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(54) **ELECTROLYSER ARRANGEMENT**

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(57) **ABSTRACT**

An electrolyser arrangement with at least one electrolytic cell, having two electrodes, namely an anode and a cathode, each of the two electrodes being in contact with an electrode compartment for filling with a liquid electrolyte, the two electrode compartments being separated by a membrane and a conveying device being provided, one for each of the two electrodes, for conveying the electrolyte in each case in a circuit, a cathode circuit and an anode circuit, through the electrode compartment via at least one collection vessel per circuit and back into the electrode chamber. A device is provided outside the electrolytic cell, for conveying an auxiliary volume flow between the cathode circuit and the anode circuit.

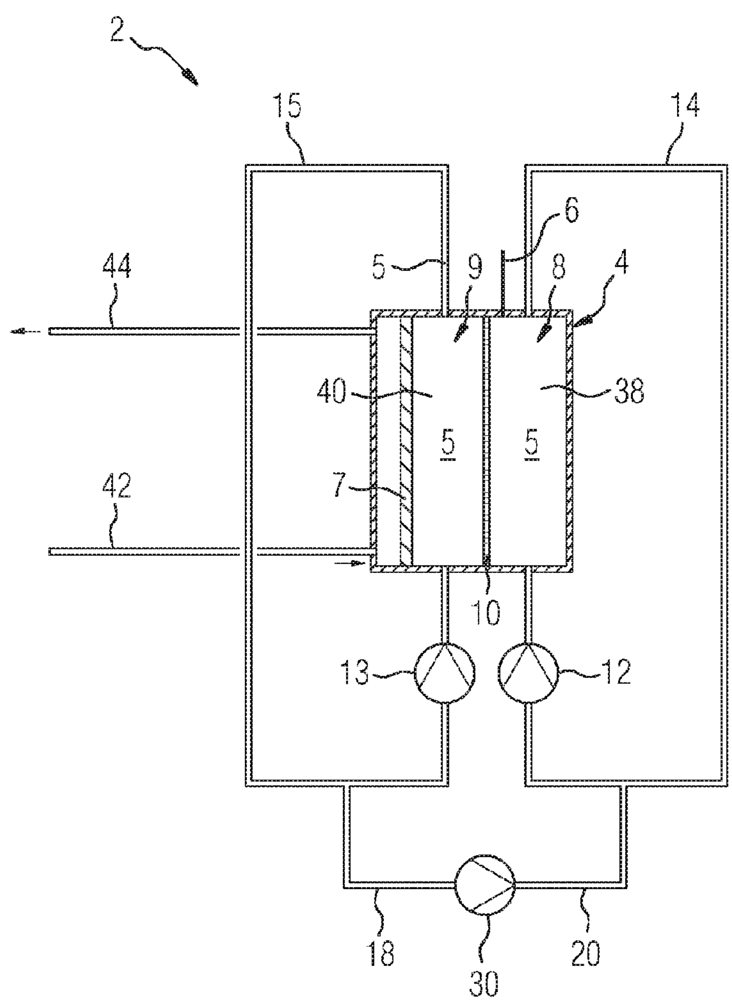


FIG 1

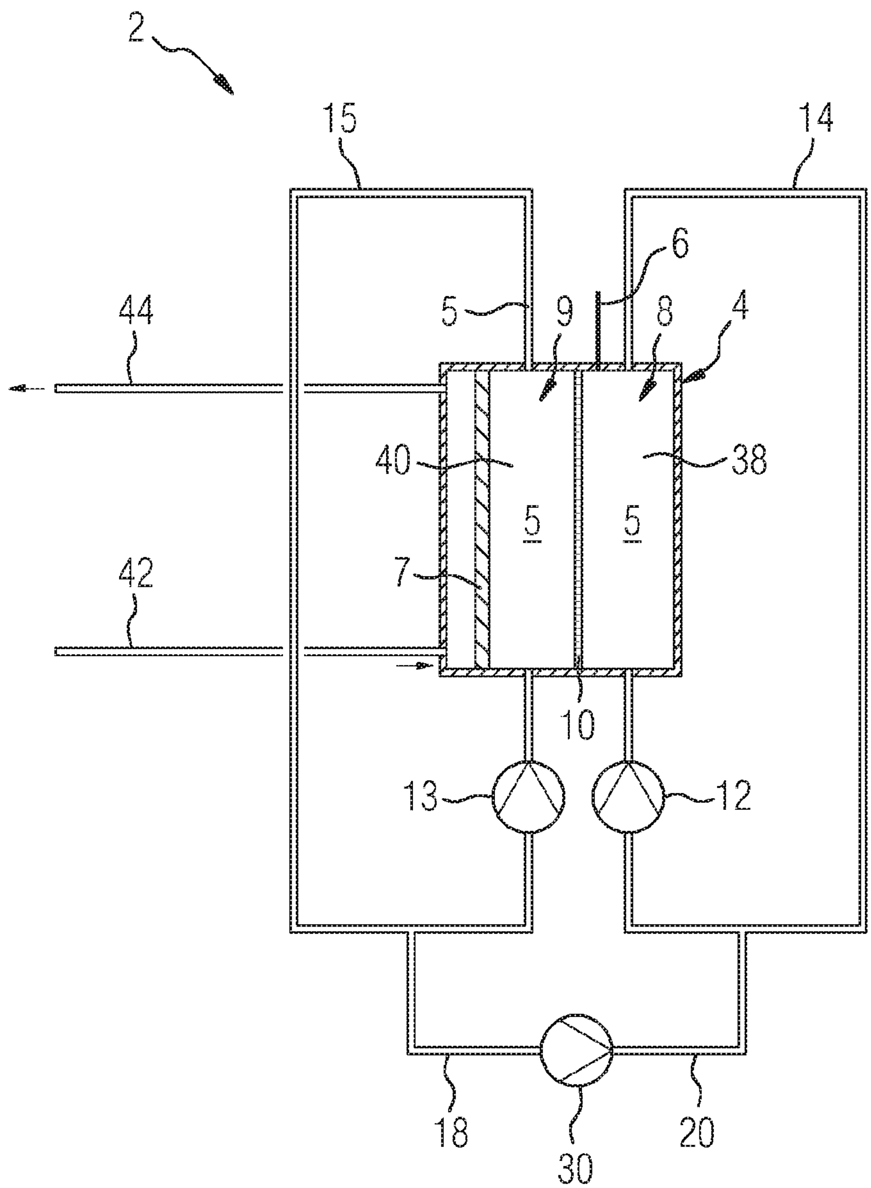


FIG 2

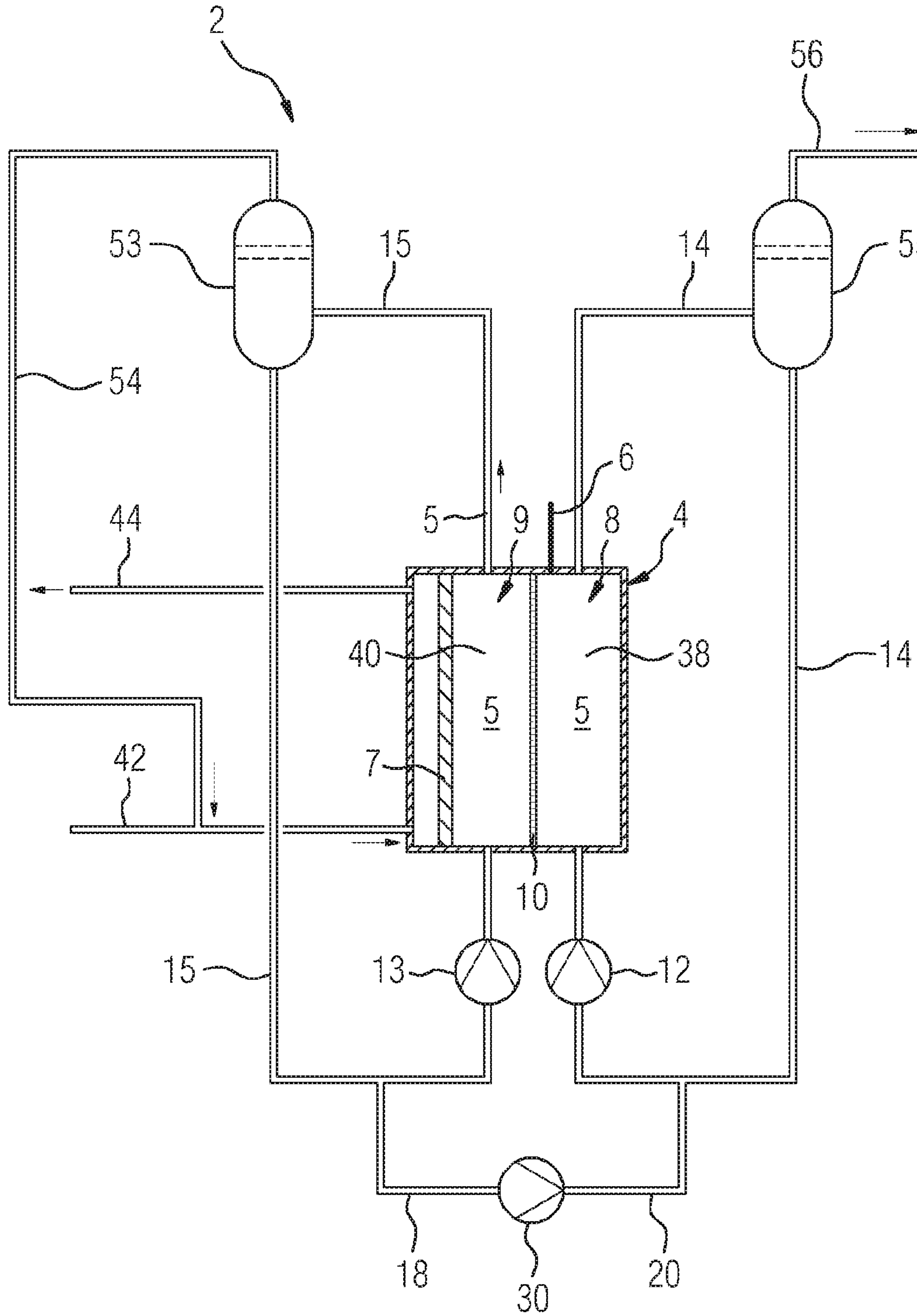


FIG 3

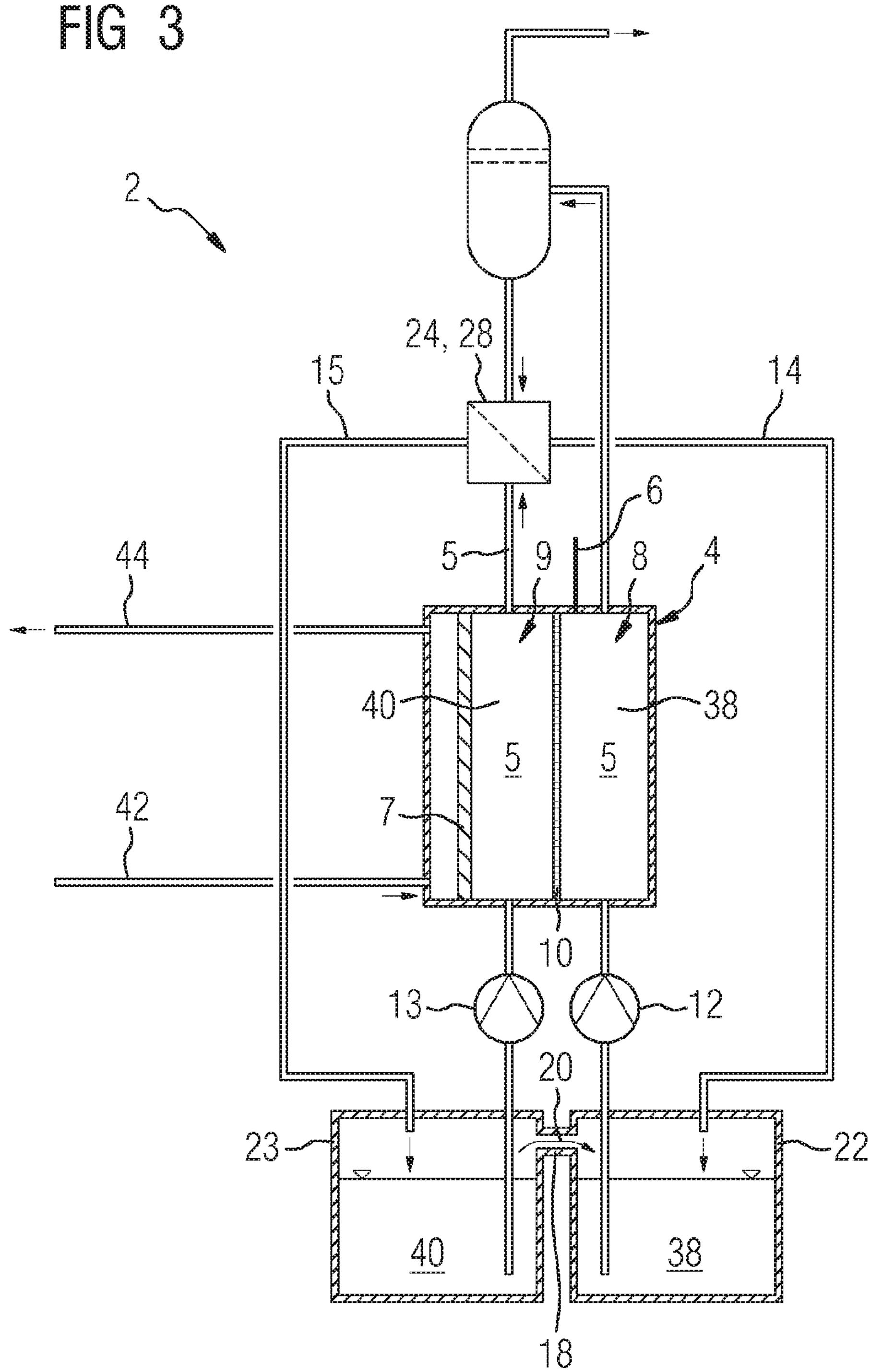


FIG 4

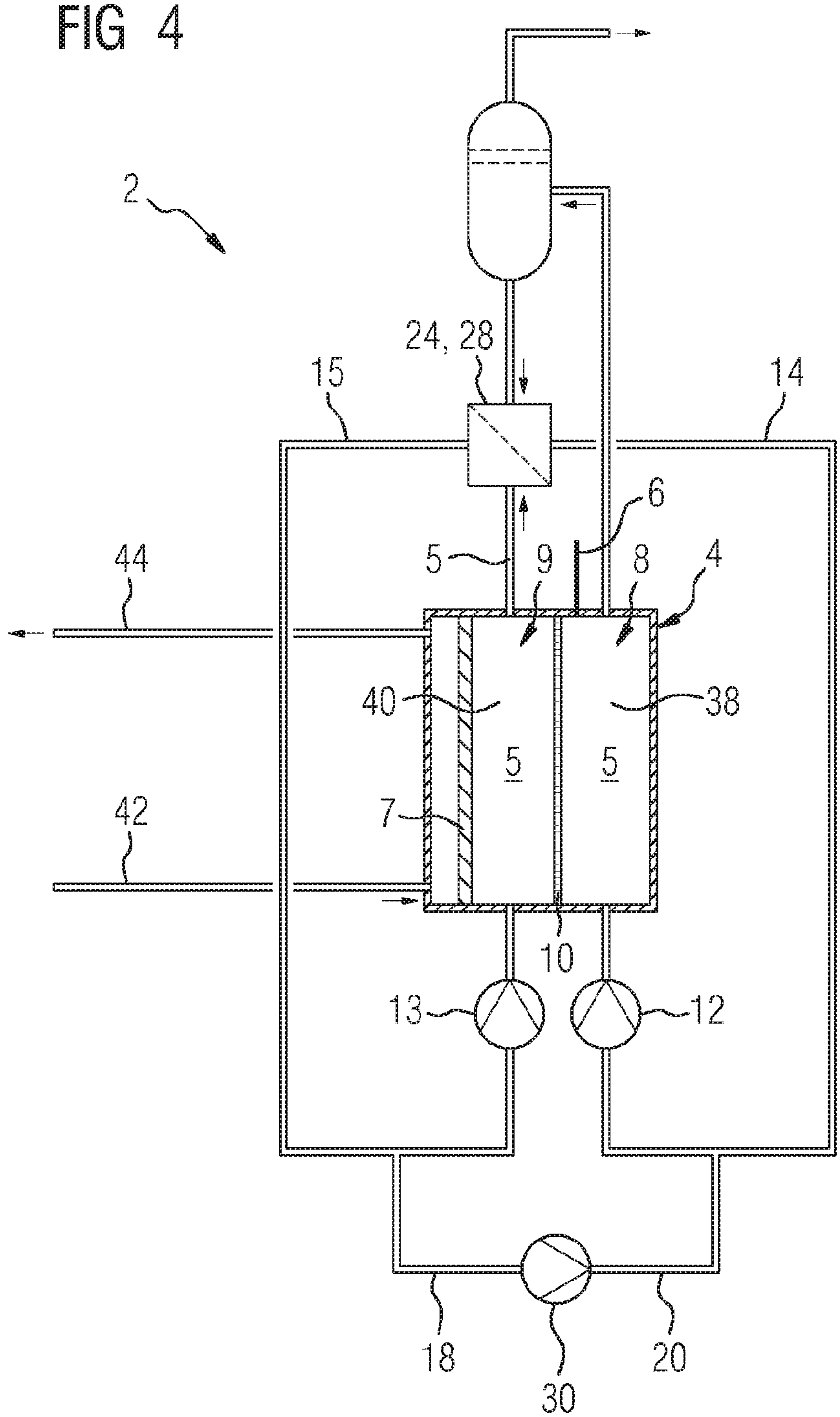


FIG 5

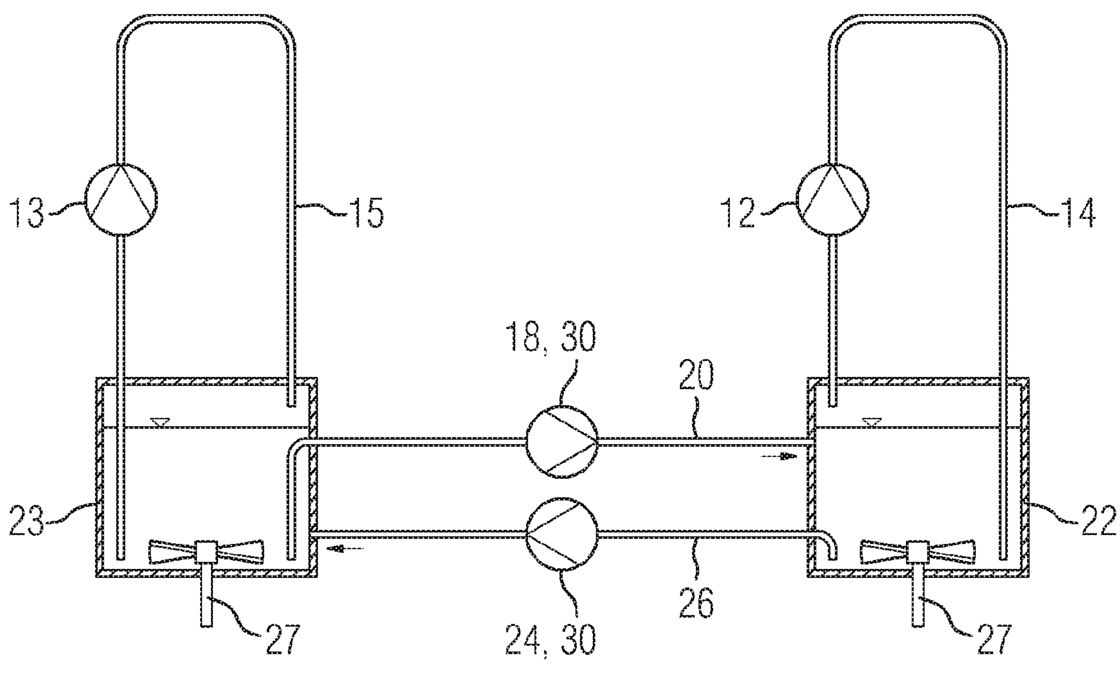
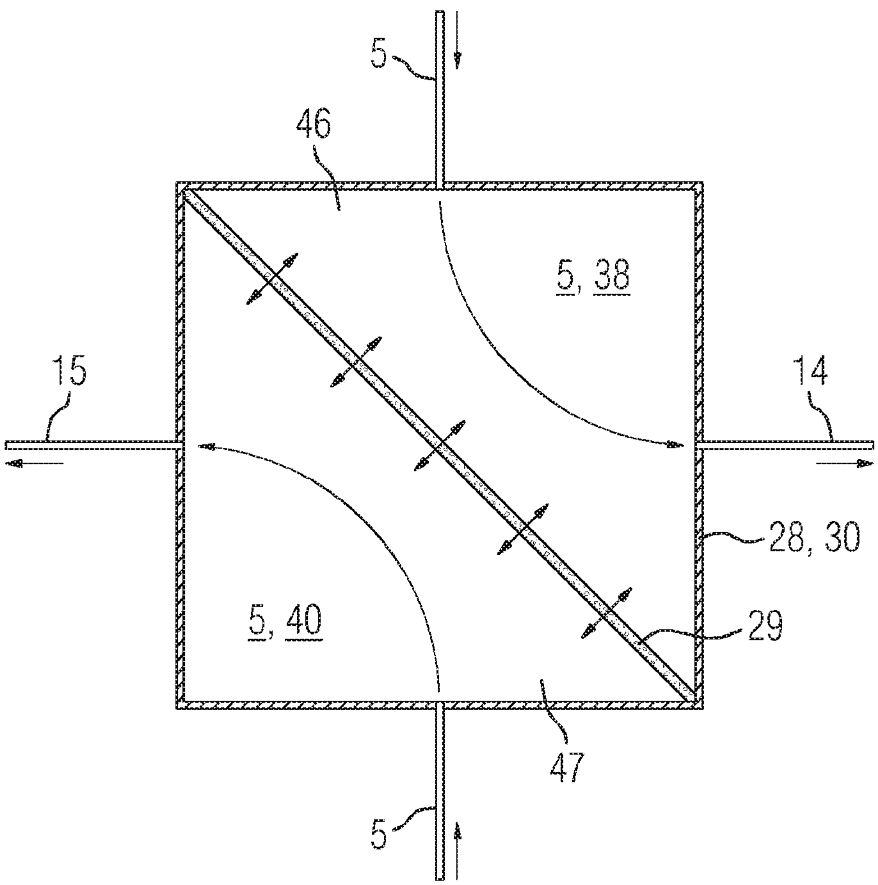


FIG 6



ELECTROLYSER ARRANGEMENT

CROSS REFERENCE TO RELATED APPLICATIONS

[0001] This application is the US National Stage of International Application No. PCT/EP2018/074697 filed 13 Sep. 2018, and claims the benefit thereof. The International Application claims the benefit of German Application No. DE 10 2017 216 710.6 filed 21 Sep. 2017. All of the applications are incorporated by reference herein in their entirety.

FIELD OF INVENTION

[0002] The invention relates to an electrolyzer arrangement and to a method for operating an electrolyzer.

BACKGROUND OF INVENTION

[0003] Major changes can be observed on the energy market at present. The use of fossil fuels is being reduced as far as possible as part of an energy turnaround, since they cause a large share of carbon dioxide emissions. At the same time, high outputs of renewable energies are available, but not always at the desired location and at the desired time. One technical challenge is to produce products of value from carbon dioxide, CO_2, using excess energies that arise in particular when renewable energies are supplied at increased levels in the network. One approach is to produce gaseous products of value, such as e.g. carbon monoxide, CO or ethylene, C_2H_4, by means of electrochemical reduction of carbon dioxide. These reactions are performed inside what are known as CO_2 electrolyzers, for example.

[0004] A typical design of CO_2 electrolyzers is based on aqueous electrolytes, which contain a conducting salt, that is to say a salt that is dissolved in the electrolyte and is electrically active. The CO_2 electrolyzers are dealt with in this instance in exemplary fashion for all electrolysis apparatuses that have a liquid electrolyte. A cation permeable membrane is used to keep the anode space and the cathode space separate from one another. This prevents a gaseous substance of value formed at the cathode from being able to get to the anode side. However, it also prevents a gas formed on the anode side, typically oxygen, from getting to the cathode side. Reciprocal mixing of the two gases is thus avoided. This is necessary in order to prevent dangerous operating states, e.g. as a result of the formation of explosive gas mixtures. However, there are other reasons to avoid mixing of the gases. By way of example, depending on the application, there are demands on gas purities of the product gas. By way of example, CO used in anerobic gas fermentation can contain only traces of oxygen.

[0005] Although the membranes used are practically impermeable to gases, they need to be permeable to ionic charge carriers. When a conducting salt is used, one frequent occurrence, however, is that the cations of the conducting salt, e.g. potassium, are transported into the foreground, i.e. the potassium cation diffuses through the membrane from the anode side to the cathode side. This in turn results in a difference in the concentration of cations between the electrolytes on the anode side and the cathode side.

[0006] By and large, it can be stated that a crossing of cations, with the exception of protons, leads to many disadvantages. It is thus desirable for the composition of anolyte, that is to say the electrolyte on the anode side, and catholyte to be kept as identical as possible. Along with the aforementioned crossing of the cations, water gets through the membrane, which leads to a thinning of the catholyte, that is to say the electrolyte on the cathode side, while the anolyte is concentrated. This effect makes keeping the composition of the anolyte and the catholyte equal as desired difficult.

[0007] It is known from the prior art that the two electrolytes can be mixed with one another in a common storage container, so that the equalization of the concentration of both ions and water is assured after passage through the electrolyzer. Since there are always gas impurities in the individual electrolyte liquids, however, said impurities resulting from the electrolysis and substantially consisting of the product gas or hydrogen and oxygen, this joint concentration equalization also holds certain risks. Additionally, a frequently required product purity is made difficult by a contamination of the product with hydrogen or with oxygen.

SUMMARY OF INVENTION

[0008] The object of the invention is to provide an electrolyzer arrangement and a method for operating an electrolyzer arrangement that are suitable for ensuring a necessary equalization of concentrations between an anolyte and a catholyte in the electrolyzer and at the same time for reducing gas contaminations.

[0009] The object is achieved by means of an electrolyzer having the features of patent claim **1** and by a method for operating the electrolyzer having the features of patent claim **12**.

[0010] The inventive electrolyzer as claimed in patent claim **1** comprises at least one electrolysis cell that in turn comprises two electrodes, namely an anode and a cathode. Each of the two electrodes is connected to what is known as an electrode space. The electrode space is suitable for being filled with a liquid electrolyte. The two electrode spaces are separated from one another by a membrane, the two electrodes comprising a conveyor apparatus for conveying the electrolyte in a respective circuit, a cathode circuit and an anode circuit. The invention is distinguished in that there is provision outside the electrolysis cell for a conveyor apparatus for conveying a secondary volume flow between the cathode circuit and the anode circuit.

[0011] The advantage of the invention described is that a secondary volume flow allows an equalization of cations and anions between the two circuits to take place. Further, it is also possible for a larger volume of water to be equalized without this resulting in substantial volumes of product gases, such as hydrogen or oxygen, being shifted between the individual circuits, so that excessive contaminations or reactive mixtures are avoided. The terms anode circuit and cathode circuit are each understood to mean an apparatus, in particular a pipeline apparatus, in particular having a pump apparatus, that is suitable for having an applicable electrolyte circulated or recirculated in it.

[0012] In one embodiment of the invention, there is provision for a respective collecting container for each of the two circuits. This has a process-engineering advantage, as care is taken to ensure that there is always sufficient electrolyte available for the two electrolyte circuits.

[0013] In one configuration of the invention, the collecting container is divided into at least two subcontainers, wherein a first subcontainer is connected to the cathode circuit and a second subcontainer is connected to the anode circuit and the secondary volume flow takes place between the first subcontainer and the second subcontainer. Equalization of the electrolytes, that is to say the anolyte and the catholyte, outside the electrolysis cell in two separate containers by means of a defined secondary volume flow, for example by a pipeline having a specific flow rate controllable by a pump, is particularly expedient because the electrolyte is collected in this subcontainer and the volume flow can easily be regulated.

[0014] In a further advantageous embodiment of the invention, there is provision for a second conveyor apparatus for producing a second secondary volume flow between the two circuits. This takes place in the opposite direction from the first secondary volume flow. This can be expedient if the first secondary volume flow routes for example water and cations from a first to a second subcontainer and equalization of anions can take place in the second secondary volume flow.

[0015] In one embodiment of the invention, the conveyor apparatus between the two circuits for producing the second secondary volume flow is configured in the form of a membrane module.

[0016] In this case, it is expedient for the membrane module to be both part of the cathode circuit and part of the anode circuit. There is likewise a membrane arranged in the membrane module, as between the two electrode spaces, said membrane being available as an exchange area for the dissolved ions. These are cations and anions.

[0017] The membrane between the electrode spaces is advantageously a cation permeable membrane. In contrast to a porous membrane, this is suitable for keeping gases from the individual electrode spaces, which arise there during the electrolysis, separate from one another. However, this also results in cations, such as for example potassium, which is part of the conducting salt, migrating through the membrane. This in turn necessitates increased equalization of concentrations between the catholyte and the anolyte outside the electrolysis cell. When a cation permeable membrane is used, the secondary volume flow advantageously takes place from the cathode circuit to the anode circuit.

[0018] A further part of the invention is a method having the features of patent claim **12**, which is suitable for operating an electrolyzer arrangement. In this case, the electrolyzer arrangement has an electrolysis cell that in turn has two electrodes, namely an anode and a cathode. The electrodes each have an electrode space through which a liquid electrolyte having a conducting salt dissolved therein is conveyed in a respective circuit, namely a cathode circuit and an anode circuit. In this case, the electrode spaces and hence also the electrolytes contained therein are separated by a membrane. The invention is distinguished in that the electrolyte is conveyed from one circuit to the second circuit in a secondary volume flow.

[0019] The method has the same advantages as have already been discussed in regard to the electrolysis arrangement. The secondary volume flow described both achieves equalization of the concentration of ions, anions and cations and also returns water, which can be in excess in one circuit, to the other circuit without at the same time producing too strong a mixing of product gases, such as oxygen and hydrogen or else carbon monoxide, in a common collecting container.

[0020] In one particular embodiment of the invention, the secondary volume flow is designed such that it has at least 0.01%, no more than 10%, advantageously between 0.1% and 1%, of the larger of the two primary volume flows, that is to say of either the volume flow of the cathode circuit or of the anode circuit. In this case, it should be noted that the term secondary volume flow is understood to mean a flow of molecules and ions, both in regard to the method and in regard to the electrolyzer arrangement. The secondary volume flow can take place in applicable pipelines, hoses or else channels, in the form of a flow of the electrolyte, in particular water-based with conducting salt contained therein and the applicable ions. On the other hand, it can also take place in the form of a diffusion through a membrane. Hence, the term conveyor apparatus for a secondary volume flow is understood to mean any apparatus suitable for providing the cited flow of molecules and ions. This firstly includes an appropriate pump, in particular, but also an appropriate line, or channel, that produces the secondary volume flow on the basis of pressure differences or gravity. Further, the term conveyor apparatus also includes a membrane that causes ions to be transferred and returned from one circuit to the other circuit.

[0021] Further, it is expedient for there to be provision for a vapor deposition container in the cathode circuit and/or in the anode circuit and for there to be provision for a connecting line from at least one of the vapor deposition containers to an educt supply apparatus. This allows anode gas and/or cathode gas, which can in turn be an educt gas for process reasons, to be supplied to the actual electrolysis process again. This has a positive influence on the economical viability of the process.

BRIEF DESCRIPTION OF THE DRAWINGS

[0022] Further embodiments and further features of the invention emerge from the drawings. These are not a limitation for the invention, since they merely describe advantageous embodiments. In the drawings:

[0023] FIG. **1** shows an electrolyzer arrangement having a secondary volume flow between the anode circuit and the cathode circuit,

[0024] FIG. **2** shows an electrolyzer arrangement as in FIG. **1** having additional deposition containers,

[0025] FIG. **3** shows an electrolyzer arrangement having two options for depicting apparatuses for a secondary volume flow having two collecting containers,

[0026] FIG. **4** shows an electrolyzer arrangement having two options for depicting apparatuses for a secondary volume flow,

[0027] FIG. **5** shows a schematic depiction of an electrolyzer arrangement, wherein two collecting containers are in the foreground, and

[0028] FIG. **6** shows a membrane module.

DETAILED DESCRIPTION OF INVENTION

[0029] FIG. **1** schematically depicts an electrolyzer arrangement **2** that has an electrolysis cell **4** in which an electrolyte **5** is arranged. The electrolysis cell **4** has two electrodes, a cathode **7**, which in this case is configured in the form of a gas permeable electrode, and an anode **6**. The two electrodes, namely the anode **6** and the cathode **7**, each adjoin an electrode space, a distinction being drawn between an electrode space **8** for the anode **6** and an electrode space **9** for the cathode **7**. The two electrode spaces **8**, **9** are separated from one another by a membrane **10**. The electrode spaces contain the electrolyte **5**, which, depending on where it is in the electrolysis cell **4**, is referred to as anolyte **38**, if it is in the electrode space **8** of the anode **6**, and which is referred to as catholyte **40** if it is in the electrode space **9** of the cathode **7**.

[0030] The electrolyte **5** or **38** and **40** is not in the electrode spaces **8** and **9** in a stationary manner, but rather is in a circuit **14**, **15**. To this end, there is provision for conveyor apparatuses **12** and **13**, which each provide the applicable volume flow of electrolyte **5** or **38** and **40** for an anode circuit **14** or a cathode circuit **15**. This involves the electrolyte **5** being moved along the respective circuit **14** (anode circuit) and **15** (cathode circuit). If the cathode circuit **15** is now considered in exemplary fashion, the catholyte **40** is pumped through the conveyor apparatus **13** from the electrode space **9** of the cathode **7** via the line provided with the reference sign **15**.

[0031] Further, there exists in the electrolyzer arrangement an educt feed **42** that introduces an educt, for example carbon dioxide, into the electrolysis cell **4**, and a product outlet **44**. During the electrolysis, which involves electric current being applied to the cathode **7** and to the anode **6**, the carbon dioxide is reduced in this example to carbon monoxide, which leaves the electrolysis cell **4** again via the product outlet **44**. During this electrolysis, both protons and the cations of a conducting salt dissolved in the electrolyte **5**, for example potassium, migrate through the membrane **10**, which is in the form of a cation permeable membrane in this embodiment. This results in the anolyte **38** and the catholyte **40** having different concentrations of cations, in particular cations of the conducting salt, as electrolysis activity increases. This can be tolerated up to a certain degree of approximately 2% difference, the economic viability and profitability of the electrolysis process no longer being ensured upward of a specific concentration difference. For this reason, it is expedient to perform a constant exchange between the anolyte **38** and the catholyte **40**. In accordance with the prior art, in an extremely simple form, a single collecting container is used that is part of both the circuit **14**, the anode circuit, and the cathode circuit **15**. In a common collecting container, which is not depicted here, good equalization of concentrations and complete mixing of the electrolyte **5** enriched or depleted in the electrolysis cell take place. However, product gases, in particular hydrogen in the cathode circuit **15** and oxygen from the anode circuit **14**, are also transferred in this common collecting container, which is not depicted here. This can lead to an explosive mixture; additionally, product gases, such as carbon monoxide, which are likewise present in the common collecting container in small amounts, are contaminated by the gases oxygen and hydrogen.

[0032] To solve this problem, there is provision for a secondary volume flow **20** to take place, which takes place via a secondary volume flow apparatus **18**. The secondary volume flow results in an exchange of concentrations taking place between the anode circuit and the cathode circuit, and vice-versa. The direction in which the secondary volume flow occurs is dependent on the respective process control. The secondary volume flow is advantageously no more than 10% of the electrolyte volume flows in the cathode circuit **15** or in the anode circuit **14**. At minimum, the secondary volume flow is 0.01% of the electrolyte volume flow; in particular, the range that covers the secondary volume flow **20** is between 0.1% an 1% of the electrolyte volume flows. If the two electrolyte volume flows are of different magnitude, then the larger of the two electrolyte volume flows is used as reference for the secondary volume flow.

[0033] It is expedient in this instance for the pH value of the anolyte to be between 4 and 5 and for the pH value of the catholyte to be between 7 and 9 in steady-state operation.

[0034] FIG. **2** provides an analogous configuration of the apparatus shown in FIG. **1**, there being provision in both the cathode circuit **15** and the anode circuit **14** for a respective deposition container **53**, **55** in which respective gaseous constituents of the electrolyte can be removed. In the case of the deposition container **53**, for example deposited carbon dioxide can be supplied to the educt supply apparatus **42** again.

[0035] In FIG. **3**, there is provision for the cathode circuit **15** to have a collecting container **23** in which the catholyte **40** is conveyed and for the anode circuit **14** to have a collecting container **22** into which the anolyte **38** is put. The two collecting containers **23** and **22** are fundamentally separate from one another, and they likewise have, but in a different embodiment, an apparatus **18** that is used to produce a secondary volume flow **20**. This apparatus **18** is depicted highly schematically in FIG. **3**; it can be configured in the form of an overflow channel, for example, that allows a small amount to get from one container to the other collecting container by means of a defined gradient or a defined slope. It is also possible for an appropriate pipeline, not depicted here, or an appropriate hose to cause a secondary volume flow **20** between the containers **22** and **23**, said flow being brought about by gravity or else by a pressure difference, for example.

[0036] FIG. **5** depicts an apparatus **18** for producing the secondary volume flow **20**, said apparatus being provided in the form of pipelines that incorporate a pump **30**. In this case, as shown in FIG. **5**, it can also be expedient, in order to ensure equalization of concentrations between the anolyte **38** and the catholyte **40** in regard to the anions, for there to be provision for a second secondary volume flow **26** produced by a second conveyor apparatus **24**, for example in the pump apparatus **30** shown in FIG. **5**. In this case, it is also expedient for the two subcontainers **22**, **23** to contain stirring apparatuses **27** that ensure uniform mixing of the electrolyte **38**, **40** in the respective containers **22** and **23**. It goes without saying that it is likewise possible to achieve good mixing inside the subcontainers without active stirring apparatuses, e.g. by means of a suitable flow guide.

[0037] If the membrane **10** used is a cation permeable membrane, a particularly large number of cations from the conducting salt migrate from the anode side, that is to say from the anolyte **38** that is in the electrode space **8** of the anode **6**, through the membrane **10** to the electrode space **9** of the cathode **7**. Together with the cations, water, what is known as drag water, also migrates through the membrane, so that equalization in particular from the cathode circuit **15** to the anode circuit **14** is necessary. In this case, when a cation permeable membrane is used, the first secondary volume flow from the cation circuit **15** to the anode circuit **14** therefore takes place. This advantageously takes place between the collecting container **23** of the cathode circuit **15** and the collecting container **22** of the anode circuit **14**, specifically in the direction described. The second secondary volume flow is then used to equalize anions that ensue between the container **22** and the container **23** via the second secondary volume flow **26**.

[0038] A further option for producing a secondary volume flow exists in the form of a membrane module **28** in which a membrane **29** is arranged (cf. FIGS. **3** and **4**). Both the cathode circuit **15** and the anode circuit **14** pass through this membrane module **28** as shown in FIG. **3**. In this case, the membrane module **28** has not only the membrane **29** but also two module chambers, a first module chamber **46** through which the anode circuit **14** runs and a second module chamber **47** through which the cathode circuit **15** goes. The module chamber **47** therefore contains the catholyte **40** and the module chamber **46** contains the anolyte **38**. The membrane **29** in this case provides an exchange area for the dissolved ions in the electrolytes **38** and **40**, specifically for cations and for anions. Porous membranes that are as thin as possible are particularly well suited to this task. These introduce a relatively low transport resistance, which means that comparatively small membrane areas are adequate. The transport in porous membranes (permeation) is caused by two different mechanisms, an externally enforced transport through pores, that is to say a purely convected transport, or a transport on the basis of diffusion of a dissolved component. The transport mechanism for the ions through the porous membrane corresponds to the diffusion, which takes place without energy consumption. What is known as the drag water can also be pushed through the membrane by convection, in principle, by applying a small differential pressure.

[0039] The necessary size of the porous membrane **29** can be ascertained by means of the maximum material flow rate of cations to be expected inside the electrolysis cell by at the same time stipulating a maximum tolerable concentration difference between the anolyte **38** and the catholyte **40** (for example 0.2 mol/l). With the aid of known passage coefficients, it is possible to estimate that when thin porous membranes **29** are used, the membrane module **28** can be configured to be distinctly smaller than the area provided therefor in the electrolysis cell **4** or the membrane **10** stretched therein. The entire membrane area of the membrane **29** is smaller than the entire electrolysis cell area of the membrane **10**, but is at least one hundredth of the membrane area of the membrane **10**. A ratio of from 1:20 between the membrane **29** and the membrane **10** to 1:5 between the membrane **29** and the membrane **10** is particularly advantageous.

[0040] The porous membrane **29** also allows water to be transported, in principle, by virtue of a lower differential pressure prevailing inside the membrane module **28**. Said pressure is advantageously less than 100 mbar.

[0041] The entire arrangement described allows intermixing of the gases produced during the carbon dioxide electrolysis of the electrolysis cell **4** to be avoided, as a result of which complex conditioning of the electrolyte **5** or of the gases produced is dispensed with. By way of example, the catholyte **40** therefore contains no oxygen that contaminates the catholyte product gas. Additionally, neither product gas (for example carbon monoxide, methane or hydrogen) nor educt gas such as carbon dioxide is lost via the anolyte **38** in practice.

[0042] Use of two separate electrolysis circuits, namely the anode circuit **14** and the cathode circuit **15**, cannot prevent a certain amount of drifting apart by the compositions and hence also by the pH values of the anolyte **38** and the catholyte **40**. In addition, drag water from the anolyte **38** gets into the catholyte **40**. Conventional conditioning would have a high level of associated energy expenditure, e.g. as a result of thermal degassing or vacuum degassing. Alternatively, the process can also have an additive added that chemically binds unwanted gases. The use of an additive has associated costs, however. In addition, it is not foreseeable to what extent possible additives influence the electrochemical process. The catalytic removal of undesirable gases likewise has a high level of associated energy expenditure. The arrangement described therefore shows a simple technical solution to ensure appropriate equalization of ions and water between the anode circuit **14** and the cathode circuit **15**.

1. An electrolyzer arrangement having comprising:
at least one electrolysis cell comprising two electrodes, namely an anode and a cathode, wherein each of the two electrodes is in contact with an electrode space for filling with a liquid electrolyte, wherein the two electrode spaces are separated by a membrane,
a conveyor apparatus for each of the two electrodes for conveying the electrolyte through the electrode space in a respective circuit, comprising a cathode circuit and an anode circuit, and
an apparatus outside the electrolysis cell for conveying a secondary volume flow between the cathode circuit and the anode circuit.

2. The electrolyzer arrangement as claimed in claim **1**, wherein the cathode circuit and the anode circuit each have a collecting container.

3. The electrolyzer arrangement as claimed in claim **2**, wherein a first secondary volume flow takes place between a first collecting container and the second collecting container.

4. The electrolyzer arrangement as claimed in claim **1**, further comprising:
a second conveyor apparatus for producing a second secondary volume flow between the two circuits, which takes place in the opposite direction from the first secondary volume flow.

5. The electrolyzer arrangement as claimed in claim **4**,
wherein the second conveyor apparatus for producing the second secondary volume flow is configured in the form of a membrane module.

6. The electrolyzer arrangement as claimed in claim **5**,
wherein both the cathode circuit and the anode circuit pass through the membrane module.

7. The electrolyzer arrangement as claimed in claim **4**, further comprising:
a pump apparatus between the collecting containers in order to bring about the second secondary volume flow.

8. The electrolyzer arrangement as claimed in claim **2**, further comprising:
an overflow or a pump apparatus between the two collecting containers in order to bring about the first partial volume flow.

9. The electrolyzer arrangement as claimed in claim **1**,
wherein the membrane between the electrode spaces is a cation permeable membrane.

10. The electrolyzer arrangement as claimed in claim **8**,
wherein the secondary volume flow takes place from the cathode circuit to the anode circuit.

11. The electrolyzer arrangement as claimed in claim **1**, further comprising:
a vapor deposition container in the cathode circuit and/or in the anode circuit and there is provision for a con-

necting line from one of the vapor deposition containers to an educt supply apparatus.

12. A method for operating an electrolyzer having at least one electrolysis cell that in turn has two electrodes, comprising an anode and a cathode, wherein each electrode has an electrode space through which a liquid electrolyte having a conducting salt dissolved therein is conveyed in a respective conveyor circuit namely in a cathode circuit and an anode circuit, in a respective primary volume flow and wherein the two electrode spaces and hence the electrolyte contained therein are separated by a membrane, the method comprising:

conveying the electrolyte from one circuit to a second circuit in a secondary volume flow.

13. The method as claimed in claim **12**,

wherein the secondary volume flow is at least 0.01 and at most 10% of the larger of the two primary volume flows.

14. The method as claimed in claim **13**,

wherein the secondary volume flow is at least 0.1 and at most 1% of the larger of the two primary volume flows.

15. The method as claimed in claim **12**, further comprising:

a collecting container in each of the two circuits;

wherein the electrolyte in the secondary volume flow is conveyed from a first collecting container to a second collecting container.

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