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(71) Applicant: **TECHNISCHE UNIVERSITEIT DELFT** [NL/NL]; Mekelweg 4, 2628 CD Delft (NL).

(72) Inventors: **SMITH, Wilson Abele**; c/o Postbus 5, 2600 AA Delft (NL). **VERMAAS, David Arie**; c/o Postbus 5, 2600 AA Delft (NL).

(74) Agent: **VOGELS, Leonard Johan Paul**; Weteringschans 96, 1017 XS Amsterdam (NL).

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(54) Title: BIPOLAR MEMBRANE ELECTRODE ASSEMBLY FOR FUEL GENERATION

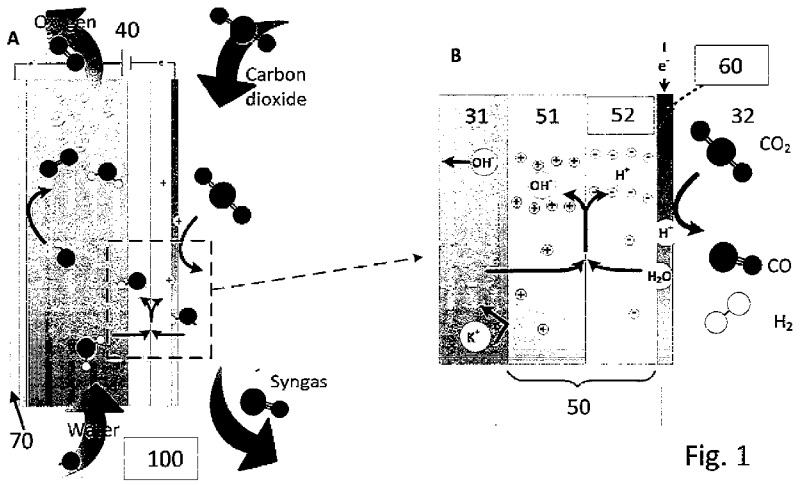


Fig. 1

(57) Abstract: The present invention is in the field of forming a chemical fuel, such as by electrolysis of water thereby forming hydrogen. Alternative approaches relate to forming hydro gen also a hydrocarbon, syngas, and an alcohol may be formed. Based on readily available fluids these chemical fuels can be produced. A source for e.g. the electrolysis may be solar radiation.

Bipolar membrane electrode assembly for fuel generation

FIELD OF THE INVENTION

The present invention is in the field of forming a chemical fuel, such as by electrolysis of water thereby forming hydrogen.

BACKGROUND OF THE INVENTION

The present invention is in the field of forming a chemical fuel, such as by electrolysis of water thereby forming hydrogen. Alternative approaches relate to forming hydrogen also a hydrocarbon, syngas, and an alcohol may be formed. Based on readily available fluids these chemical fuels can be produced. A source for e.g. the electrolysis may be solar radiation.

Electrolysis of a species as water relates to decomposition of the species (water) into its constituents (oxygen and hydrogen) by providing an electric current through said species. Thereto the species is typically in fluid form. In case of water an objective is to produce hydrogen. Electrolysis can be used to skim off excess power, such as from wind energy.

It is noted that at this point in time production of hydrogen from water is considered not competitive. There are various production methods, such as steam reforming, production of hydrogen from hydrocarbons, biological production, various forms of electrolysis, photo electrochemical water splitting, by concentrating solar energy, catalytic production, etc.

EP 0 459 820 A2 recites a bipolar membrane comprising a cation-exchange membrane closely adhered to an anion exchange membrane in which cation-exchange groups present at least at the adhered surface of the cation exchange membrane have been ion-exchanged with a heavy metal ion, has a water splitting current efficiency of not less then 80% and a water splitting membrane potential of not more than 2.0 V.
Some documents recites water-splitting devices.

US 2007/023290 A recites an electrochemical cell comprising an ion exchange membrane having anion and cation exchange materials. The membrane can have separate anion and cation exchange layers that define a heterogeneous water-splitting interface there between. In one version, the mem-

brane has a textured surface having a pattern of texture features comprising spaced apart peaks and valleys. The membranes can also have an integral spacer. A cartridge can be fabricated with a plurality of the membranes for insertion in a housing of the electrochemical cell. The housing can also have a detachable lid that fits on the cartridge. The electrochemical cell can be part of an ion controlling apparatus.

The above two documents relate at the best to a membrane, but not to a fuel generating system.

CN 102 912 374 A2 recites an electrochemical reduction CO_2 electrolytic tank using a bipolar membrane as a diaphragm and an application of the electrochemical reduction CO_2 electrolytic tank. The electrolytic tank comprises a cathode electrolysis compartment, catholyte, an anode electrolysis compartment, anolyte and the bipolar membrane for dividing the cathode electrolysis compartment and the anode electrolysis compartment. The electrode materials of the cathode electrolysis compartment include Pb, In and Cu etc., and the catholyteis an alkaline aqueous solution; and the electrode materials of the anode electrolysis compartment include Pt and Pd etc., and the anolyte is an acidic aqueous solution containing iodate. The hydroxy radicals in the cathode electrolysis compartment and the protons in the anodic electrolysis compartment are diffused to the bipolar membrane to generate water so as to form a voltage drop, so that the working voltage in the electrolytic tank is reduced. Compared with an anodic reaction that water and the electricity are oxidized to generate oxygen, the iodide ions are oxidized to generate an iodine elementary substance with low potential, small over-potential and quick dynamic process, so that the working voltage in the electrolytic tank is further reduced. CO_2 is electrically reduced in a cathode compartment so as to generate small molecular fuels, such as formate, methanol, and methane; and the iodide ions are electrically oxidized to generate the elementary substance iodine in an anode compartment.

Some typical issues with prior art systems and processes are losses in yield, having various causes. Also, when gasses are produced, mixing of gasses is sometimes difficult to prevent. Further, selectivity towards a desired end prod-

uct of a given process is often limited. And typical catalysts used can not be operated optimally.

The present invention therefore relates to an improved process and system for generating chemical fuels, which solve one or more of the above problems and drawbacks of the prior art, providing reliable results, without jeopardizing functionality and advantages.

SUMMARY OF THE INVENTION

The present invention relates to a system for producing a chemical fuel according to claim 1, and a method of making said chemical fuel according to claim 13.

In the present system use is made of a physical double layer structure (also referred to as "double layer"), such as a bipolar membrane. The double layer structure comprises two layers directly attached to one and another. Advantages of using a double layer structure with an attached electrode are; that the double layer structure provides a physical support layer for an embedded electrode, and minimizes the thickness of the electrode; the double layer structure and embedded electrode have a negligible distance between the structure (or put different, are physically attached or connected to one and another; see e.g. fig. 1), which supplies protons, and the electrode, which is found to minimize ohmic losses; the double layer structure acts as a barrier for the produced species (oxygen) at one side and the produced products (e.g., hydrogen, syngas) at another side of the structure. The prior art cited above does not have any electrode attached to the double layer structure. Clearly the numbering of specific elements of the present system may be reversed and is therefore not limiting. Hence, the electrodes can be positioned close together without mutual mixing of the produced gasses, which is found to further decrease ohmic losses; the double layer structure enables to use different electrolytes at either side, or to even use a gaseous phase at one side. Hence, the supply of species, e.g. CO_2 and H_2O, can be tuned in all ratios, which directly influences the product selectivity; the double layer structure creates an extreme pH-difference, which allows the cathode to operate in acidic conditions and the anode to operate in alkaline conditions; the double layer in an example chemically/physically

separates a base being present at one side thereof from an acid being present at another side thereof. This is found favourable for common earth abundant catalysts, such as Ni-based oxygen evolution catalysts, while CO and H_2 production are promoted at low pH; the anion-cation layered structure creates a strong barrier for any other ions than H^+ or OH^-. This feature is found to minimize contamination of the cathode with dissolved species of the anodic compartment and therefore increases the stability of the system.

The present double layer structure, in an example the present bipolar membrane, is typically not electrically conducting, or in an alternative having a high ohmic resistance (i.e. being an electrical insulator).

In the present description the term "attached" indicates that an area of a first element e.g. an electrode is at a negligible distance of an area of a second element e.g. a double membrane, stays at such a distance if a small force is applied to detach the elements, such as a force of two times gravity; i.e. if a first element is fixed the second element does not fall of if exposed to gravity. Attachment can be achieved by e.g. forming a second element from a solution or gas on the first element, such as by chemical reaction, by deposition, by (thermo-)pressing the first element on the second element, by applying an adhesive, and vice versa. The attachment can be established over a full area of the smaller of the two elements, or over a part thereof. Attachment may be disrupted, e.g. due to a sub-optimal attachment process, the nature of an element, incompatibility of elements, etc.

The present chemical fuel can in principle be any fuel that can be made from fluids by electrolysis; the fuel provides energy when it is oxidized.

The present system relates to a stack-like structure of various components. The stack may be in any suitable form, such as a stack, a tube-like stack, etc. The first electrode and first compartment are in ionic contact, hence adjacent to one and another. The first electrode may in principle be any electrode capable of oxidizing the first fluid being present in the first compartment. The electrode optionally is high surface/volume electrode.

With the term "fluid" any gaseous or liquid is indi-

cated. With the term "adjacent" it is implied two elements that are adjacent are in direct or indirect contact with one and another

In the present system the first and second compartment are physically separated by a double layer structure, such as a bipolar membrane. The double layer structure comprises two layers, a first layer adjacent to and in contact with the first fluid or first electrode, acting as an anion exchange layer (AEL), and a second layer adjacent to and in contact with the cathode, acting as a cation exchange layer (CEL). The first electrode may therefore be attached to the present double layer structure, or, in an alternative, may be separated from the double layer structure by the first fluid in the first compartment. Optionally a further layer may be present. The AEL and CEL are typically parallel and adjacent to one and another, and in an example in contact with one and another. In an example of the double layer structure a bipolar membrane is used. The double layer acts as a barrier at least for oxidized species.

With respect to the present bipolar membrane the following is noted. A bipolar membrane is typically applied in the production of acid and base. Occasionally, a bipolar membrane has been used for microbial fuel cells and fuel cells, which processes are different from the present technology. For example, the water is not being dissociated when using it in a fuel cell, but instead H^+ and OH^- recombine to water in this case. This is considered an important difference, not only because the process is opposite in goal (e.g. making water versus the present splitting water), but also because water will be accumulated in the interfacial layer in the bipolar membrane, which causes blistering and high electrical resistance of the membrane; hence the bipolar membrane is not suitable in this respect. On the contrary, in the present technology, the bipolar membrane remains stable.

A bipolar membrane has been reported for fuel production, but in both cases there was no electrode attached to the membrane. In both cases, an aqueous solution was applied at both sides of the bipolar membrane.

The present system further comprises a second electrode, which is attached to the CEL, that is in ionic contact

with said layer and adjacent to said layer.

The first electrode is typically the anode, whereas the second electrode is typically the cathode.

Adjacent to the second electrode a second compartment for reduction is present, the compartment comprising a second fluid for reduction in cation exchange contact with the second electrode. The second electrode and second compartment are in ionic contact, hence adjacent to one and another. The second electrode may in principle be any electrode capable of reducing the second fluid being present in the second compartment.

In order to provide electrical power a power source in electrical contact with the first and second electrode is provided. The power source can retrieve its power in principle from any electrical power source; if a DC source is used preferably a transformer is provided.

With the present system chemical fuels can be generated using electrochemical reduction and oxidation. The most common example thereof is the oxidation and reduction of water, where hydrogen and oxygen are evolved. As an alternative, CO_2 can be reduced, together with water oxidation, to obtain carbon monoxide. As a third example, the combination of these reactions can yield hydrocarbons, which can be used as fuel.

The present system provides a stable and selective reaction at high rate, using e.g. a system with a bipolar membrane and an embedded electrode in this membrane. A possible design of such a system 100 is described in Fig. 1.

The present invention provides design flexibility. Several variations on the present system can be created. In particular, the use of tubular membranes, with a gas phase inside the tube, is considered. The tube may be cylindrical (circular cross-section), polygonal cross-sectional, such as square, rectangular, triangular, hexa-angular, oval, etc. It provides the following advantages:

- It allows a pressurized gas phase as tubes allow a large pressure difference over the membrane.
- The tube provides a relatively short average distance for an arbitrary molecule/atom in the fluid to the electrode; similar, it provides a relatively large area with respect to

the volume. This is in particular beneficial when the gaseous phase is filled with a porous conductive material, as a charge collector for the cathode. For example, porous carbon (graphite) can be used to fill the tubes.

- The tube allows selection of a larger or smaller area for the cathode compared to the anode, respectively. This is considered beneficial if one of the two electrodes is limiting for the process selected.
- A limited number of tubes provides the possibility of having a solar panel behind the system, even when the bipolar membranes and cathodes are not transparent. Only a part of the light is interrupted by the tubes, while light can freely pass at the positions where no tube interrupts the light.

Thereby the present invention provides a solution to one or more of the above mentioned problems and drawbacks.

Advantages of the present description are detailed throughout the description.

DETAILED DESCRIPTION OF THE INVENTION

The present invention relates in a first aspect to a system for producing a chemical fuel according to claim 1.

In an example of the present system the first fluid is an aqueous solution, such as comprising a hydroxide, or a gas.

In an example of the present system the double layer is a bipolar membrane.

In an example of the present system the second fluid is a gas, such as hydrogen, carbon monoxide, carbon dioxide, and combinations thereof.

In an example of the present system the first fluid provides hydrogen ions, in order to make a chemical fuel thereof.

In an example of the present system the second fluid provides a carbon dioxide molecule.

In an example of the present system the power source is a PV-source, such as a PV-element attached to the first electrode (adjacent to the first liquid) and/or a PV-element attached to the second electrode. As such integration with present PV-systems can be made without any difficulty.

In an example of the present system the double layer is curved, such as a tube, wherein the anion exchange layer

is on a convex side of the double layer and the cation exchange layer is on a concave side of the double layer.

In an example of the present system the concave side is filled with a porous material, such as with carbon.

In an example of the present system a first electrode (70) comprises Ni or NiFe. The present system provides herewith as further advantage that good affordable materials as Ni and Fe can be used without a risk of deposition of these materials on a second electrode (cathode).

In an example of the present system a second electrode (60) comprises at least one of Cu, Pt and Ag.

In a second aspect the present invention relates to a method of producing a chemical fuel according to claim 13. The method comprises the steps of providing a system according to the invention, having a suitable first fluid and a suitable second fluid, and producing a chemical fuel, the chemical fuel being selected from hydrogen, a hydrocarbon, CO, a combination of CO and H_2, such as syngas, an alcohol, and combinations thereof. The production is established under suitable conditions. In an example a Faradaic efficiency (e_{output}/e_{input} * 100%) results in at least 30% selectivity of the production of CO. In an example a Faradaic efficiency results in at least 50% selectivity of the production of formic acid.

In an example of the present method the electric power is provided by at least one PV-system. As can be seen from e.g. fig. 1 integration of a PV-element within the present system is now possible.

In an example of the present method one or more of a cathode area, an anode area, a number of electrodes, a ratio between to be oxidized first fluid and to be reduced second fluid, and product selectivity, are tuned. Therewith reaction conditions and as a consequence e.g. yield and selectivity are further optimized.

In an example of the present method the cathode operates in acidic conditions, the pH preferably being between 1-7, more preferably between 3-6.5, such as 4-6 or 5, and/or wherein the anode operates in basic conditions, the pH preferably being between 9-14, more preferably between 10-13.5, such as between 11-13. Acidic conditions typically relate to

a pH<6, more typically a pH<4, depending on the chemical species involved, whereas the basic conditions typically relate to a pH>9, more typically a pH>10. In an example acidic conditions are a pH of 6.5 and basic conditions are a pH of 14; this is considered a unique feature of the present system, as prior art systems typically operate at much larger pH-differences c.q. gradients. In other words, under relatively mild conditions the present system provides good yields. In an example, involving CO_2, the acidic conditions are a pH of 5-6.5. At this pH a relatively large concentration of CO_2 can be present, whereas the CO_2 is typically present as HCO_3^-, which is a favourable species in view of yield.

The one or more of the above examples and embodiments may be combined, falling within the scope of the invention.

EXAMPLES

The below relates to examples, which are not limiting in nature.

With reference to figure 3 the following experimental verification was obtained under the following conditions:

A cathode 60 was made from a copper wire mesh, cut in a round shape with a diameter of approximately 5 cm and a weight of 0.98 gram. The cathode was attached to the present membrane 50 by positioning the copper mesh on top of the bipolar membrane and pipetting approximately 2 mL Nafion solution (5% w/w, dissolved in lower aliphatic alcohols and water) on the same bipolar membrane with copper mesh. After 1 hour, the solvent was evaporated, leaving the copper mesh attached to the bipolar membrane due to the dried Nafion polymer. See figure 3 for an illustration of this process. This method is only one example to create a membrane electrode assembly (MEA); other methods for attaching the cathode in the bipolar membrane are possible as well.

An anode 70 was made of a Ti plate with a Pt coating, prepared by magnetron sputtering. This anode was situated in a first compartment 31 filled with 1 M KOH and had a geometric area of approximately 4 cm^2. A second compartment 32 was separated from the first compartment by using a bipolar membrane, in which the anion exchange layer was facing

the anode and the cation exchange layer was attached to the cathode as described previously. The second compartment, which was in contact with the cathode, was filled with either CO_2 gas (99.995% pure) or 0.1 M $KHCO_3$ (99.995% pure) solution that was continuously purged with CO_2 gas. In all cases, the CO_2 gas flow was 4 mL/min. An Ag/AgCl (Radiometer) reference electrode 21 was installed at the anodic compartment 31 via a capillary 21a that was positioned at approximately 1 mm distance from the anion exchange layer 51 of the bipolar membrane 50. The potential between this reference electrode and the cathode was controlled using a potentiostat 24 (Princeton Applied Research), applying -1.9 V at the cathode versus the Ag/AgCl reference electrode, which corresponds to -0.9 V versus reversible hydrogen electrode (RHE). The current was measured with the same potentiostat. The effluent gas composition from the cathodic compartment 32 was measured using gas chromatography (GC, ThermoScientific) and is vented after measuring. The setup is illustrated in Figure 4.

The temperature in these experiments was 25 (±2)°C, and the pressure was near atmospheric (100 kPa)(strictly speaking a slight overpressure to circulate the gas flow).

For comparison reasons, the same cell was constructed using a copper cathode of the same size that was not attached to the bipolar membrane, but at approximately 10 mm distant from the bipolar membrane. A solution of 0.1 M $KHCO_3$ was used in the cathodic compartment. All other elements were equal to the cases where the cathode was attached to the bipolar membrane.

The performance of these prototypes is indicated by the current density and the fuel production rates, in this case measured by CO and H_2 output concentrations. Figure 5 shows the current density (mA/cm^2) as a function of time (h), when applying -0.9 V vs RHE to the cathode for Cu in HCO_3^- (bottom line), for BPM MEA in HCO_3^- (middle fluent line) and for BPM MEA in CO_2 gas (middle irregular line). Figure 6 shows the concentrations for the products CO and H_2 in the effluent gas, for the same cases as shown in Figure 5.

Figure 6 shows that the yield of fuel products (H_2 and CO in parts per thousand/million, respectively) as a function of time (h) using a HCO_3^- solution is significantly

higher for the BPM MEA case (top line) and when the cathode is attached to the bipolar membrane (BPM) compared to the reference case (bottom line) when the cathode is not attached to the bipolar membrane. This higher yield is also reflected in the current density (Figure 5), which is higher for the BPM MEA in HCO_3^- compared to the Cu electrode that was not attached to the BPM.

The case in which the membrane electrode assembly (MEA) is operated in a gaseous CO_2 environment has a higher current density than the reference case and shows much higher yield for H_2 as well. The CO yield is lower than for the other cases, but slightly increases over time, whereas the production rate for the reference case decreases over time. Therefore, it is found that operation in gaseous environment also has a higher yield in the long term than the reference case. Moreover, the reference case, where the Cu is not attached to the membrane, cannot operate in a gaseous environment at all, because a closed electrical circuit cannot be realized without the use of water in the reference case.

The invention is further detailed by the accompanying figures, which are exemplary and explanatory of nature and are not limiting the scope of the invention. To the person skilled in the art it may be clear that many variants, being obvious or not, may be conceivable falling within the scope of protection, defined by the present claims.

FIGURES

The invention although described in detailed explanatory context may be best understood in conjunction with the accompanying figures.

Fig. 1 shows an illustration of bipolar membrane with embedded cathode.

Figure 2: System design with bipolar membrane and embedded electrode in tubular shape and a photovoltaic (PV)-panel at the back side.

Figure 3: a method for attaching cathode to bipolar membrane. CEL and AEL represent the cation exchange layer and anion exchange layer, respectively.

Figure 4: illustration of experimental setup. The anion exchange layer 51 and cation exchange layer 52 are together referred as the bipolar membrane 50.

Figure 5: current density as a function of time, at fixed cathode potential of -0.9 V vs RHE, for the bipolar membrane with attached Cu electrode (BPM MEA) in CO_2 gas, in 0.1 M HCO_3^-, and a Cu electrode (in 0.1 M HCO_3^-) that was not attached to the bipolar membrane as a reference case.

Figure 6: fuel production rate, indicated by the H_2 and CO product concentrations in the effluent gas, for the same cases as in figure 3.

DETAILED DESCRIPTION OF THE FIGURES

In the figures:

100	water splitting device
21	reference electrode
21a	capillary
24	potentiostat
31	first compartment
32	second compartment
40	power source
50	bipolar membrane
51	anion exchange layer (AEL)
52	cation exchange layer (CEL)
60	second electrode (cathode)
70	first electrode (anode)
80	tube
91	first solar system
92	second solar system

Fig. 1 shows an illustration of a system 100 having a bipolar membrane with embedded cathode. Therein a bipolar membrane 50, having an anion exchange layer (AEL) 51 and a cation exchange layer (CEL) 52, a cathode 60 attached to the CEL, a first compartment 31 adjacent to the AEL, a second compartment 32 adjacent to the cathode, and an anode 70 adjacent to the first compartment. It is used for CO_2 reduction (in gas phase) to CO and H_2 (syngas). The same system could be applied for water splitting (i.e., only H_2 and O_2 evolution). Also an aqueous phase at both sides of the membranes is possible.

In figure 1A (left side) a typical system set-up is shown. On a top side a power source is indicated, connected to the anode (left) and cathode (right). In the aqueous left side oxygen is generated (top left) and removed from the sys-

tem. The membrane allows passage of hydrogen. On a right side carbon dioxide (top right) is provided, and syngas and water are generated.

In figure 1B (right side) a section of fig. 1A is enlarged. Therein the aqueous solution, comprising K^+ and OH^-, the AEL, the CEL, the cathode, and the gas phase are shown) from left to right). In the gas phase further H_2 is generated.

Figure 2: System design with a bipolar membrane and embedded electrode in a tubular shape and an optional photovoltaic (PV)-panel at the back side. Light is absorbed by the photo anode and remaining light is absorbed by the PV-panel. This drives a redox reaction, in this case of CO_2 reduction and H_2 evolution. Combination with an additional power supply are possible as well.

In fig.2, from the inside to the outside a tube 80 is shown, comprising a compartment 32 with a gas phase with carbon dioxide and carbon monoxide, the tube comprising an cathode 60, attached to the cathode a CEL 52, an AEL 51, the tube-structure being in an aqueous solution 31, comprising water. In the aqueous solution an anode 70 is provided. In an example the anode is connected to a (first) solar system 91, whereas, as an alternative or in combination, a second solar system 92 is provided in contact with the cathode. As such use can be made of light for generating a chemical fuel.

The figures have been detailed throughout the description.

CLAIMS

1. System (100) for producing a chemical fuel, comprising a stack like structure, the stack comprising
a first electrode (70), typically an anode,
adjacent to the first electrode a first compartment (31), the compartment comprising a first fluid for oxidation,
adjacent to the first compartment or first electrode, a double layer structure (50), the double layer acting as a gas barrier, having
an anion exchange layer (51) at an anode side and in anion exchange contact with the first fluid, and
a cation exchange layer (52) adjacent to the anion exchange layer,
a second electrode (60) attached to said cation exchange layer, typically a cathode,
adjacent to the second electrode a second compartment (32) for reduction, the compartment comprising a second fluid for reduction in cation exchange contact with the second electrode, and
a power source (40) in electrical contact with the first and second electrode.

2. System according to claim 1, wherein the first fluid is an aqueous solution or a gas.

3. System according to any of claims 1-2, wherein the double layer (50) is a bipolar membrane.

4. System according to any of claims 1-3, wherein the second fluid is a gas.

5. System according to any of claims 1-4, wherein the first fluid provides hydrogen ions.

6. System according to any of claims 1-5, wherein the second fluid provides a carbon dioxide molecule

7. System according to any of the preceding claims, wherein the power source (40) is a PV-source (91,92), such as a PV-element attached to the first electrode (adjacent to the first liquid)

8. System according to any of the preceding claims, wherein a PV-element is attached to the second electrode.

9. System according to any of the preceding claims, wherein the double layer (50) is curved, such as a tube,

wherein the anion exchange layer (51) is on a convex side of the double layer and the cation exchange layer (52) is on a concave side of the double layer.

10. System according to claim 9, wherein the concave side is filled with a porous material, such as with carbon.

11. System according to any of the preceding claims, wherein a first electrode (70) comprises Ni or NiFe.

12. System according to any of the preceding claims, wherein a second electrode (60) comprises at least one of Cu, Pt and Ag.

13. Method of producing a chemical fuel, comprising the steps of

providing a system (100) according to any of claims 1-12, having a suitable first fluid and a suitable second fluid,

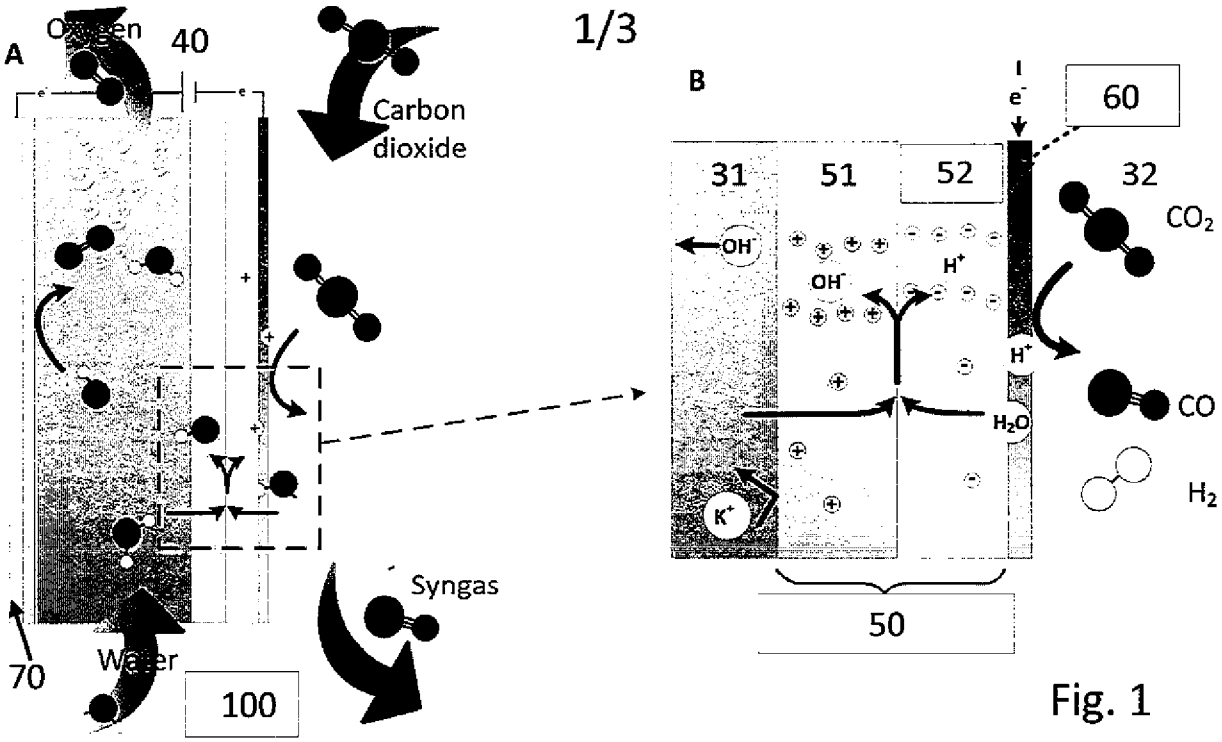
and producing a chemical fuel, the chemical fuel being selected from hydrogen, a hydrocarbon, CO, syngas, an alcohol, and combinations thereof.

14. Method according to claim 13, wherein the electric power is provided by at least one PV-system (91,92).

15. Method according to claim 13 or 14, wherein one or more of a cathode area, an anode area, a number of electrodes, a ratio between to be oxidized first fluid and to be reduced second fluid, and product selectivity, are tuned.

16. Method according to any of claims 13-15, wherein the cathode operates in acidic conditions, the pH preferably being between 1-7, more preferably 5-7.

17. Method according to any of claims 13-16, wherein the anode operates in basic conditions, the pH preferably being between 9-14.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40
Carbon dioxide
Syngas
Water
70
100
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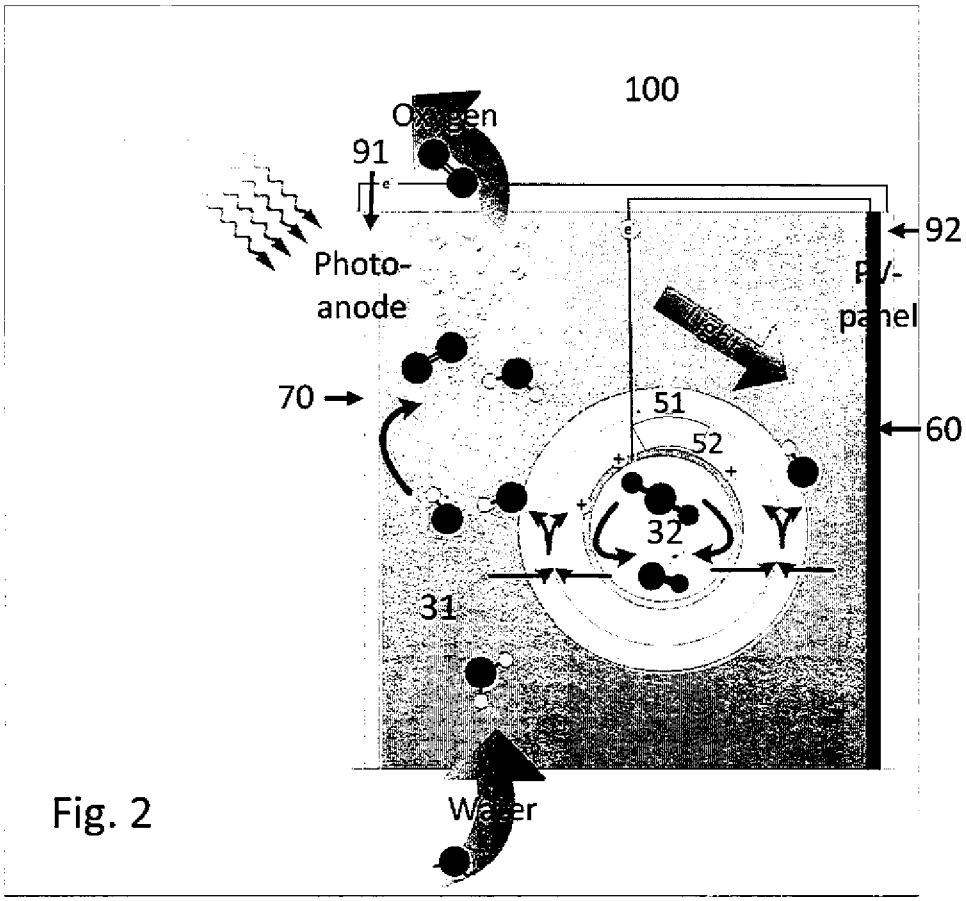
CO
H_2
K^+
50

Fig. 1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Oxygen
91
Photo-anode
92
PV-panel
70
51
52
60
32
31
Water

Fig. 2

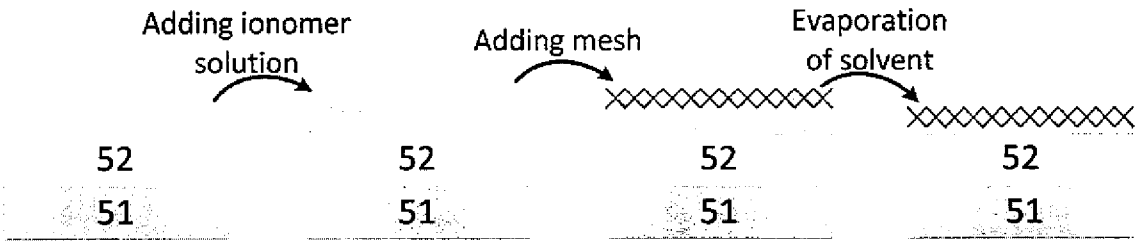

Adding ionomer solution
Adding mesh
Evaporation of solvent
52
51
52
51
52
51
52
51

Fig. 3

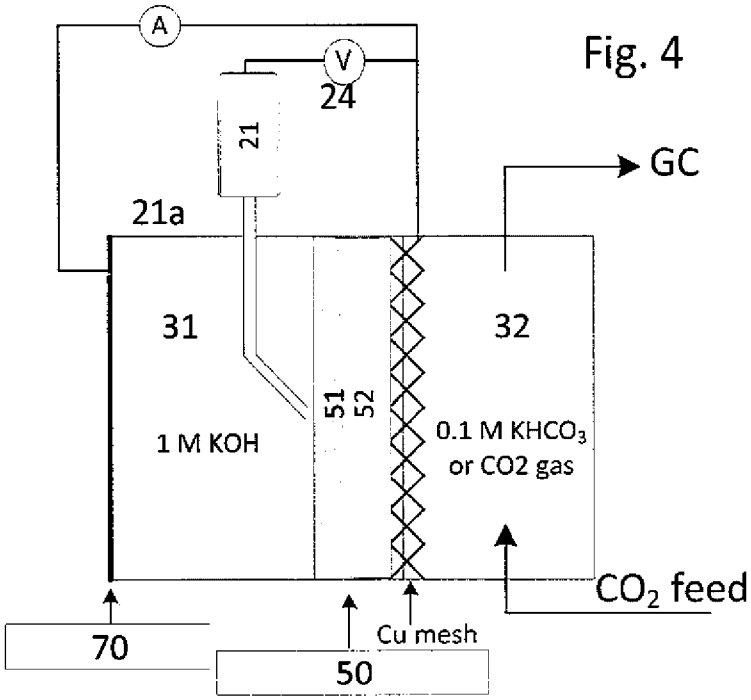

A
V
24
21
21a
GC
31
32
1 M KOH
51
52
0.1 M KHCO3
or CO2 gas
CO2 feed
70
Cu mesh
50

Fig. 4

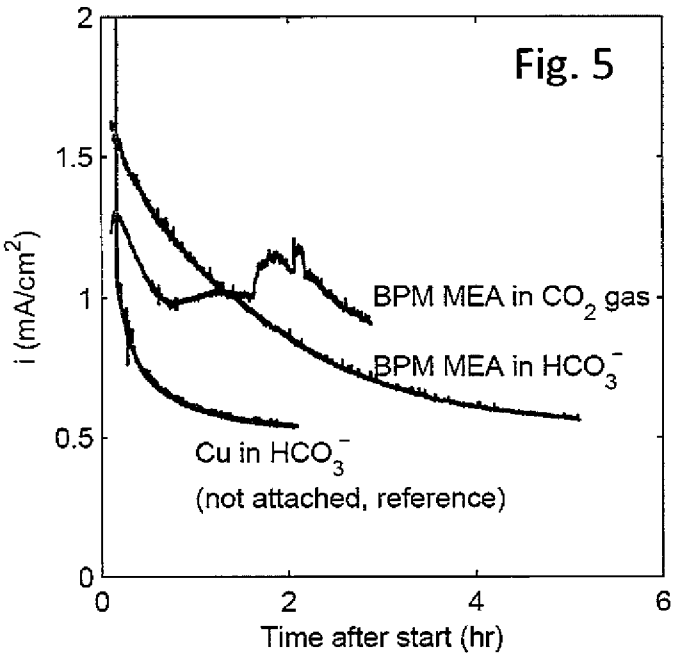
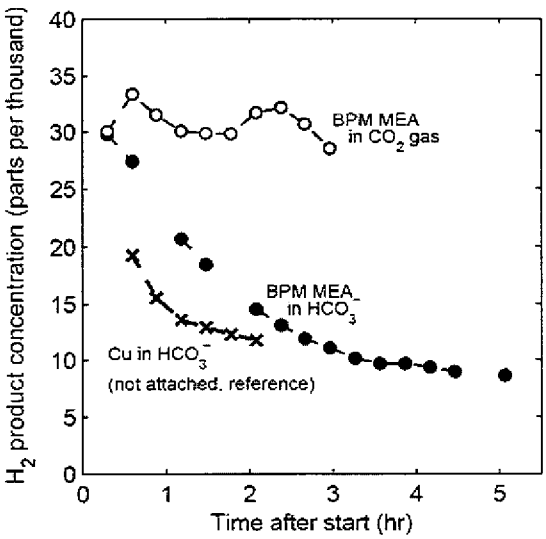


Fig. 5
i (mA/cm^2)
Time after start (hr)
BPM MEA in CO_2 gas
BPM MEA in HCO_3^-
Cu in HCO_3^-
(not attached, reference)


H_2 product concentration (parts per thousand)
Time after start (hr)
BPM MEA in CO_2 gas
BPM MEA in HCO_3^-
Cu in HCO_3^-
(not attached. reference)

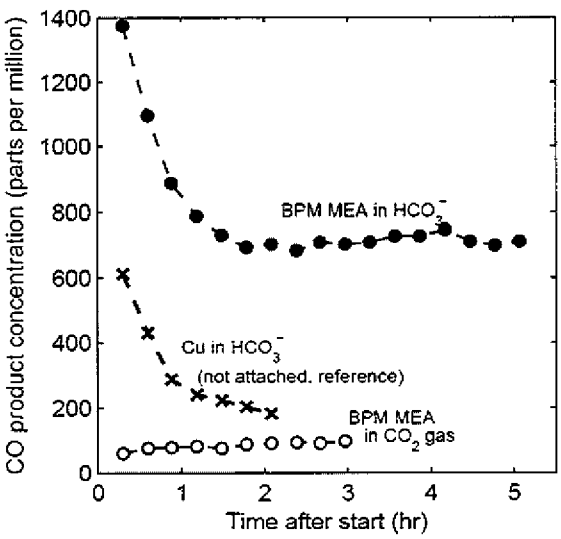

CO product concentration (parts per million)
Time after start (hr)
BPM MEA in HCO_3^-
Cu in HCO_3^-
(not attached. reference)
BPM MEA in CO_2 gas

Fig. 6

INTERNATIONAL SEARCH REPORT

International application No
PCT/NL2016/050189

A. CLASSIFICATION OF SUBJECT MATTER
INV. C25B13/04 C25B1/10 C25B9/10 C25B1/00
ADD.

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

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
C25B

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

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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Y	claim 6 paragraphs [0010] - [0013], [0057], [0063], [0066]	6-10,14

Y	EP 0 459 820 A2 (TOKUYAMA SODA KK [JP]) 4 December 1991 (1991-12-04) claims 1-4,6 examples 1-7 page 2, lines 14,52-57 page 6, lines 52-56	1-17

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[X] Further documents are listed in the continuation of Box C.

[X] See patent family annex.

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Date of the actual completion of the international search	Date of mailing of the international search report
27 July 2016	09/08/2016

Name and mailing address of the ISA/	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016	Perednis, Dainius

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INTERNATIONAL SEARCH REPORT

International application No
PCT/NL2016/050189

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

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Y	US 2007/023290 A1 (HAWKINS JOHN [US] ET AL) 1 February 2007 (2007-02-01) claims 1,5 examples 1-6 pages 1,6,; figures 1,6,12 paragraphs [0039], [0071]	1-17
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		WO 2014114806 A1	31-07-2014