCl and 1 mol L^{-1} NaOH compartment on either side of the bipolar membrane

To separate the HCl from the anolyte, an anion exchange membrane, sourced from Fumatech, was used. The spacers used as acid and base channels were identical in active area and thickness to the electrolyte spacers.

The conventional redox flow battery and the device for boosting a redox flow battery according to the invention were electrochemically tested under identical conditions. Before applying the current, the open circuit voltage of the cell was recorded. Following this, the following testing method was implemented:

- Electrolyte flow rate (400 mL min^{-1})
- Acid/Base flow rate (120 mL min^{-1})
- Applied current (12.5 A $m^{-2} \leq I_a \leq$ 100 A m^{-2})
- Charging and discharging time (600 seconds (10 minutes)). Total cycle time (20 minutes)
- Cell voltage was measured every 1 second.

The method above was implemented via a potensiostat, sourced from Autolab. The conventional device was tested first and then, fresh electrolytes were used for the device for boosting a redox flow battery.

The cell was also tested at an elevated temperature of 40 °C. This was facilitated by heating all the streams entering the cell, i.e. acid, base, salt and the two electrolytes over a hotplate. The temperature of the reservoirs was controlled by a temperature sensor dipped into the Fe-TE electrolyte and connected to the hotplate.

The testing method, as described in above, resulted in a current-voltage signal shown in Figure 5 for 12.5 A m^{-2} to 50 A m^{-2} and Figure 6 for 50 A m^{-2} to 100 A m^{-2}. Figure 5 shows in Figure 5A a profile of the current sweep applied to the two different cells for charging as well as discharging until 50 A m^{-2}, and Figure 5B shows the corresponding voltage profiles, as recorded for both. The electrolyte was refreshed for 50 A m^{-2}.

The lines at a cell voltage of 0 V to 1.45 V are directed to the conventional device and the cell voltage of 1.8 V to 2.3 V are directed to the device for boosting a redox flow battery according to the invention.

Figure 6A shows a profile of the current sweep applied to the two different cells for charging as well as discharging from 50 to 100 A m^{-2}, and Figure 6B shows the corresponding voltage profiles, as recorded for both devices.

From the cell voltage data in Figures 5 and 6, it is evident that the voltage, upon addition of a bipolar membrane, an increases by a step of $\approx$ 0.8V as the open circuit voltage is achieved. A measure of the maximum potential of a battery rises nearly 60%.

Heating the flow streams influences the voltage response of the cell as well, as shown in Figure 7. An increase in total cell voltage in the device for boosting a redox flow battery may operate at lower current density, for example a lower current density of about 60% compared to a conventional system and deliver the same power Thus, the gross power from the device for boosting a redox flow battery according to the invention increases. It is unexpected that for the same current density, the gross power output for the method and device for boosting a redox flow battery is significantly higher compared conventional methods and devices. This is a clear advantage over the conventional methods and devices.

The present invention is by no means limited to the above described preferred embodiments and/or experiments thereof. The rights sought are defined by the following claims within the scope of which many modifications can be envisaged.

CLAUSES

1. Method for boosting a redox flow battery with a membrane device, the method comprising the steps of:
   - providing the redox flow battery with the membrane device comprising:
     - n triplet of membranes, wherein n is an integer of 1 or more, and wherein each triplet of membranes consists of an anion exchange membrane, a bipolar membrane, and a cation exchange membrane; and
     - $3n + 1$ compartments, wherein one of the compartments comprises a cathode and wherein another of the compartments comprises an anode, and wherein the $3n + 1$ compartments are at least partly delineated by one of the triplet of membranes;
   - circulating a catholyte to the compartment with the cathode;
   - circulating an anolyte to the compartment with the anode; and
   - charging the redox flow battery by applying an electrical potential difference between the anode and the cathode and/or providing charged catholyte to the compartment with the cathode and anolyte to the compartment with the anode.

2. Method according to clause 1, further comprising the step of discharging the redox flow battery.

3. Method according to any one of the preceding clauses, further comprising the steps of providing freshwater including an acid to the compartment delineated by the anion membrane and bipolar membrane and providing freshwater including a base to the compartment delineated by the cation membrane and bipolar membrane.

4. Method according to clause 3, wherein the acid is one or more selected from the group of HCl, H_2SO_4, HI, and HF.

5. Method according to clause 3 or 4, wherein the base is sodium hydroxide and/or potassium hydroxide.

6. Method according to any one of the clauses 3 to 5, wherein the concentration of the acid and base are independently selected from the range of 0.5 mol L^{-1} to 3 mol L^{-1}, preferably 0.5 mol L^{-1} to 2 mol L^{-1}, more preferably 1 mol L^{-1} to 1.5 mol L^{-1}.

7. Method according to any one of the preceding clauses, further comprising the steps of providing freshwater to the compartment delineated by the anion membrane and bipolar membrane and providing freshwater to the compartment delineated by the cation membrane and bipolar membrane.

8. Method according to any one of the preceding clauses, wherein *n* is an integer of 2 or more, and wherein two triplets of membranes delineate an intermediate compartment.

9. Method according to clause 8, further comprising the step of providing freshwater or salt water to the intermediate compartment.

10. Method according to any one of the preceding clauses, wherein the catholyte comprises one or more selected from the group of $Na_4[FeCN]_6$, $K_3[FeCN]_6$, Br_2/Br^-, VO^{2+}.

11. Method according to any one of the preceding clauses, wherein the anolyte comprises one or more selected from the group of $FeCl_2$, $FeCl_3$, triethanolamine, V^{2+}/V^{3+}, H_2/H^+, Zn/Zn^{2+}.

12. Method according to any one of the preceding clauses, wherein the anolyte and/or catholyte comprises one or more bases independently selected from potassium hydroxide and sodium hydroxide.

13. Method according to clause 12, wherein the concentration of the base is in the range of 1 mol L^{-1} to 5 mol L^{-1}, preferably 2 mol L^{-1} to 4 mol L^{-1}, more preferably 2.5 mol L^{-1} to 3.5 mol L^{-1}.

14. Method according to any one of the preceding clauses, wherein the anolyte and/or catholyte are heated to a temperature in the range of 25 °C to 80 °C, preferably in the range of 35 °C to 60 °C, more preferably in the range of 35 °C to 50 °C.

15. Method according to any one of the preceding clauses, wherein circulating the anolyte and circulating the catholyte is independently performed with a flushing rate in the range of 50 L min^{-1} per cubic decimetre of the compartment to 500 L min^{-1} per cubic decimetre of the compartment the anolyte or catholyte is circulated in, preferably in the range of 100 L min^{-1} per cubic decimetre of the compartment to 450 L min^{-1} per cubic decimetre of the compartment the anolyte or catholyte is circulated in, more preferably in the range of 250 L min^{-1} per cubic decimetre of the compartment to 450 L min^{-1} per cubic decimetre of the compartment the anolyte or catholyte is circulated in.

16. Device for boosting a redox flow battery, configured to perform the method according to any one of the preceding clauses, comprising:

- n triplet of membranes, wherein n is an integer of 1 or more, and wherein each triplet of membranes consists of an anion exchange membrane, a bipolar membrane, and a cation exchange membrane;
- $3n + 1$ compartments, wherein one of the compartments comprises a cathode and wherein another of the compartments comprises an anode, and wherein the $3n + 1$ compartments are at least partly delineated by one of the triplet of membranes; and
- means for applying an electrical potential difference between the cathode and the anode,

wherein the compartment comprising the anode is configured to, during use, be provided with an anolyte and the compartment comprising the cathode is configured to, during use, be provided with a catholyte.

17. Device according to clause 16, wherein n is an integer of 2 or more, and wherein two triplets of membranes delineate an intermediate compartment.

18. Device according to clause 17, wherein the intermediate compartment is configured to, during use in a discharge state, be provided with freshwater, and configured to, during use in a charge state, be provided with salt water.

19. Membrane stack for boosting a redox flow battery, the stack comprising a number of cells which are configured to perform the method according to any one of the clauses 1 to 15.

20. System for boosting a redox flow battery, comprising:

- a device according to any one of the clauses 16 to 18;
- a first pump configured for circulating an anolyte;
- a second pump configured for circulating a catholyte;
- an anolyte storage which is operatively coupled with an anolyte buffer of the device and the first pump; and
- a catholyte storage which is operatively coupled with a catholyte buffer of the device and the second pump.

21. System according to clause 20, wherein the means to provide an electrical potential difference is a windmill, a solar panel, and/or tidal power station.

CONCLUSIES

1. Werkwijze voor het stimuleren van een redox flow batterij met een membraaninrichting, de werkwijze omvattende de stappen van:
   - het verschaffen van de redox flow batterij met de membraaninrichting omvattende:
     - *n* triplet van membranen, waarbij *n* een integer is van 1 of meer, en waarbij iedere triplet van membranen bestaat uit een anion uitwisselingsmembraan, een bipolairmembraan, en een kation uitwisselingsmembraan; en
     - $3n + 1$ compartimenten, waarbij één van de compartimenten een kathode omvat en waarbij een ander van de compartimenten een anode omvat, en waarbij de $3n + 1$ compartimenten ten minste ten dele worden afgebakend door één van de triplet van membranen;
   - het circuleren van een katholiet naar het compartiment met de kathode;
   - het circuleren van een anoliet naar het compartiment met de anode; en
   - het laden van de redox flow batterij door het toepassen van een elektrisch potentiaalverschil tussen de anode en de kathode en/or het verschaffen van geladen katholiet aan het compartiment met de kathode en anoliet aan het compartiment met de anode.

2. Werkwijze volgens conclusie 1, verder omvattende de stap van het ontladen van de redox flow batterij.

3. Werkwijze volgens één der voorgaande conclusies, verder omvattende de stappen van het verschaffen van zoetwater bevattende een zuur aan het compartiment afgebakend door het anionmembraan en bipolairmembraan en het verschaffen van zoetwater bevattende een base aan het compartiment afgebakend door het kationmembraan en bipolairmembraan.

4. Werkwijze volgens conclusie 3, waarbij het zuur één of meer is geselecteerd uit de groep van HCl, H_2SO_4, HI, en HF.

5. Werkwijze volgens conclusie 3 of 4, waarbij de base natrium hydroxide en/of kaliumhydroxide is.

6. Werkwijze volgens één der conclusies 3 tot en met 5, waarbij de concentratie van het zuur en base onafhankelijk zijn geselecteerd uit het bereik van 0,5 mol L^{-1} tot en met 3 mol L^{-1}, bij voorkeur 0,5 mol L^{-1} tot en met 2 mol L^{-1}, bij meer voorkeur 1 mol L^{-1} tot en met 1,5 mol L^{-1}.

7. Werkwijze volgens één der voorgaande conclusies, verder omvattende de stappen van het verschaffen van zoetwater aan het compartiment afgebakend door het anionmembraan en bipolairmembraan en het verschaffen van zoetwater aan het compartiment afgebakend door het kationmembraan en bipolairmembraan.

8. Werkwijze volgens één der voorgaande conclusies, waarbij *n* een integer is van 2 or meer, en waarbij de triplets van membranen een tussenliggend compartiment afbakenen.

9. Werkwijze volgens conclusie 8, verder omvattende de stap van het verschaffen van zoetwater of zoutwater aan het tussenliggend compartiment.

10. Werkwijze volgens één der voorgaande conclusies, waarbij het katholiet één of meer geselecteerd uit de groep van $Na_4[FeCN]_6$, $K_3[FeCN]_6$, Br_2/Br^-, VO^{2+} omvat.

11. Werkwijze volgens één der voorgaande conclusies, waarbij het anoliet één of meer geselecteerd uit de groep van $FeCl_2$, $FeCl_3$, triethanolamine, V^{2+}/V^{3+}, H_2/H^+, Zn/Zn^{2+} omvat.

12. Werkwijze volgens één der voorgaande conclusies, waarbij het anoliet en/of katholiet één of meer basen omvatten die onafhankelijk zijn geselecteerd uit de groep van kalium hydroxide en natrium hydroxide.

13. Werkwijze volgens conclusie 12, waarbij de concentratie van de base in het bereik ligt van 1 mol L^{-1} tot en met mol L^{-1}, bij voorkeur 2 mol L^{-1} tot en met 4 mol L^{-1}, bij meer voorkeur 2,5 mol L^{-1} tot en met 3,5 mol L^{-1}.

14. Werkwijze volgens één der voorgaande conclusies, waarbij het anoliet en/of katholiet wordt verwarmd tot een temperatuur in het bereik van 25 °C tot en met 80 °C, bij voorkeur in het bereik van 35 °C tot en met 60 °C, bij meer voorkeur in het bereik van 35 °C tot en met 50 °C.

15. Werkwijze volgens één der voorgaande conclusies, waarbij het circuleren van het anoliet en het circuleren van het katholiet onafhankelijk wordt uitgevoerd met een spoelsnelheid in het bereik van 50 L min^{-1} per kubieke decimeter van het compartiment tot en met 500 L min^{-1} per kubieke decimeter van het compartiment waar het anoliet of katholiet in wordt gecirculeerd, bij voorkeur in het bereik van 100 L min^{-1} per kubieke decimeter van het compartiment tot en met 450 L min^{-1} per kubieke decimeter van het compartiment waar het anoliet of katholiet in wordt

gecirculeerd, bij meer voorkeur in het bereik van 250 L min^{-1} per kubieke decimeter van het compartiment tot en met 450 L min^{-1} per kubieke decimeter van het compartiment waar het anoliet of katholiet in wordt gecirculeerd.

16. Inrichting voor het stimuleren van een redox flow batterij, ingericht voor het uitvoeren van de werkwijze volgens één der voorgaande conclusies, omvattende:

- *n* triplet van membranen, waarbij *n* een integer is van 1 of meer, en waarbij iedere triplet van membranen bestaat uit een anion uitwisselingsmembraan, een bipolairmembraan, en een kation uitwisselingsmembraan;
- $3n + 1$ compartimenten, waarbij één van de compartimenten een kathode omvat en waarbij een ander van de compartimenten een anode omvat, en waarbij de $3n + 1$ compartimenten ten minste ten dele worden afgebakend door één van de triplet van membranen; en
- middelen voor het toepassen van een elektrisch potentiaalverschil tussen de kathode en de anode,

waarbij het compartiment omvattende de anode is ingericht om, tijdens gebruik, te worden verschaft van een anoliet en het compartiment omvattende de kathode is ingericht om, tijdens gebruik, te worden verschaft van een katholiet.

17. Inrichting volgens conclusie 16, waarbij *n* een integer is van 2 of meer, en waarbij twee triplets van membranen een tussenliggend compartiment afbakenen.

18. Inrichting volgens conclusie 17, waarbij het tussenliggende compartiment is ingericht om, tijdens gebruik in een ontladingstoestand, te worden verschaft van zoetwater, en ingericht om, tijdens gebruik in een ladingstoestand, te worden verschaft met zoutwater.

19. Membraanstapel voor het stimuleren van een redox flow batterij, de stapel omvattende een aantal van cellen die zijn ingericht om de werkwijze volgens één der conclusies 1 tot en met 15 uit te voeren.

20. Systeem voor het stimuleren van een redox flow batterij, omvattende:

- een inrichting volgens één der conclusies 16 tot en met 18;
- een eerste pump die is ingericht voor het circuleren van een anoliet;
- een tweede pump die is ingericht voor het circuleren van een katholiet;
- een anolietopslag die werkzaam is gekoppeld met een anolytebuffer van de inrichting en de eerste pomp; en

- een katholietopslag die werkzaam is gekoppeld met een katholietbuffer van de inrichting en de tweede pomp.

21. Systeem volgens conclusie 20, waarbij de middelen voor het verschaffen van een elektrisch potentiaalverschil een windmolen, een zonnepaneel, en/of een getijdecentrale is.

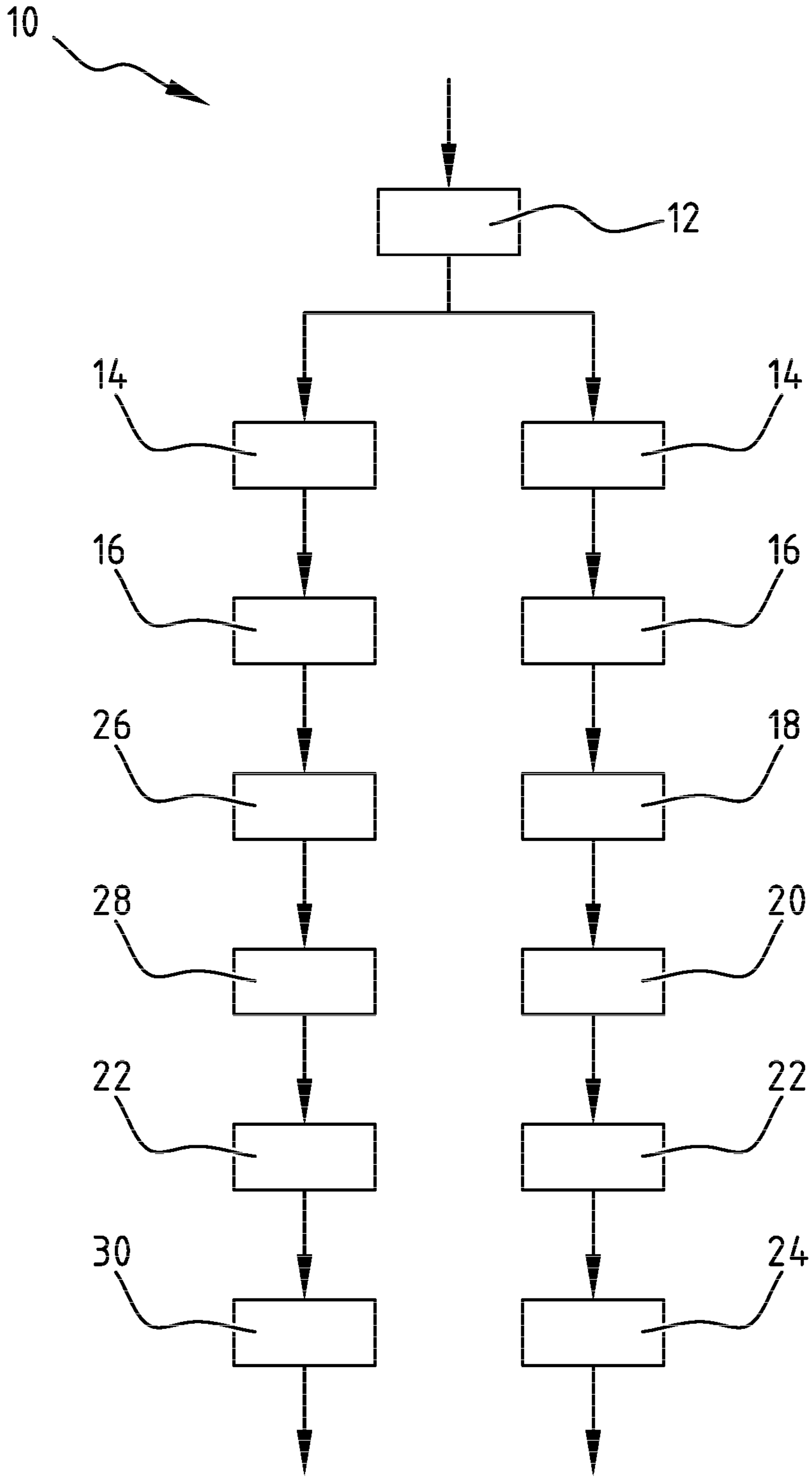


FIG. 1

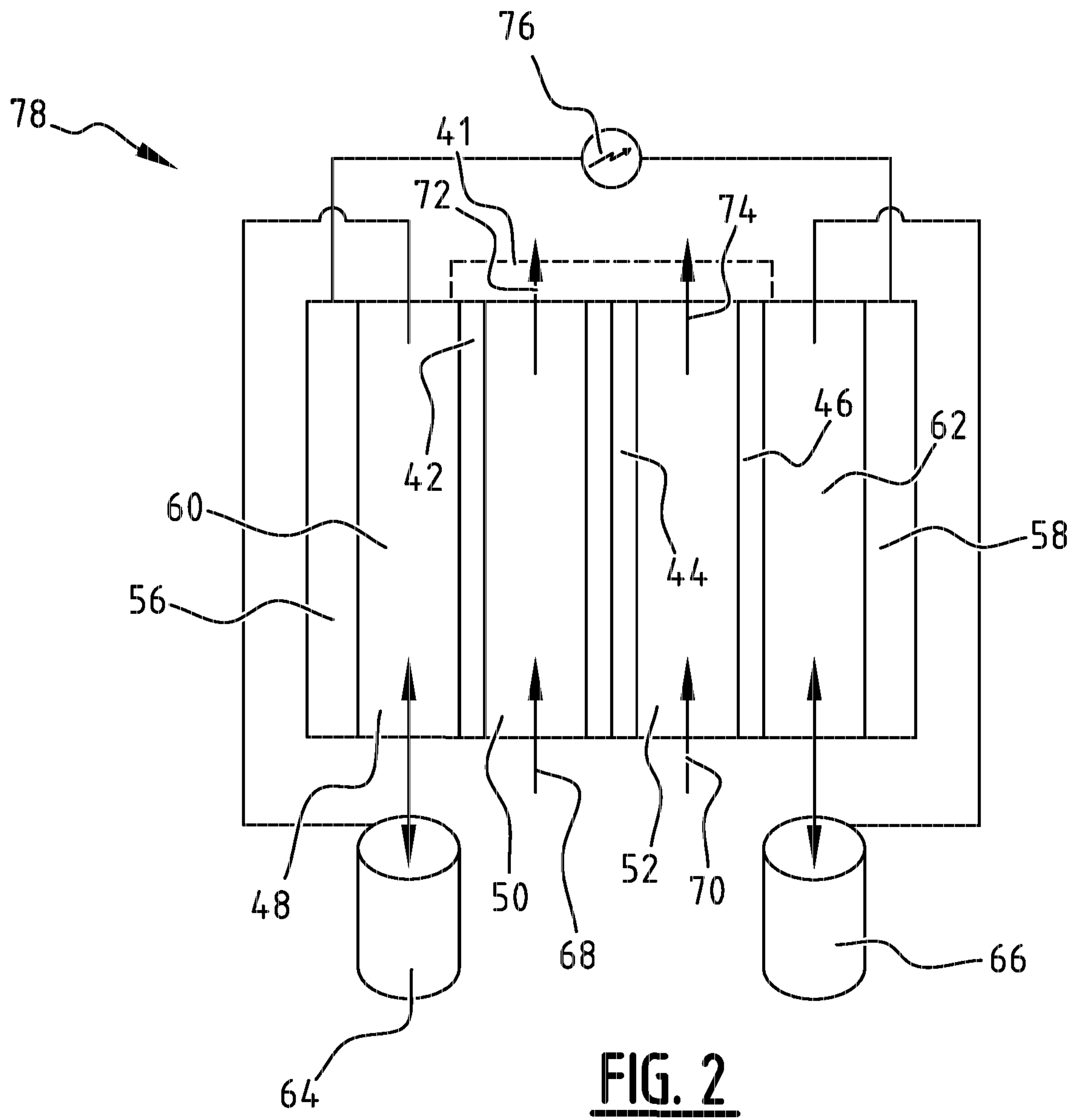
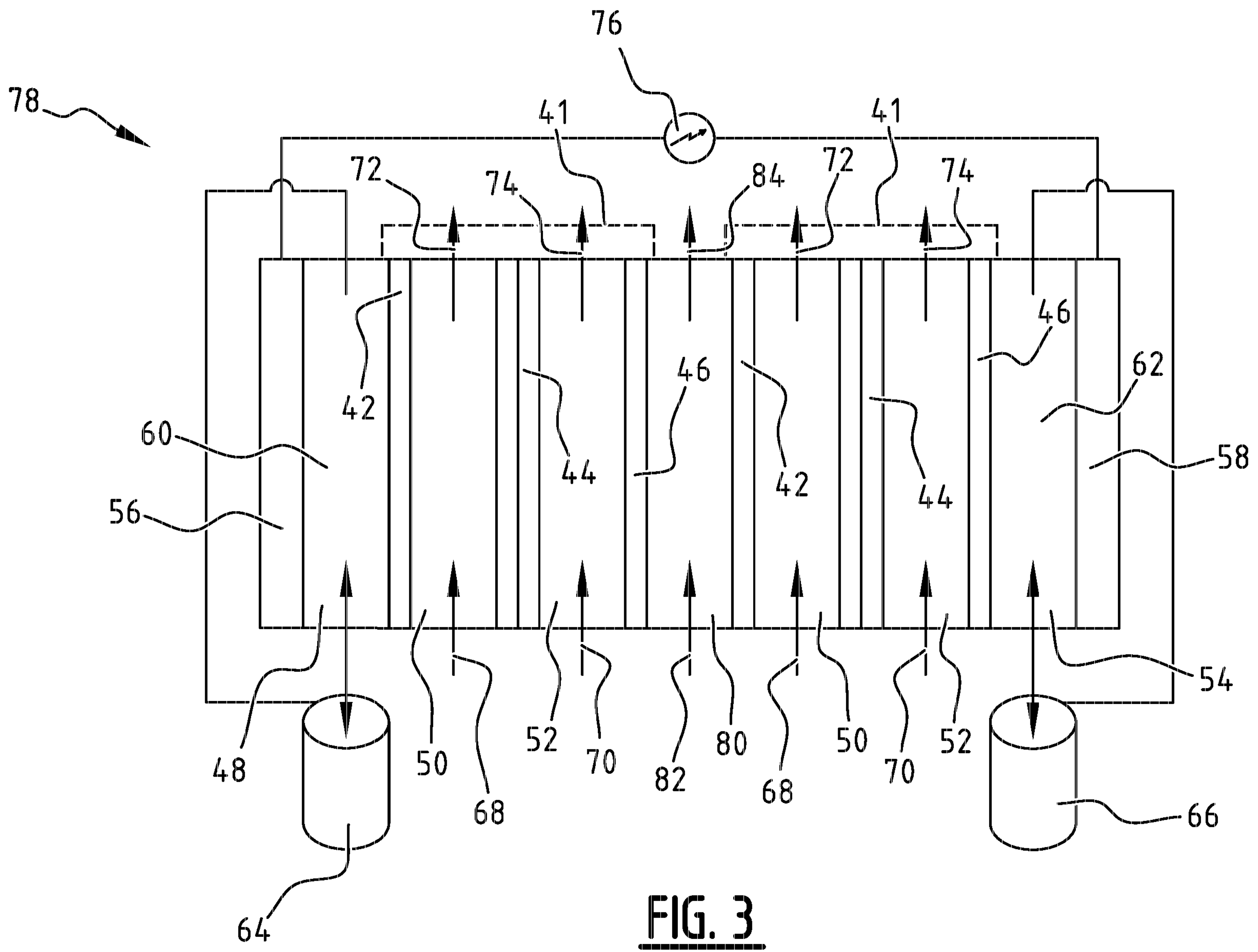


FIG. 2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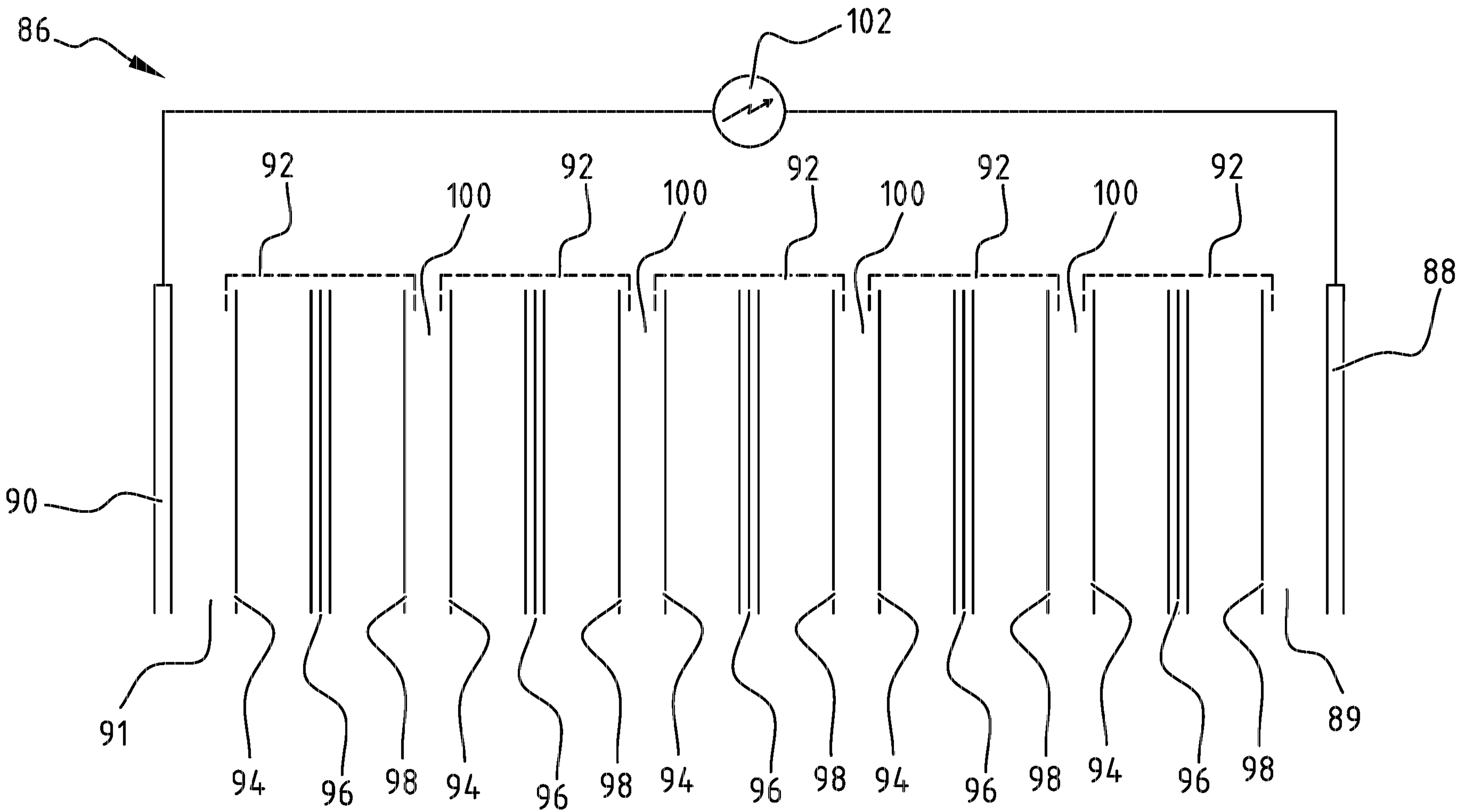
66
64

FIG. 3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92
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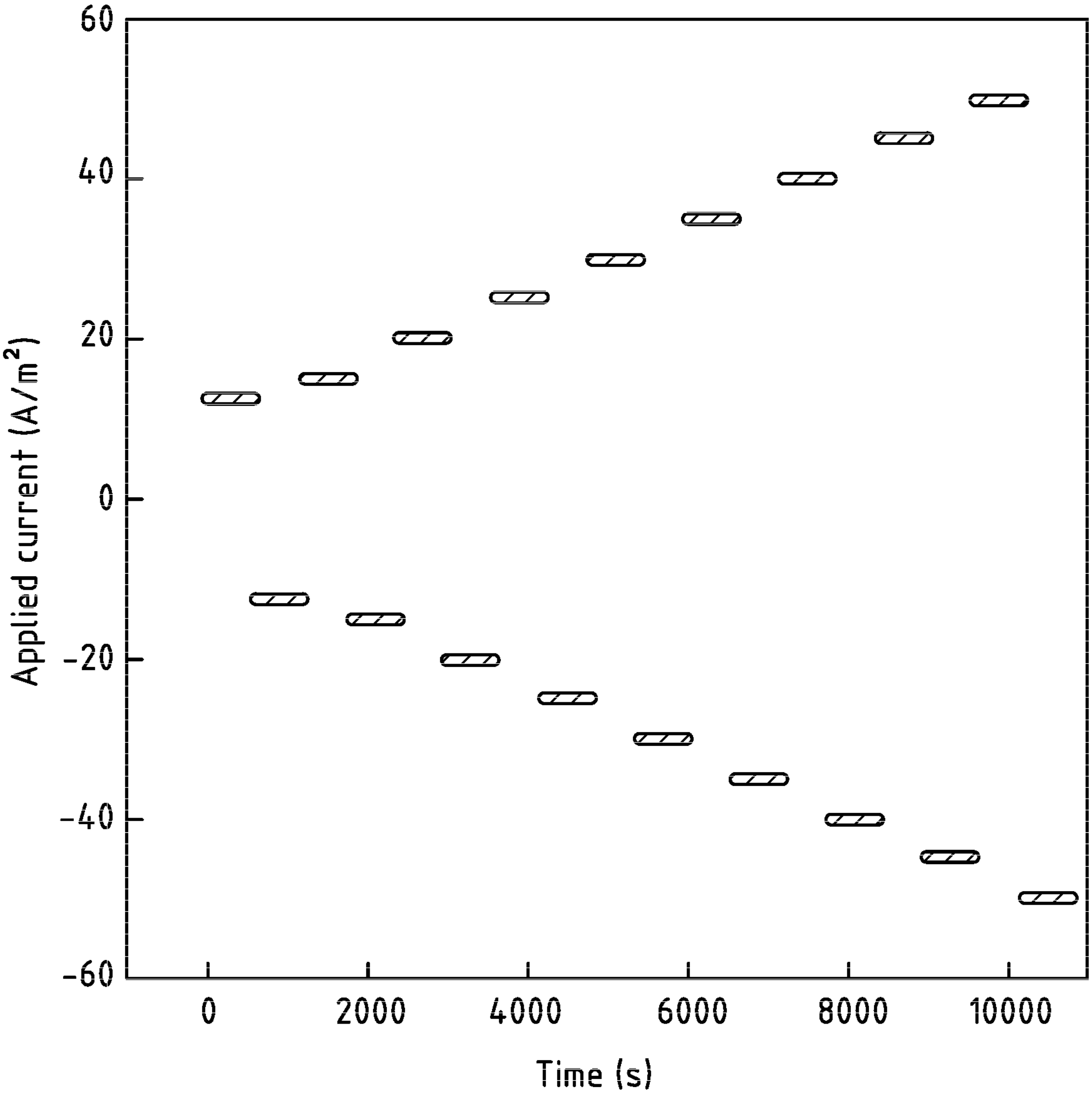
98
89

FIG. 4



60
40
20
0
-20
-40
-60
Applied current (A/m²)
0
2000
4000
6000
8000
10000
Time (s)

FIG. 5A

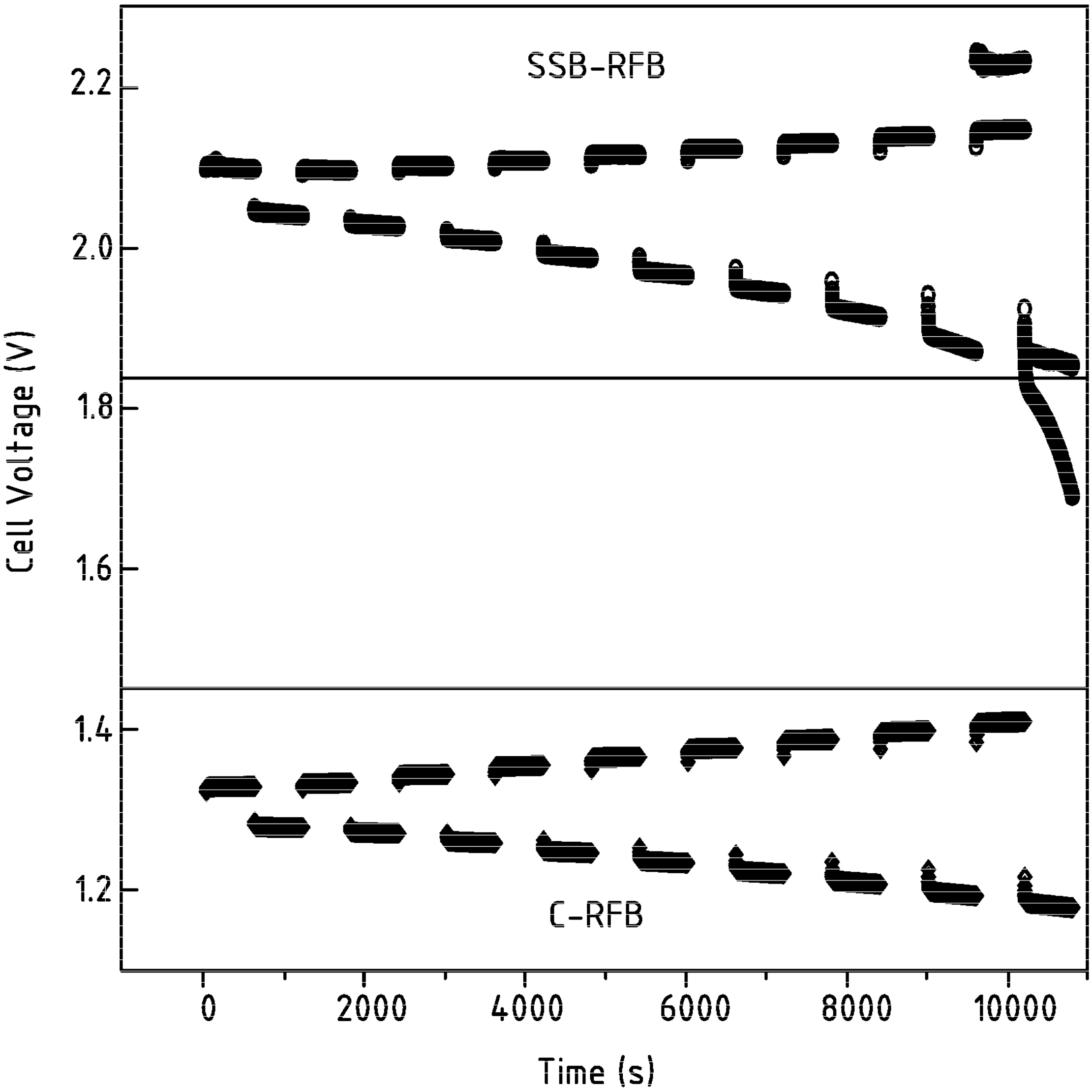
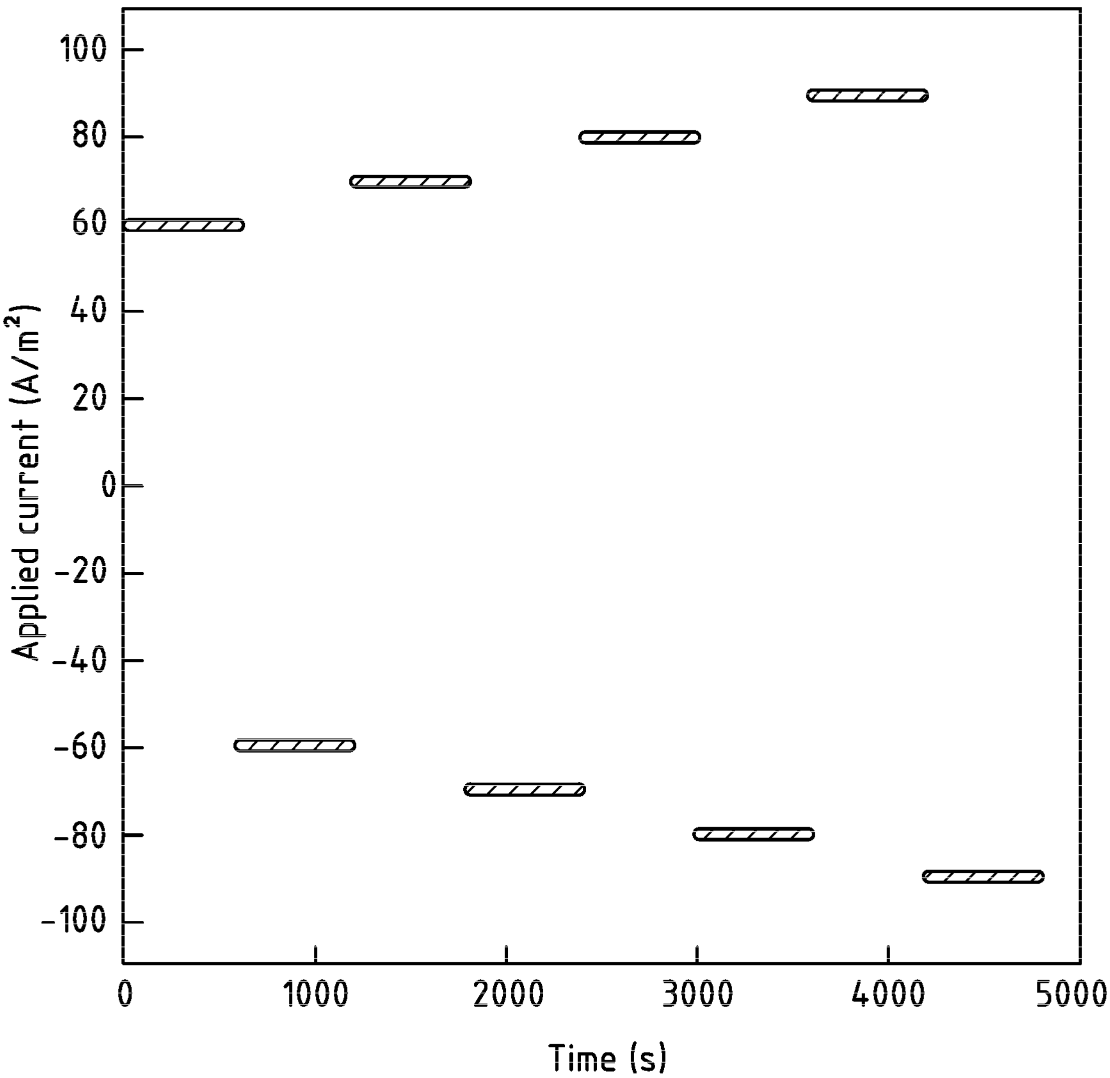

SSB-RFB
C-RFB
Cell Voltage (V)
2.2
2.0
1.8
1.6
1.4
1.2
0
2000
4000
6000
8000
10000
Time (s)

FIG. 5B



100
80
60
40
20
0
-20
-40
-60
-80
-100
Applied current (A/m²)
0
1000
2000
3000
4000
5000
Time (s)

FIG. 6A

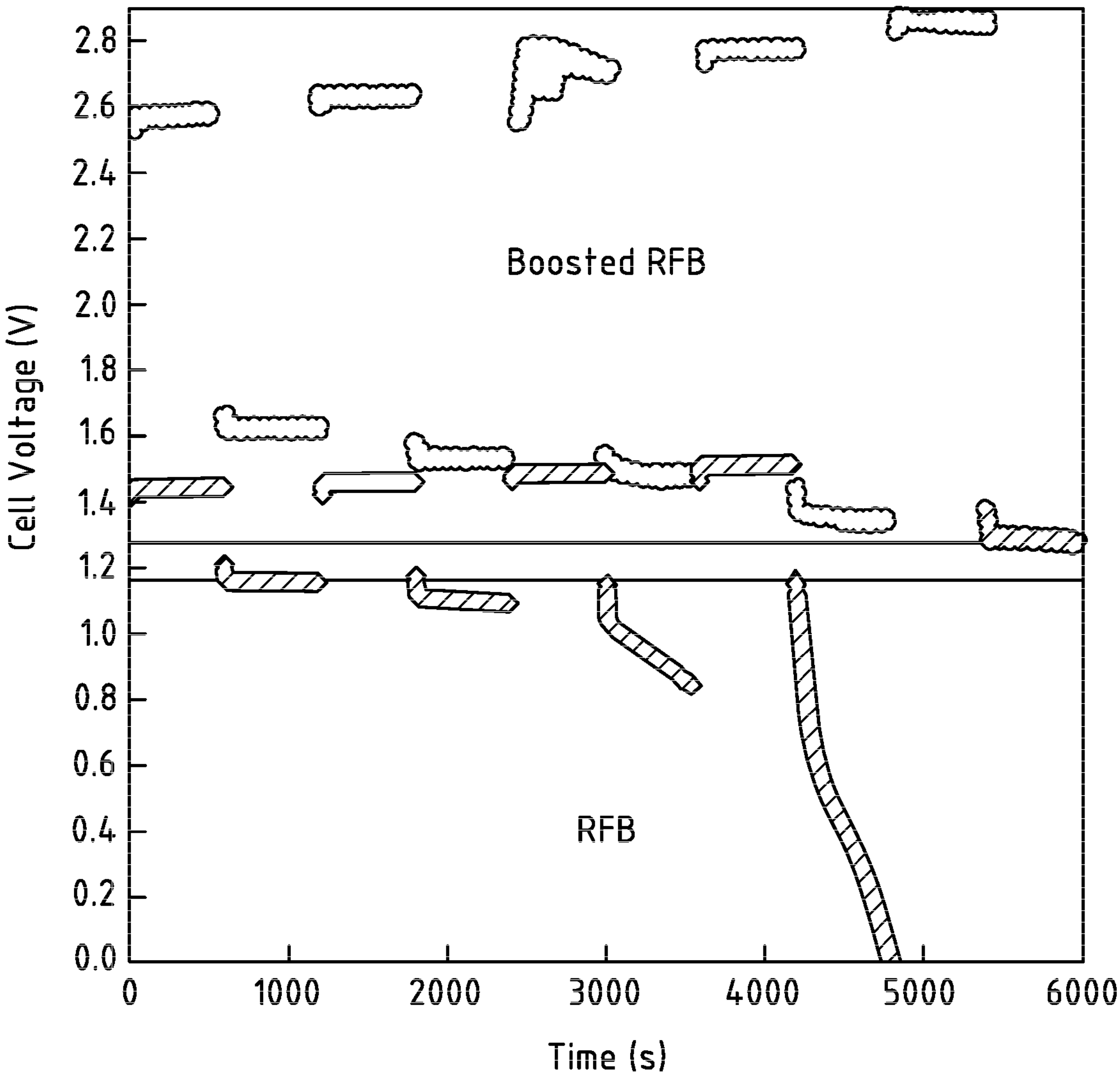

Cell Voltage (V)
2.8
2.6
2.4
2.2
2.0
1.8
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
Boosted RFB
RFB
0
1000
2000
3000
4000
5000
6000
Time (s)

FIG. 6B

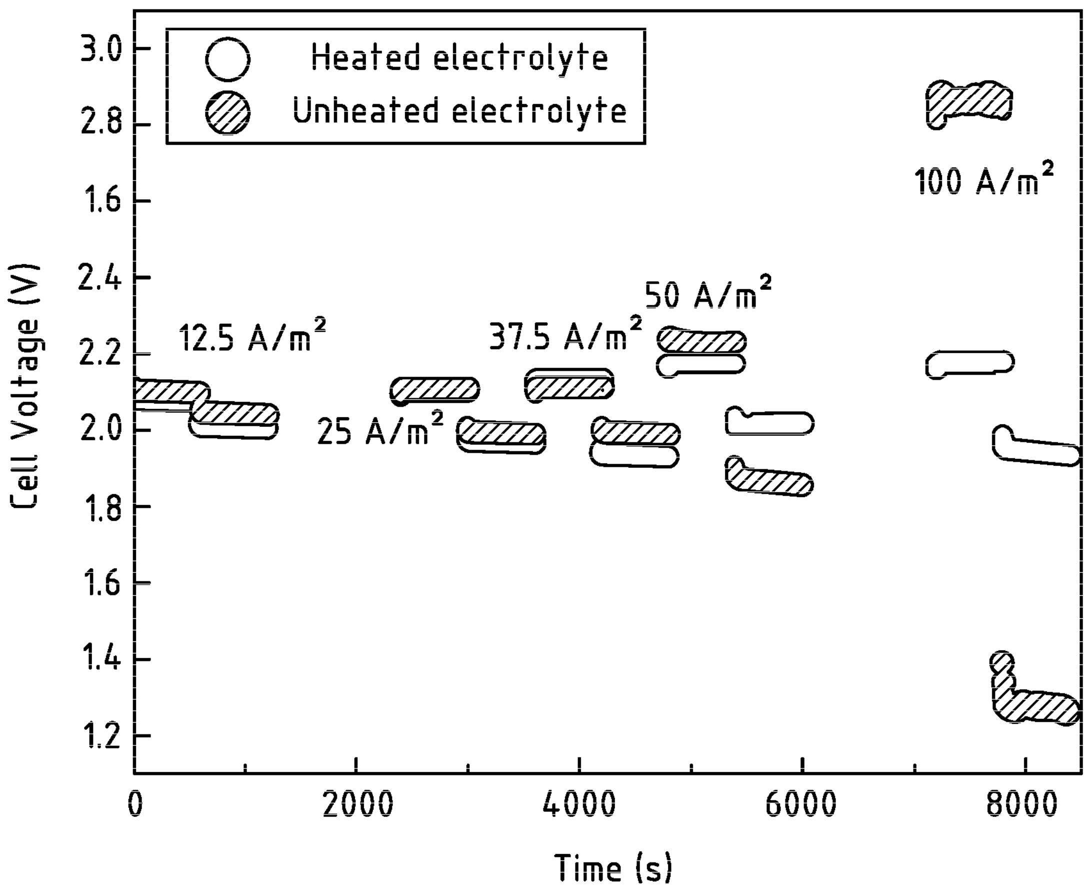

Heated electrolyte
Unheated electrolyte
100 A/m²
50 A/m²
37.5 A/m²
12.5 A/m²
25 A/m²
Cell Voltage (V)
Time (s)
3.0
2.8
1.6
2.4
2.2
2.0
1.8
1.6
1.4
1.2
0
2000
4000
6000
8000

FIG. 7

SAMENWERKINGSVERDRAG (PCT)

RAPPORT BETREFFENDE NIEUWHEIDSONDERZOEK VAN INTERNATIONAAL TYPE

IDENTIFICATIE VAN DE NATIONALE AANVRAGE	**KENMERK VAN DE AANVRAGER OF VAN DE GEMACHTIGDE**
Nederlands aanvraag nr. **2034448**	Indieningsdatum **28-03-2023**
	Ingeroepen voorrangsdatum
Aanvrager (Naam) **Aquabattery B.V.**	
Datum van het verzoek voor een onderzoek van internationaal type **15-04-2023**	Door de Instantie voor Internationaal Onderzoek aan het verzoek voor een onderzoek van internationaal type toegekend nr. **SN83655**

I. CLASSIFICATIE VAN HET ONDERWERP (bij toepassing van verschillende classificaties, alle classificatiesymbolen opgeven)	
Volgens de internationale classificatie (IPC) **Zie onderzoeksrapport**	
II. ONDERZOCHTE GEBIEDEN VAN DE TECHNIEK	
	Onderzochte minimumdocumentatie
Classificatiesysteem	Classificatiesymbolen
IPC	**Zie onderzoeksrapport**
Onderzochte andere documentatie dan de minimum documentatie, voor zover dergelijke documenten in de onderzochte gebieden zijn opgenomen	

III.		**GEEN ONDERZOEK MOGELIJK VOOR BEPAALDE CONCLUSIES**	(opmerkingen op aanvullingsblad)
IV.		**GEBREK AAN EENHEID VAN UITVINDING**	(opmerkingen op aanvullingsblad)

Form PCT/ISA 201 A (11/2000)

ONDERZOEKSRAPPORT BETREFFENDE HET RESULTAAT VAN HET ONDERZOEK NAAR DE STAND VAN DE TECHNIEK VAN HET INTERNATIONALE TYPE

Nummer van het verzoek om een onderzoek naar de stand van de techniek
NL 2034448

A. CLASSIFICATIE VAN HET ONDERWERP
INV. H01M8/18 H01M10/38 H01M8/22 B01D61/44 C02F1/461
ADD. H02J15/00

Volgens de Internationale Classificatie van octrooien (IPC) of zowel volgens de nationale classificatie als volgens de IPC.

B. ONDERZOCHTE GEBIEDEN VAN DE TECHNIEK

Onderzochte miminum documentatie (classificatie gevolgd door classificatiesymbolen)
H01M H02J B01D C02F

Onderzochte andere documentatie dan de mimimum documentatie, voor dergelijke documenten, voor zover dergelijke documenten in de onderzochte gebieden zijn opgenomen

Tijdens het onderzoek geraadpleegde elektronische gegevensbestanden (naam van de gegevensbestanden en, waar uitvoerbaar, gebruikte trefwoorden)

EPO-Internal, WPI Data

C. VAN BELANG GEACHTE DOCUMENTEN

Categorie °	Geciteerde documenten, eventueel met aanduiding van speciaal van belang zijnde passages	Van belang voor conclusie nr.
X	PÄRNAMÄE RAGNE ET AL: "The Acid-Base Flow Battery: Sustainable Energy Storage via Reversible Water Dissociation with Bipolar Membranes", MEMBRANES, deel 10, nr. 12, 10 december 2020 (2020-12-10), bladzijde 409, XP093007666, DOI: 10.3390/membranes10120409 * bladzijde 3, alinea 2 - bladzijde 8, alinea 5; figuur 2; tabellen 1,2 * * bladzijde 16, alinea 1 * -----	1-13, 15-21
X	WO 98/09002 A1 (SACHEM INC [US]) 5 maart 1998 (1998-03-05) * figuren 1-6; voorbeelden 1-5 * -----	1,2,8, 10,11, 14,16, 17,19,20

-/--

[x] Verdere documenten worden vermeld in het vervolg van vak C.

[x] Leden van dezelfde octrooifamilie zijn vermeld in een bijlage

° Speciale categorieën van aangehaalde documenten

"A" niet tot de categorie X of Y behorende literatuur die de stand van de techniek beschrijft

"D" in de octrooiaanvrage vermeld

"E" eerdere octrooi(aanvrage), gepubliceerd op of na de indieningsdatum, waarin dezelfde uitvinding wordt beschreven

"L" om andere redenen vermelde literatuur

"O" niet-schriftelijke stand van de techniek

"P" tussen de voorrangsdatum en de indieningsdatum gepubliceerde literatuur

"T" na de indieningsdatum of de voorrangsdatum gepubliceerde literatuur die niet bezwarend is voor de octrooiaanvrage, maar wordt vermeld ter verheldering van de theorie of het principe dat ten grondslag ligt aan de uitvinding

"X" de conclusie wordt als niet nieuw of niet inventief beschouwd ten opzichte van deze literatuur

"Y" de conclusie wordt als niet inventief beschouwd ten opzichte van de combinatie van deze literatuur met andere geciteerde literatuur van dezelfde categorie, waarbij de combinatie voor de vakman voor de hand liggend wordt geacht

"&" lid van dezelfde octrooifamilie of overeenkomstige octrooipublicatie

Datum waarop het onderzoek naar de stand van de techniek van internationaal type werd voltooid	Verzenddatum van het rapport van het onderzoek naar de stand van de techniek van internationaal type
25 oktober 2023	

Naam en adres van de instantie	De bevoegde ambtenaar
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016	Barenbrug-van Druten

1

Formulier PCT/ISA/201 (tweede blad) (Januari 2004)

ONDERZOEKSRAPPORT BETREFFENDE HET RESULTAAT VAN HET ONDERZOEK NAAR DE STAND VAN DE TECHNIEK VAN HET INTERNATIONALE TYPE

Nummer van het verzoek om een onderzoek naar de stand van de techniek
NL 2034448

C.(Vervolg). VAN BELANG GEACHTE DOCUMENTEN

Categorie °	Geciteerde documenten, eventueel met aanduiding van speciaal van belang zijnde passages	Van belang voor conclusie nr.
X	WO 2022/226323 A1 (HARVARD COLLEGE [US]) 27 oktober 2022 (2022-10-27) * bladzijde 7, regel 7 - bladzijde 12, regel 25; figuren 1-5 *	1-12, 16-21

1

Formulier PCT/ISA/201 (vervolg tweede blad) (Januari 2004)

ONDERZOEKSRAPPORT BETREFFENDE HET RESULTAAT VAN HET ONDERZOEK NAAR DE STAND VAN DE TECHNIEK VAN HET INTERNATIONALE TYPE

Informatie over leden van dezelfde octrooifamilie

Nummer van het verzoek om een onderzoek naar de stand van de techniek

NL 2034448

In het rapport genoemd octrooigeschrift		Datum van publicatie	Overeenkomend(e) geschrift(en)			Datum van publicatie
WO 9809002	A1	05-03-1998	AT	E350139	T1	15-01-2007
			AU	4237297	A	19-03-1998
			CA	2264086	A1	05-03-1998
			CN	1234080	A	03-11-1999
			DE	69737203	T2	18-10-2007
			EP	0944747	A1	29-09-1999
			HK	1023374	A1	08-09-2000
			JP	4320055	B2	26-08-2009
			JP	2000517379	A	26-12-2000
			KR	20000035915	A	26-06-2000
			MY	115681	A	30-08-2003
			TW	411368	B	11-11-2000
			US	5833832	A	10-11-1998
			WO	9809002	A1	05-03-1998
WO 2022226323	A1	27-10-2022	GEEN			

WRITTEN OPINION

File No.	Filing date *(day/month/year)*	Priority date *(day/month/year)*	Application No.
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Applicant
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This opinion contains indications relating to the following items:

- ☒ Box No. I — Basis of the opinion
- ☐ Box No. II — Priority
- ☐ Box No. III — Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- ☐ Box No. IV — Lack of unity of invention
- ☒ Box No. V — Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- ☐ Box No. VI — Certain documents cited
- ☒ Box No. VII — Certain defects in the application
- ☒ Box No. VIII — Certain observations on the application

	Examiner
	Barenbrug-van Druten

Application number

WRITTEN OPINION

NL2034448

Box No. I Basis of this opinion

1. This opinion has been established on the basis of the latest set of claims filed before the start of the search.
2. With regard to any **nucleotide and/or amino acid sequence** disclosed in the application, this opinion has been established on the basis of a sequence listing:

   a. ☐ forming part of the application as filed.

   b. ☐ furnished subsequent to the filing date for the purposes of search,

   ☐ accompanied by a statement to the effect that the sequence listing does not go beyond the disclosure in the application as filed.

3. ☐ With regard to any nucleotide and/or amino acid sequence disclosed in the application, this opinion has been established to the extent that a meaningful opinion could be formed without a WIPO Standard ST.26 compliant sequence listing.
4. Additional comments:

Box No. V Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty	Yes:	Claims	13, 15
	No:	Claims	1-12, 14, 16-21
Inventive step	Yes:	Claims	
	No:	Claims	1-21
Industrial applicability	Yes:	Claims	1-21
	No:	Claims	

2. Citations and explanations

   see separate sheet

Box No. VII Certain defects in the application

see separate sheet

Box No. VIII Certain observations on the application

see separate sheet

1 **<u>Re Item V</u>**

Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1.1 Reference is made to the following documents:

D1	PÄRNAMÄE RAGNE ET AL: "The Acid-Base Flow Battery: Sustainable Energy Storage via Reversible Water Dissociation with Bipolar Membranes", MEMBRANES, deel 10, nr. 12, 10 december 2020 (2020-12-10), bladzijde 409, XP093007666, DOI: 10.3390/membranes10120409
D2	WO 98/09002 A1 (SACHEM INC [US]) 5 maart 1998 (1998-03-05)
D3	WO 2022/226323 A1 (HARVARD COLLEGE [US]) 27 oktober 2022 (2022-10-27)

1.2 The present application does not meet the criteria of patentability, because the subject-matter of claim 1 is not new.

D1 discloses (see page 3, paragraph 2 - page 8, paragraph 5, page 16, first paragraph, figure 2 and tables 1 and 2):

Werkwijze voor het stimuleren van een redox flow batterij met een membraaninrichting, de werkwijze omvattende de stappen van:
- het verschaffen van de redox flow batterij met de membraaninrichting omvattende:
- n triplet van membranen, waarbij n een integer is van 1 of meer, en waarbij iedere triplet van membranen bestaat uit een anion uitwisselingsmembraan, een bipolairmembraan, en een kation uitwisselingsmembraan; en
- 3n + 1 compartimenten, waarbij één van de compartimenten een kathode omvat en waarbij een ander van de compartimenten een anode omvat, en waarbij de 3n + 1 compartimenten ten minste ten dele worden afgebakend door één van de triplet van membranen;
- het circuleren van een katholiet naar het compartiment met de kathode;
- het circuleren van een anoliet naar het compartiment met de anode; en
- het laden van de redox flow batterij door het toepassen van een elektrisch potentiaalverschil tussen de anode en de kathode en/or het verschaffen van geladen katholiet aan het compartiment met de kathode en anoliet aan het compartiment met de anode.

The corresponding "inrichting" of claim 16 and "membraanstapel" of claim 19 are further disclosed in D1 as are the additional features of claims 2-5, 7-9, 12, 17-21 (see page 3, paragraph 2 - page 8, paragraph 5, page 16, first paragraph, figure 2 and tables 1 and 2).

Starting from D1, the additional features of claims 6, 10, 11, 13 and 15 relate to simple design details that are obvious to the skilled person and the subject-matter of these claims thus does not involve an inventive step.

1.3 The subject-matter of claims 1, 2, 8, 10, 11, 14, 16, 17, 19, 20 is further known from document D2 (see figures 1-6 and examples 1-5) and the subject-matter of claims 1-12, 16-21 is known from document D3 (see page 7, line 7 - page 12, line 25 and figures 1-5).

2 **Re Item VII**

Certain defects in the application

2.1 Independent claim 1 is not in the two-part form, which in the present case would be appropriate, with those features known in combination from the prior art being placed in the preamble and the remaining features being included in the characterising part.

2.2 The features of the claims are not provided with reference signs placed in parentheses.

2.3 The relevant background art disclosed in D1-D3 is not mentioned in the description, nor are these documents identified therein.

3 **Re Item VIII**

Certain observations on the application

3.1 Claims 1, 3, 7, 9, 18-20 are not clear for the following reasons.

3.2 The terms zoetwater and zoutwater used in claims 3, 7, 9 and 18 are unclear and leave the reader in doubt as to the meaning of the technical features to which they refer, thereby rendering the definition of the subject-matter of said claims unclear. The reason being that no salt concentration range is given within which the water is considered to be "zoet" or "salt". From the description page 4, lines 8 and 9 it is only clear that "zoetwater" is considered to be "naturally occurring water that is not salty, and is suitable for consumption if cleaned or processed" without defining what is considered to be "not salty" leading to doubt as to the scope of the terms "zoetwater" and "zoutwater".

3.3 Claim 19 does not meet the requirements of clarity in that the matter for which protection is sought is not clearly defined. The functional statement "ingericht om de werkwijze volgens één der conclusies 1 tot en met 15 uit te voeren" does not enable the skilled person to determine which technical features are necessary to perform the stated function. A membraanstapel that would be able to perform this function would appear to be identical to the "inrichting" of claim 16, and there is thus overlapping scope between the two independent product claims.

3.4 Claim 1, line 16 "en/or" is clearly a typographical error that should be replaced by "en/of". In claim 20 two instances of "pump" are clearly intended to be "pomp".

3.5 The subject-matter described on page 3, lines 13 and 15 does not fall within the scope of the claims as the "preferred" features are features on the independent claim 1 and are thus features of the invention. This inconsistency between the claims and the description leads to doubt concerning the matter for which protection is sought, thereby rendering the claims unclear.