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ELECTROCHEMICAL UTILIZATION OF  
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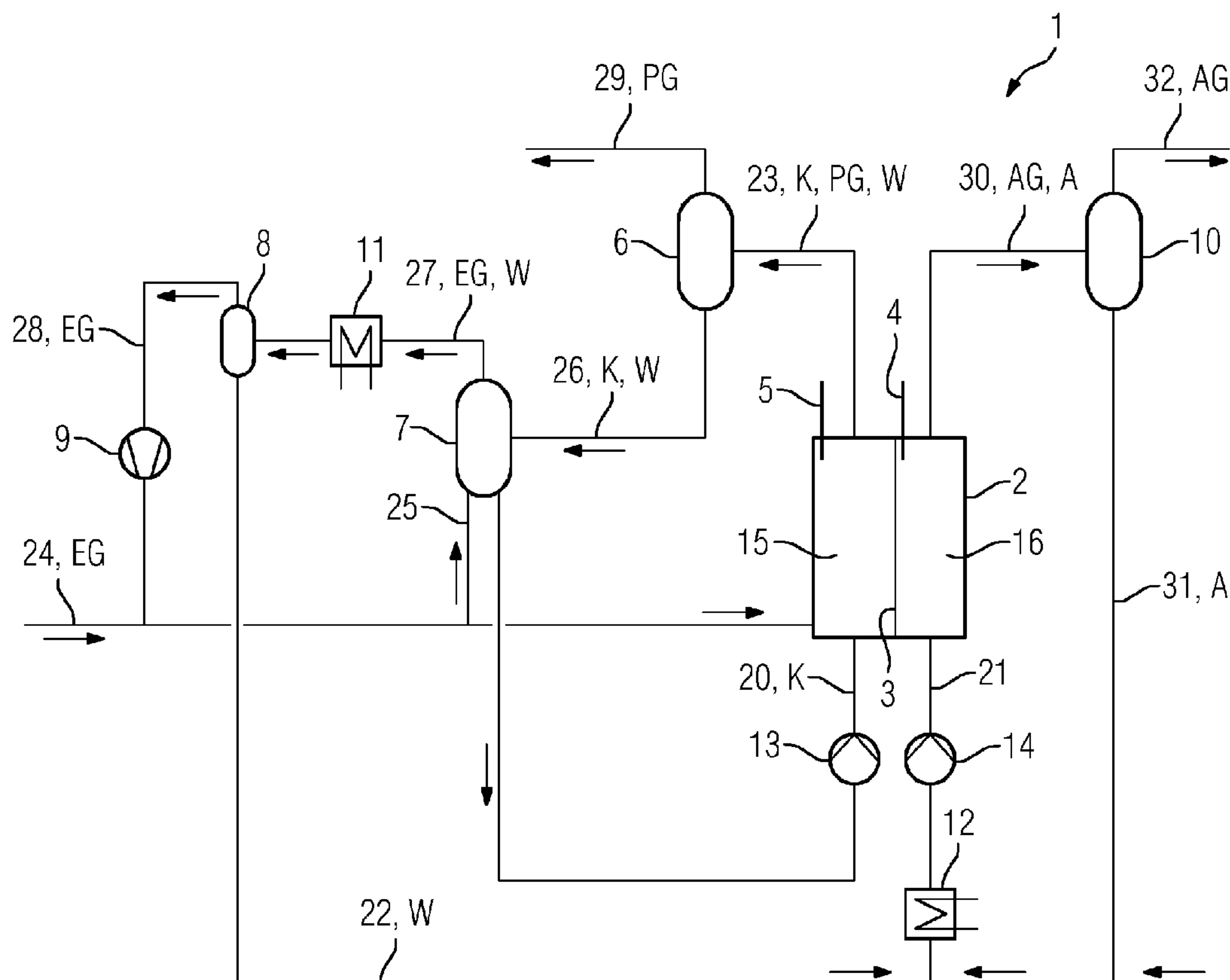
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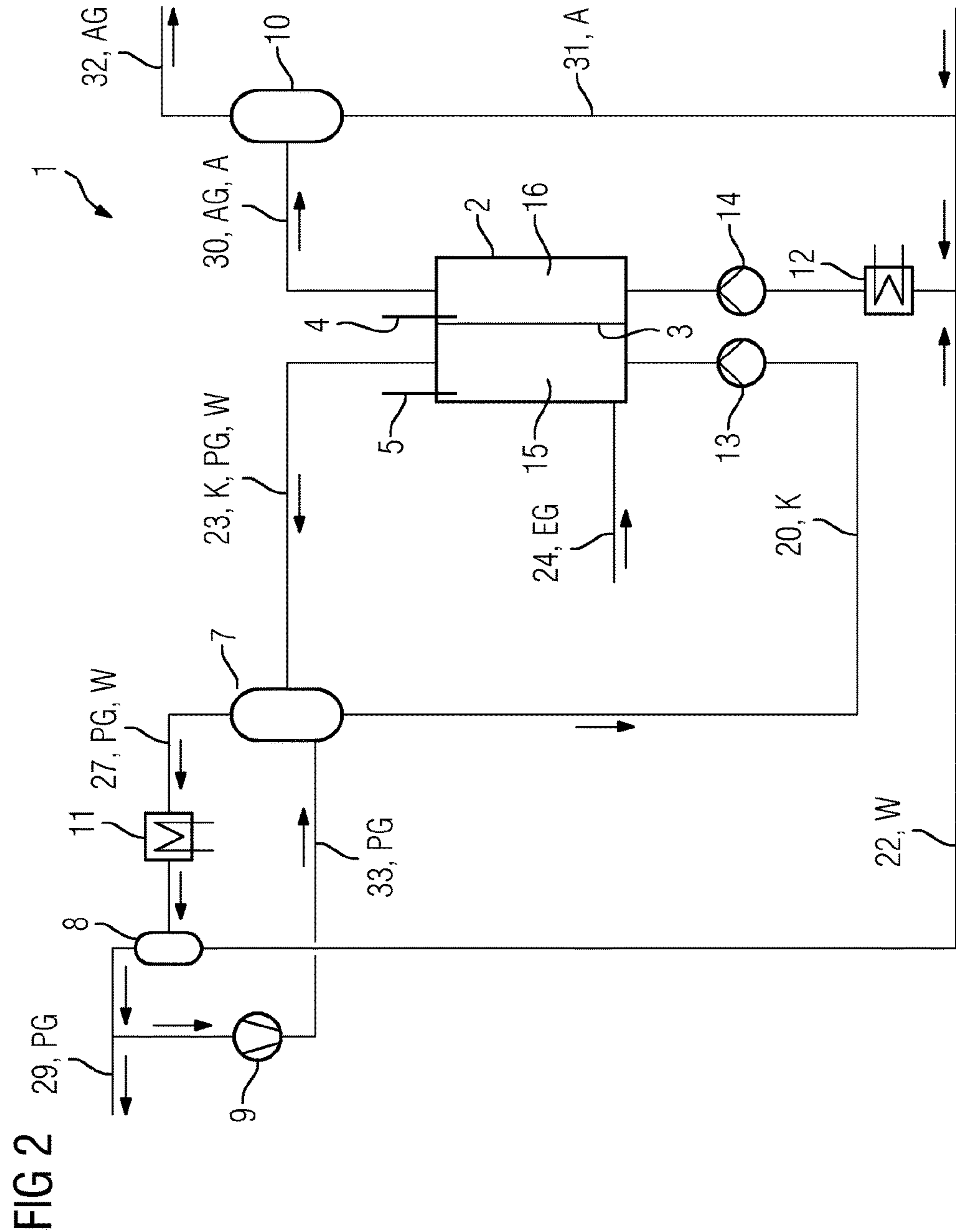
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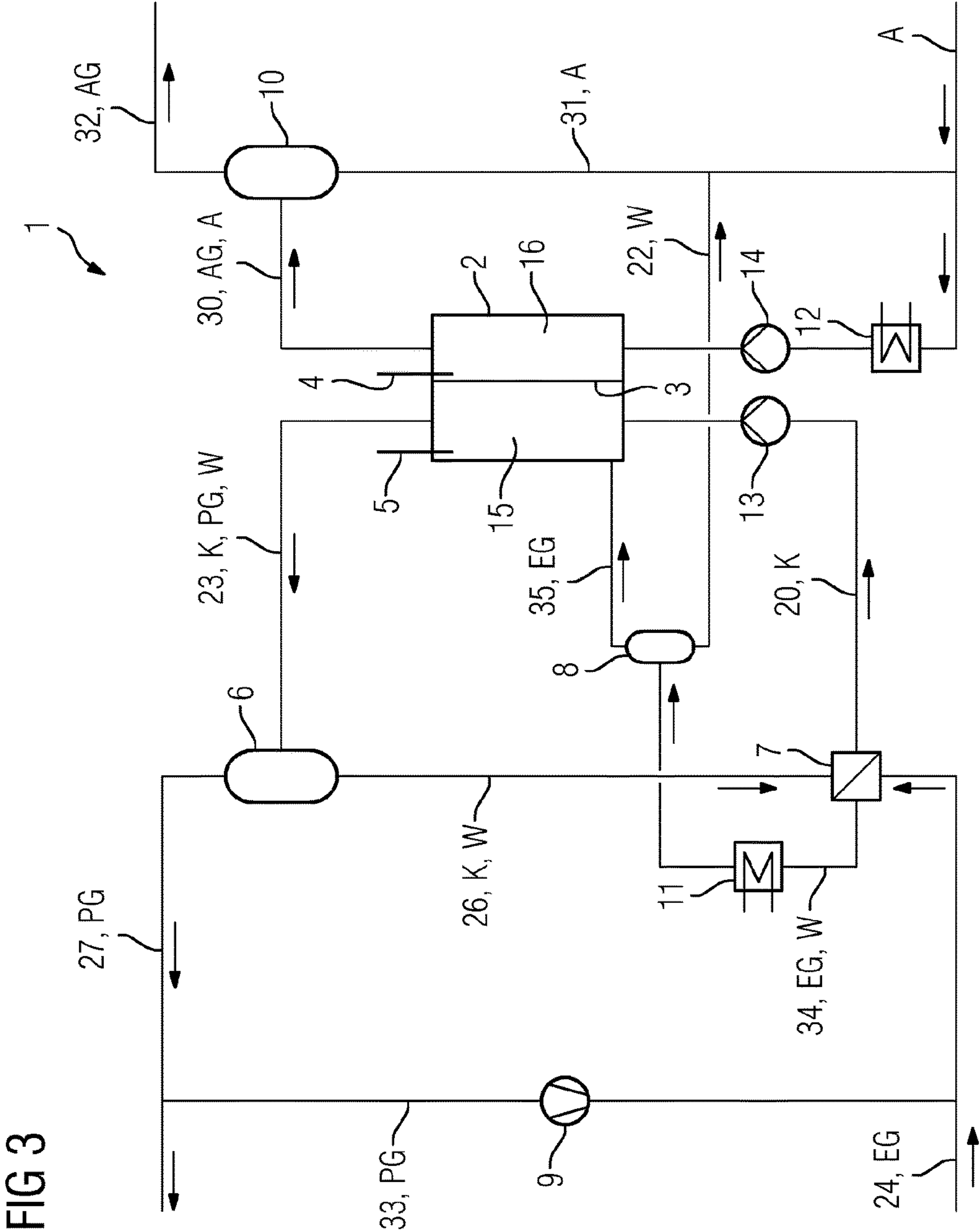
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Various embodiments may include a method comprising: reducing carbon dioxide to a product gas comprising carbon monoxide in a electrolysis cell; separating the product gas from the catholyte in a first separation apparatus; separating off excess water in the catholyte enriched with water from the anolyte during the reduction of carbon dioxide with a stripping gas in a second separation apparatus; separating the water from the stripping gas in a third separation apparatus; and recycling the water from the third separation apparatus into the anode space.











# METHOD AND DEVICE FOR THE ELECTROCHEMICAL UTILIZATION OF CARBON DIOXIDE

## CROSS-REFERENCE TO RELATED APPLICATIONS

[0001] This application is a U.S. National Stage Application of International Application No. PCT/EP2017/053742 filed Feb. 20, 2017, which designates the United States of America, and claims priority to DE Application No. 10 2016 202 840.5 filed Feb. 24, 2016, the contents of which are hereby incorporated by reference in their entirety.

## TECHNICAL FIELD

[0002] The present disclosure relates to chemical processes. Various embodiments may include a method and/or an apparatus for electrochemical utilization of carbon dioxide, wherein carbon dioxide is introduced into an electrolysis cell and reduced at a cathode.

## BACKGROUND

[0003] The demand for power varies significantly over the course of the day. There is also variation in the generation of power, with an increasing proportion of power from renewable energies during the course of the day. In order to be able to compensate for an oversupply of power in periods with a lot of sun and strong wind when demand for power is low, controllable power plants or storage means are required to store this energy.

[0004] One of the solutions currently contemplated is conversion of electrical energy to products of value which can serve as platform chemicals or synthesis gas which comprises carbon monoxide and hydrogen. One possible technique for conversion of electrical energy to products of value is electrolysis. The electrolysis of water to hydrogen and oxygen is a method known in the prior art. But the electrolysis of carbon dioxide to carbon monoxide has also been a subject of research for a number of years, and there are efforts to develop an electrochemical system that can reduce an amount of carbon dioxide in accordance with economic interests.

[0005] At present, about 80% of global energy demand is covered by the combustion of fossil fuels, and the processes of combustion thereof cause global emissions of about 34 000 million tons of carbon dioxide into the atmosphere per year. Carbon dioxide is one of the “greenhouse gases”, the adverse effects of which on the atmosphere and the climate are a matter of discussion. Utilization of this carbon dioxide is therefore desirable.

[0006] Some electrolysis units include a low-temperature electrolyzer in which carbon dioxide as reactant gas is metered into a cathode space with the aid of a gas diffusion electrode. The carbon dioxide is reduced to carbon monoxide at a cathode of the electrochemical cell, and water is oxidized to oxygen at an anode. Owing to diffusion limitations at the cathode, use of an aqueous electrolyte can result not only in the formation of carbon monoxide but also in the formation of hydrogen, since the water in the aqueous electrolyte is likewise electrolyzed. The cathode space and the anode space may be separated by a membrane in order to guide the product gases formed, the carbon monoxide and hydrogen on the cathode side and oxygen on the anode side,

out of the electrolysis cell separately from one another in order to prevent them from contaminating one another.

[0007] During the electrolysis, the transport of cations, especially of hydrogen ions, through the membrane is accompanied by simultaneous transport of water molecules. Three to ten water molecules permeate from the anode side to the cathode side for every cation. The aqueous electrolyte therefore loses water on the anode side, while the aqueous electrolyte on the cathode side becomes ever further diluted by water, and so the conductivity here disadvantageously falls considerably. There is a disadvantageous increase in ohmic potential drops in the catholyte, which prevents sustained continuous operation.

## SUMMARY

[0008] The teachings of the present disclosure provide a method and an apparatus enabling sustained continuous operation of the carbon dioxide electrolyzer with sufficiently high conductivity of the electrolyte, especially of the catholyte. For example, some embodiments of the teachings herein include a method of electrochemical utilization of carbon dioxide (CO<sub>2</sub>), comprising the following steps: providing an electrolysis cell (2), wherein the electrolysis cell (2) comprises a membrane (3) which separates an anode space (16) from a cathode space (15), guiding a first aqueous electrolyte as catholyte (K) into the cathode space (15) and guiding a second aqueous electrolyte as anolyte (A) into the anode space (16), reducing the carbon dioxide (CO<sub>2</sub>) to a product gas (PG) comprising carbon monoxide (CO), separating the product gas (PG) from the catholyte (K) in a first separation apparatus (6), separating off excess water in the catholyte (K) enriched with water (W) from the anolyte (A) during the reduction of carbon dioxide (CO<sub>2</sub>) by means of a stripping gas in a second separation apparatus (7), separating the water (W) from the stripping gas in a third separation apparatus (8), and recycling the water (W) from the third separation apparatus (8) into the anode space (16).

[0009] In some embodiments, the stripping gas used is a reactant gas (EG) comprising carbon dioxide (CO<sub>2</sub>) and/or the product gas (PG) from the electrolysis cell (2).

[0010] In some embodiments, the volume flow rate of the stripping gas is increased with the aid of at least one fan unit (9).

[0011] In some embodiments, the stripping gas is at least partly recycled into the electrolysis cell (2) as reactant gas (EG) after the water (W) has been separated off.

[0012] In some embodiments, the stripping gas is heated up prior to entry into the second separation apparatus (7).

[0013] In some embodiments, the catholyte (K) separated from excess water is recycled from the third separation apparatus (8) into the cathode space (15).

[0014] In some embodiments, a fraction not exceeding one eighth of the excess water (W) is already separated from the product gas (PG) by means of a condenser downstream of the first separation apparatus (6).

[0015] As another example, some embodiments include an electrolysis apparatus (1) for reducing carbon dioxide (CO<sub>2</sub>) to carbon monoxide (CO), comprising: an electrolysis cell (2), wherein the electrolysis cell (2) comprises a membrane (3) which separates an (16) from a cathode space (15), a first conduit (20) for guiding a first electrolyte as catholyte (K) into the cathode space (15), a second conduit (21) for guiding a second electrolyte as anolyte (A) into the anode space (16), a first separation apparatus (6) for separating a



product gas (PG) comprising carbon monoxide (CO) from the catholyte (K), a second separation apparatus (7) for separating off the excess water in the catholyte (K) by means of a stripping gas, a third separation apparatus (8) for separating the water (W) from the stripping gas, and a third conduit (22) from the third separation apparatus (8) to the anode space (16) of the electrolysis cell (2) for recycling the water (W) into the anode space (16).

[0016] In some embodiments, the second separation apparatus (7) comprises a pervaporation unit, a falling-film evaporator or a bubble column.

[0017] In some embodiments, the membrane (3) comprises an anode (A).

[0018] In some embodiments, there is at least one fan unit (9) disposed in the conduits that guide stripping gas to increase the volume flow rate of the stripping gas.

[0019] In some embodiments, the first separation apparatus (6) is integrated into the second separation apparatus (7).

#### BRIEF DESCRIPTION OF THE DRAWINGS

[0020] Further configurations, forms, and features of the embodiments of the teachings herein are elucidated in detail by the figures which follow. These are purely illustrative configurations; forms and combinations of features do not imply any restriction of the scope of protection. Features having the same mode of action and the same name but in different configuration forms are given the same reference signs. All apparatus features are referred to by numbers, all components by letters.

[0021] The figures show:

[0022] FIG. 1 a schematic arrangement of the electrolysis apparatus, wherein the stripping gas used is the reactant gas, according to teachings of the present disclosure;

[0023] FIG. 2 a schematic arrangement of the electrolysis apparatus, wherein the stripping gas used is the product gas, according to teachings of the present disclosure;

[0024] FIG. 3 a schematic arrangement of the electrolysis apparatus, wherein the stripping gas used is both the reactant gas and the product gas, according to teachings of the present disclosure.

#### DETAILED DESCRIPTION

[0025] Various embodiments include and/or enable electrochemical utilization of carbon dioxide. Some embodiments include a method, in which, first of all, an electrolysis cell is provided, wherein the electrolysis cell comprises a membrane which separates an anode space from a cathode space. Then a first aqueous electrolyte as catholyte is guided into the cathode space, and a second aqueous electrolyte as anolyte into the anode space. The carbon dioxide is then reduced to a product gas comprising carbon monoxide in the cathode space. The product gas and the catholyte are then guided into a first separation apparatus. In the first separation apparatus, the product gas is separated from the catholyte. The catholyte is then guided into a second separation apparatus. The catholyte enriched with water from the anolyte during the reduction of carbon dioxide is separated from this excess water by means of a stripping gas in this second separation apparatus. In a third separation apparatus, the water is subsequently separated from the stripping gas. This water is subsequently recycled from the third separation apparatus into the anode space.

[0026] In some embodiments, an electrolysis apparatus for reducing carbon dioxide to carbon monoxide comprises an electrolysis cell, wherein the electrolysis cell comprises a membrane which separates an anode space from a cathode space. It further comprises a first conduit into the cathode space for guiding a first electrolyte as catholyte into the cathode space. It further comprises a second conduit into the anode space for guiding a second electrolyte as anolyte into the anode space. The electrolysis apparatus further comprises a first separation apparatus for separating a product gas comprising carbon monoxide from the catholyte. It further comprises a second separation apparatus for separating off excess water in the catholyte by means of a stripping gas in a second separation apparatus. It further comprises a third separation apparatus for separating the water from the stripping gas. The electrolysis apparatus further comprises a third conduit from the third separation apparatus to the anode space of the electrolysis cell for recycling the water into the anode space.

[0027] In some embodiments, the first separation apparatus comprises a precipitation vessel. In the precipitation vessel, gaseous components are separated from liquid components. The third separation apparatus may comprise a condenser in which the water is condensed out of the stripping gas. In some embodiments, at least two membranes in the form of membrane stacks are used in the electrolysis cell.

[0028] In some embodiments, the water-diluted catholyte is separated from the excess water with the aid of a stripping gas. The catholyte is thus concentrated again, which increases its conductivity. Continuous operation of the carbon dioxide electrolyzer is possible in this way. Furthermore, the operation of the carbon dioxide electrolyzer is more economically viable compared to noncontinuous operation. Dilution effects resulting from the water may be avoided, and so the conductivity of the catholyte remains at a sufficiently high level to assure continuous operation. Separation solely of water in the first separation apparatus in which the product gas is separated from the catholyte is insufficient. At an estimate, only a maximum of one eighth of the necessary amount of water is discharged from the process here. Thus, a predominant portion of the water that has gone through the membrane still remains in the catholyte.

[0029] In some embodiments, a second separation apparatus enables the separating of a sufficiently large proportion of water from the catholyte to keep the conductivity of the catholyte at a sufficiently high level. The third separation apparatus may provide the recovery of the water from the stripping gas in order to recycle it into the electrolysis process and especially to use it again in the anolyte space. The stripping gas is also dried and can be reused in the process.

[0030] In some embodiments, the stripping gas used is a reactant gas comprising carbon dioxide and/or the product gas from the electrolysis cell that likewise comprises as yet unconverted carbon dioxide. It is advantageous here to avoid the use of a further stripping gas. Carbon dioxide is thus present both in the reactant gas and in the product gas and can be used as stripping gas for the water.

[0031] In some embodiments, the reactant gas or product gas as stripping gas can be recycled into the electrolysis cell after the stripping, such that any product gas components that have escaped into the gas phase are recycled back into



the electrolyzer and thus may not be lost. Moreover, when the reactant gas or product gas is used as stripping gas, no additional cooling of the catholyte downstream of the electrolysis cell or, alternatively, only a cooling apparatus with low output is needed for cooling of the catholyte from the electrolysis cell. In some embodiments, the water-saturated stripping gas leaves the second apparatus and is separated in the third apparatus, especially a gas/liquid separator, to give condensate and a gas phase. The heat from the catholyte may thus be discharged via the stripping gas and this condenser.

**[0032]** In some embodiments, the volume flow rate of the stripping gas is increased with the aid of at least one fan unit. A fan unit may increase the volume flow rate, and so the volume flow rate of the reactant gas and/or the product gas is increased such that this volume flow alone as stripping gas separates the water from the catholyte in a sufficient amount. The fan unit is typically disposed in a conduit that guides the stripping gas. The fan is may be arranged such that a portion of the carbon dioxide in the reactant gas or product gas is removed and circulated. The fan unit is thus within this mass transfer cycle.

**[0033]** In some embodiments, the stripping gas is at least partly recycled into the electrolysis cell as reactant gas after the water has been separated off. In this way, components that have also been transferred into the gas phase during the stripping but are products of value are recycled into the electrolysis cell. Both the components of the product gas and those of the reactant gas can be recycled into the electrolysis cell as reactant gas. Even though recycling of product gas into a reactant stream can lead to dilution effects, there are nevertheless advantages that balance these disadvantages. For example, such a measure can increase the selectivity of carbon dioxide electrolyzers with regard to carbon monoxide and suppress the side reaction for formation of hydrogen. In some embodiments, this also avoids the loss of, for example, carbon monoxide or carbon dioxide which is used as reactant.

**[0034]** In some embodiments, the stripping gas is heated up prior to entry into the second separation apparatus. This is helpful especially when the temperature of the catholyte downstream of the electrolysis cell is insufficiently high to preheat the stripping gas such that the water is transferred from the catholyte to the stripping gas.

**[0035]** In some embodiments, the catholyte separated from excess water is recycled from the third separation apparatus into the cathode space. In some embodiments, the catholyte is thus reused, and so the continuous supply of new additional catholyte into the electrolysis cell is avoided.

**[0036]** In some embodiments, a fraction not exceeding one eighth of the excess water is already separated by means of a condenser from the product gas which is guided out of the first separation apparatus. This water can be recycled into the anode space. Owing to the phase equilibrium in the first separation apparatus, the gas phase also comprises water as well as the product gas. It is thus possible to reduce the amount of stripping gas required in the second separation apparatus. It is possible either for an additional condenser to be disposed in the product gas conduit downstream of the first separation apparatus or for the product gas comprising the water to be guided into a condenser downstream of the second separation apparatus. In this case, the two streams, the product gas stream and the water-laden stripping gas, are guided separately and in parallel through the condenser, such that there is no mass transfer.

**[0037]** In some embodiments, the second separation apparatus is a pervaporation unit, a falling-film evaporator or a bubble column. A bubble column may be suitable when solely the product gas is used as stripping gas. In this case, the first separation apparatus and the second separation apparatus have been combined. In this apparatus, the product gas is separated out and a portion of the water is stripped out of the catholyte at the same time. This construction dispenses with components in the apparatus, which contributes to the simplicity of the apparatus and saves energy.

**[0038]** The pervaporation unit is advantageous when both product gas and reactant gas are used for stripping of the water out of the catholyte. The pervaporation unit provides a large area for contact between the stripping gas and the catholyte can be provided in a relatively small design volume.

**[0039]** In some embodiments, during the electrolysis, there is transport of cations, especially protons, and simultaneous transport of water through the membrane **3**. Three to ten molecules of water are transferred from the anode space **16** into the cathode space **15** for every cation. Thus, the anolyte A loses water W, while the catholyte K is diluted over time. The conductivity of the catholyte K is distinctly reduced by the dilution with water W, which increases ohmic potential drops. In order nevertheless to be able to assure sustained operation of the carbon dioxide electrolyzer, the water has to be separated from the catholyte K. The three working examples show how the water W can be effectively separated.

**[0040]** FIG. **1** shows, in schematic form, an electrolysis apparatus having an electrolysis cell **2**. The electrolysis apparatus comprises a membrane **3**, wherein the membrane **3** separates a cathode space **15** from an anode space **16**. It further comprises an anode **4** and a cathode **5**. In this configuration, the anode **4** has been applied to the membrane **3**. This reduces the necessary amount of anolyte A in the anode space **16**. The cathode **5** may comprise a gas diffusion electrode.

**[0041]** In addition, the electrolysis apparatus **1** comprises a first separation apparatus **6**. The fourth conduit **23** leads from the cathode space **15** into the first separation apparatus **6**. The catholyte K, the product gas PG and water W are guided within the fourth conduit **23**. The first separation apparatus **6** may comprise a precipitation apparatus. The gaseous product gas PG is separated here from the catholyte K and water W. The tenth conduit **29** then guides the product gas PG out of the electrolysis apparatus **1**. The seventh conduit **26** guides the catholyte K separated off in the first separation apparatus **6** and the water KW into the second separation apparatus **7**.

**[0042]** In this example, the second separation apparatus **7** is configured as a falling-film evaporator. The sixth conduit **25**, branching off from the fifth conduit **24**, guides the reactant gas EG into the second separation apparatus **7**. The reactant gas EG functions here as stripping gas. The reactant gas EG is laden with water W in the second separation apparatus and then leaves the second separation apparatus **7** via the eighth conduit **27**. Moist components are first condensed out of the reactant gas EG in a heat exchanger **11**. In a further, third separation apparatus **8**, especially a gas-liquid separator, water W is now separated from the reactant gas EG. The water W is recycled via the third conduit **22** to the anode space **16**. The reactant gas EG is guided in the ninth



conduit **28** from the third separation apparatus **8** to the fifth reactant gas-guiding conduit **24**.

[0043] A fan unit **9** is present in the ninth conduit **28**. In some embodiments, this fan unit **9** increases the volume flow rate of the reactant gas EG, such that the reactant gas EG separates a sufficient amount of water W from the catholyte K. The anode gas AG is also separated from the anolyte A in a fourth separation apparatus **10**.

[0044] In this carbon dioxide electrolysis cell **2**, the carbon dioxide CO<sub>2</sub> which is the main component of the reactant gas EG is reduced to carbon monoxide CO. The product gas PG may comprise carbon monoxide CO and unreduced carbon dioxide CO<sub>2</sub>. Oxygen O<sub>2</sub> is produced as anode gas AG at the anode A. Catholytes used may include aqueous solutions of potassium sulfate, potassium hydrogencarbonate, and/or potassium phosphate. It is also possible to use further sulfates, hydrogencarbonates and phosphates. Mixtures of these substances are also possible. In general, conductivity of the aqueous catholyte must be at a maximum without precipitation of salts. Typically, these aqueous solutions are present with salt concentrations of 0.5 mol/L to 1 mol/L. As a result of the direct application of the anode to the membrane, water is used as anolyte. However, it is likewise possible to use the salts mentioned above for the catholyte.

[0045] FIG. 2 shows one embodiment with an electrolysis apparatus **1** in which the product gas PG is used for stripping of the water W out of the catholyte K. As shown in FIG. 1, the electrolysis apparatus **1** comprises an electrolysis apparatus **2** comprising a membrane **3**, an anode **4**, a cathode **5**, and an anode space **16** separated from the cathode space **15** by the membrane **3**.

[0046] The reactant gas EG is guided into the electrolysis cell **2** via a fifth conduit **24**. The reactant gas EG comprises carbon dioxide CO<sub>2</sub> which is reduced to carbon monoxide CO in the cathode space **15**. The catholyte K that has been enriched with water W leaves the electrolysis cell **2** for the second separation apparatus **7** together with the product gas PG comprising carbon monoxide CO and unreduced carbon dioxide CO<sub>2</sub> via the fourth conduit **23**.

[0047] In this working example, the first separation apparatus **6** and the second separation apparatus **7** are combined in one apparatus. In some embodiments, the second separation apparatus **7** comprises a bubble column. The product gas PG comprising carbon monoxide CO and carbon dioxide CO<sub>2</sub> is guided here by means of the fourteenth conduit **33** into the second separation apparatus **7**. The product gas PG is enriched with water W therein and leaves the second separation apparatus **7** via the eighth conduit **27**. The product gas PG and the water W are then guided through a first heat exchanger **11**, where a fraction of water W is already condensed. The two phases are then separated from one another in the third separation apparatus **8**. The water W is recycled via the third conduit **22** into the anode space **16**. The product gas PG is partly conducted out of the system via the tenth conduit **29**. A second fraction is recycled to the second separation apparatus **7** via the fourteenth conduit **33**.

[0048] In order to increase the volume flow rate, a fan unit **9** is introduced into the fourteenth conduit **33**. In some embodiments, the stripping gas is present in a sufficient amount to separate the excess water W from the catholyte K. The catholyte K can then be recycled into the cathode space **15** via the first conduit **20**. The pump **13** ensures the transport of the catholyte K. The oxygen gas formed at the

anode, anode gas AG, is guided via the eleventh conduit **30** into the fourth separation apparatus **10**. In the fourth separation apparatus **10**, the anode gas AG oxygen is separated from the anolyte A. The anolyte A is recycled into the anode space **16** via a twelfth conduit **31**. The anode gas AG oxygen leaves the electrolysis apparatus **1** via a thirteenth conduit **32**.

[0049] FIG. 3 shows an electrolysis apparatus **1** in which the stripping gas used is both the product gas PG and the reactant gas EG. In some embodiments, the second separation apparatus **7** in that case is a pervaporation unit. The electrolysis cell **2** comprises, as already set out in the first two examples, a membrane **3**, an anode **4**, a cathode **5**, an anode space **16** and a cathode space **15**. The catholyte K that has been enriched with water W during the electrolysis of carbon dioxide CO<sub>2</sub> to carbon monoxide CO is guided via the fourth conduit **23** into the first separation apparatus **6**. First of all, the product gas is discharged therein via the tenth conduit **29**. The catholyte K and the water W leave the first separation apparatus **6** via the seventh conduit **26** and reach the second separation apparatus **7**. The reactant gas EG and a portion of the product gas PG which has been recycled via the fourteenth conduit **33** are guided into the pervaporation unit into the second separation apparatus **7**, the pervaporation unit. The water W is now transferred from the catholyte K through the membrane into the reactant gas and product gas EG, PG.

[0050] The reactant gas and product gas EG, PG and the water W are now guided through the fifteenth conduit **34** at first via a first heat exchanger **11** and then, in the third separation apparatus **8**, liquid components are separated from gas components. The water W is recycled via the third conduit **22** into the anode space **16**. The reactant gas and the product gas EG, PG are recycled via the sixteenth conduit **35** into the cathode space **15**. In the heat exchanger **12**, the anolyte is cooled to such an extent that it is recycled into the anode space **16** at a temperature of 20° C. to 95° C. More preferably, it is recycled into the anode space **16** at a temperature of 70° C.

What is claimed is:

1. A method of electrochemical utilization of carbon dioxide, the method comprising:

- providing a first aqueous electrolyte as catholyte into a cathode space of an electrolysis cell;
- providing a second aqueous electrolyte as anolyte into an anode space of the electrolysis cell, wherein the electrolysis cell comprises a membrane separating the anode space from a cathode space;
- reducing carbon dioxide to a product gas comprising carbon monoxide in the electrolysis cell;
- separating the product gas from the catholyte in a first separation apparatus;
- separating off excess water in the catholyte enriched with water from the anolyte during the reduction of carbon dioxide with a stripping gas in a second separation apparatus;
- separating the water from the stripping gas in a third separation apparatus; and
- recycling the water from the third separation apparatus into the anode space.

2. The method as claimed in claim 1, wherein the stripping gas comprises carbon dioxide and/or the product gas from the electrolysis cell.



3. The method as claimed in claim 2, further comprising increasing the volume flow rate of the stripping gas with a fan.

4. The method as claimed in claim 2, further comprising at least partially recycling the stripping gas into the electrolysis cell as reactant gas after the water has been separated off.

5. The method as claimed in claim 1, further comprising heating the stripping gas prior to entry into the second separation apparatus.

6. The method as claimed in claim 1, further comprising recycling the catholyte separated from excess water from the third separation apparatus into the cathode space.

7. The method as claimed in claim 1, wherein a fraction not exceeding one eighth of the excess water is separated from the product gas by a condenser downstream of the first separation apparatus.

8. An electrolysis apparatus for reducing carbon dioxide to carbon monoxide, the apparatus comprising:

- an electrolysis cell comprising a membrane separating an anode space from a cathode space;
- a first conduit for delivering a first electrolyte as catholyte into the cathode space;
- a second conduit for delivering a second electrolyte as anolyte into the anode space;

a first separation apparatus for separating a product gas comprising carbon monoxide from the catholyte;

a second separation apparatus for separating off excess water in the catholyte by means of a stripping gas;

a third separation apparatus for separating water from the stripping gas; and

a third conduit connecting the third separation apparatus to the anode space of the electrolysis cell for recycling the water into the anode space.

9. The electrolysis apparatus as claimed in claim 8, wherein the second separation apparatus comprises at least one of: a pervaporation unit, a falling-film evaporator, or a bubble column.

10. The electrolysis apparatus as claimed in claim 8, wherein the membrane comprises an anode.

11. The electrolysis apparatus as claimed in claim 8, further comprising a fan disposed in the conduits that guide stripping gas to increase the volume flow rate of the stripping gas.

12. The electrolysis apparatus as claimed in claim 8, wherein the first separation apparatus is integrated into the second separation apparatus.

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