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|------|--|---|
| (71) | Applicant: Siemens Energy Global GmbH & Co. KG 81739 München (DE) | Pastusiak, Remigiusz 81549 München (DE) Tawil, Angelika 80935 München (DE) |

(54) BIPOLAR MULTILAYER ELECTRODE DESIGN FOR STABLE ELECTROCHEMICAL REDUCTION OF CO2 TO HYDROCARBONS

(57)The present invention pertains to a multilayer electrode for CO₂ electrolysis, in particular a bipolar multilayer electrode for stable electrochemical reduction of CO₂ to hydrocarbons as well as a corresponding electrochemical cell. Accordingly, a multilayer electrode (10) for CO₂ electrolysis is suggested, comprising a gas diffusion layer (12) with a predefined pore size adapted for CO₂ diffusion, a catalyst layer (14) adjacent to the gas diffusion layer (12) and comprising a copper-based cathode catalyst, and a conductive layer (16) adjacent to the catalyst layer (14), wherein the gas diffusion layer (12), the catalyst layer (14), and the conductive layer (16) together form a gas diffusion electrode, and wherein the catalyst layer (14) comprises a predefined amount of anion exchange ionomer (18) and the conductive layer (16) comprises at least one layer comprising a predefined amount of cation exchange ionomer (24) so as to form a bipolar gas diffusion electrode.



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Description

Technical Field

[0001] The invention relates to a multilayer electrode for CO_2 electrolysis, in particular a bipolar multilayer electrode for stable electrochemical reduction of CO_2 to hydrocarbons as well as a corresponding electrochemical cell.

Technological Background

[0002] Electrochemical carbon dioxide (CO_2) reduction reaction (CO_2RR) to hydrocarbons poses a promising alternative to other energy storage strategies in the future. The conversion of CO_2 by electrochemistry is an attractive means by which renewable electricity, such as solar energy and wind energy, can be used and CO_2 can be bound as a product. However, it has been a challenge to develop systems that are sufficiently selective, efficient and stable, in particular in view of selective hydrocarbon formation.

[0003] In this regard, catalysts for CO₂ reduction reaction can particularly suffer from poor stability of the desired products' formation. From the group of potential catalysts including e.g. copper (Cu), silver (Ag), gold (Au), palladium (Pd), tin (Sn), copper is the only transition metal catalyst for CO₂RR to value added C₂+ products, such as ethylene, ethanol, or propanol. Such copperbased catalysts are commonly used on the cathode side of the electrochemical cells, wherein a typical electrochemical cell generally consists of a gap for the gas feedin, a cathode gas diffusion electrode including the copper-based catalyst, a separator membrane and an anode gas diffusion electrode. There can be another gap for an electrolyte between the membrane and the cathode electrode, as well as between the anode electrode and e.g. an anode end plate. In the cathode, CO₂ may be reduced to a variety of compounds, including carbon monoxide, formate, ethylene, ethanol, propanol and some other minor products, such as methane or allyl alcohol. However, undesired side reactions typically occur at the cathode, including a hydrogen evolving reaction (HER). In this regard, the thermodynamical equilibrium potentials versus standard hydrogen of the abovementioned reactions unfortunately suggest that the CO₂ reduction to CO₂RR products and the occurrence of hydrogen formation are thermodynamically favored in the same and very narrow potential range. This situation creates a significant challenge to an aimed product selectivity, e.g. to primarily obtain the value-added product ethylene. While there have been ongoing efforts for modifying the catalyst and the electrode to control the product selectivity e.g. towards ethylene, up until today there has been no breakthrough in CO₂ reduction to hydrocarbons which can be applied in industry scale to provide a selective operation towards specific product(s) in longer durations (e.g. >10000h).

[0004] In order to enable a CO_2 -rich gas-phase environment at the cathode from the gas side a gas diffusion electrode (GDE) is typically provided, wherein the gas diffusion electrode comprises a porous gas diffusion layer and an active catalyst layer. To provide an efficient CO_2 reduction reaction, this facilitates the formation of three-phase boundaries consisting of gaseous CO_2 , a solid copper-based catalyst, and water with an existing electrical potential as the driving force for electrochemi-

cal reaction. However, current copper-based electrodes or catalysts are not stable in an industrial relevant long-term duration (e.g. >10000 h). Depending e.g. on the electrode, the catalyst and operation conditions, faradaic efficiencies for CO₂ reduction reactions decrease and the
 hydrogen evolving reaction is favored already after 1 to

hydrogen evolving reaction is favored already after 1 to 200 hours.

[0005] The loss of stability may e.g. be caused by a surface restructuring of the catalyst, a flooding of the gas diffusion electrode blocking the gas CO_2 feed diffusion, and/or the accumulation of impurities or salts resulting in

20 and/or the accumulation of impurities or salts resulting in a functional loss of the electrode. Furthermore, a loss of selectivity may be caused e.g. by permeation of the electrolyte into the pores of the gas diffusion electrode, thereby disrupting the three-phase boundary and block-²⁵ ing the pores available for CO₂ transport and/or local

ing the pores available for CO₂ transport and/or local catalytically active sites for CO₂ reduction. **[0006]** While there have been efforts to improve the stability and selectivity of the CO₂ reduction, most developments have been shown to result in undesirable salt

³⁰ formation and/or are only suitable for small-scale or nano-scale conditions. Such conditions, e.g. requiring strong alkaline conditions (e.g. pH>13) or acidic conditions (e.g. pH<1) have been found not to be suitable and/or scalable to an industrial level.

³⁵ **[0007]** Accordingly, a need exists to provide an improved selectivity and long-term stability of CO₂ reduction to hydrocarbons.

Summary of the invention

[0008] It is an object of the present invention to provide an improved electrode design for CO_2 reduction. In particular, it may be an object to provide an improved reduction reaction selectivity and stability of such electrode.

⁴⁵ [0009] Accordingly, in a first aspect, a multilayer electrode for CO₂ electrolysis is suggested, comprising a gas diffusion layer with a predefined pore size adapted for CO₂ diffusion, a catalyst layer adjacent to the gas diffusion layer and comprising a copper-based cathode cat-

⁵⁰ alyst, and a conductive layer adjacent to the catalyst layer. The gas diffusion layer, the catalyst layer, and the conductive layer together form a gas diffusion electrode, wherein the catalyst layer comprises a predefined amount of anion exchange ionomer and the conductive

⁵⁵ layer comprises at least one layer comprising a predefined amount of cation exchange ionomer so as to form a bipolar gas diffusion electrode.

[0010] The inventors have found that the provision of a

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bipolar gas diffusion electrode by means of the anion exchange ionomers and the cation exchange ionomers is particularly advantageous to enable the desired selectivity and stability of the CO₂ reduction reactions. In particular, when implemented in an electrochemical cell, the use of cation exchange ionomers in the conductive layer of the electrode, which preferably is formed as an outer layer, enables that OH-anions being generated in the active catalyst layer are locally enriched therein, by limiting and/or slowing down their transportation away from the active catalyst layer towards the anode in the direction of electroosmosis. The enrichment of OH-anions in the active catalyst layer provides a more alkaline environment due to the corresponding higher local pH, which was found to be beneficial for the desired C-C coupling reaction mechanism yielding ethylene (C₂H₄), for example, as a product.

[0011] These advantageous effects may be further increased by the optional combination with electrically conductive polymers, e.g. polyacetylene, polyphenylene vinylene, polypyrrole, polythiophene, polyaniline, or polyphenylene sulfide, and/or with highly hydrophobic polymers, such as polytetrafluoroethylene (PTFE), polyviny-lidene fluoride (PVDF), polypropylene (PP), or polyethylene (PE). Alternatively, the cation exchange ionomer and/or the anion exchange ionomer may be replaced by one or more of these polymers, in particular by a highly hydrophobic polymer.

[0012] Furthermore, the use of these cation exchange ionomers specifically or exclusively in the conductive layer of the electrode impairs the carbonate anion availability in the adjacent catalyst layer. In this regard, the use of anion exchange ionomers in the active catalyst layer, which preferably forms an inner layer of the electrode, limits the availability of potassium ions (K+) inside this layer. As a result, in particular in the active catalyst layer and in the gas diffusion layer, the coexistence of potassium ions and carbonate anions is effectively avoided.

[0013] Thereby, the occurrence or likelihood of typical salt formation resulting from the coexistence of potassium ions and carbonate anions in the active catalyst layer and in the gas diffusion layer is significantly reduced. By reducing the salt formation, undesirable flooding of the gas diffusion layer may be prevented or at least reduced to the largest possible extent, such that a durable and efficient three-phase boundary with the gaseous CO₂ may be ensured.

[0014] In addition, while the implementation of both anionic and cationic exchange ionomers ensures that the required level of permeance of anions or cations may be provided, it also optimizes the hydrophobicity of the respective layer as well as their water uptake capacity due to their swelling nature. This is because the ion exchange materials are preferably formed of a hydrophobic polymer backbone with a hydrophilic ion exchange functional group. Accordingly, the provision of sufficient three-phase boundaries may be further ensured by means of the corresponding hydrophobicity and e.g. a reduced water availability within pores of the gas diffusion layer. The electrode design according to the invention hence provides a synergistic solution to the current problems of poor selectivity and short stability.

- ⁵ The invention enables a selective permeability, a control of the local pH level, an effective barrier e.g. for saltforming carbonate and potassium compounds, and a well-balanced hydrophobicity to facilitate the availability of three-phase boundaries. Thereby, the selective pro-
- 10 duction of e.g. ethylene, ethanol, and/or propanol may be significantly improved for much longer durations, such that the production may be brought to a high technology readiness level (TRL).

[0015] The multilayer configuration of the bipolar gas diffusion electrode may be considered as a stack of layers, such that gaseous CO₂, which is preferably wet or humidified, may pass through the gas diffusion layer first before reaching the catalyst layer. The catalyst layer is copper-based and is preferably formed of copper-oxide

- 20 (CuO). The conductive layer in the stack is accordingly arranged at a side of the catalyst layer opposing the side of the catalyst layer being adjacent to the gas diffusion layer.
- **[0016]** The amount of anion exchange ionomer and cation exchange ionomer may be chosen to provide a predefined hydrophobicity of the overall gas diffusion electrode, wherein the respective ionomers may be provided within the respective layer as a single layer, multiple layers, or as a gradient.

30 [0017] The ionomers may furthermore also be provided as a corresponding membrane, i.e. an anion exchange membrane or cation exchange membrane, for example to define a respective layer that may be simply applied to another layer of the multilayer electrode. How-

- ³⁵ ever, the ionomer is preferably integrated or embedded in a respective layer, e.g. using ink drop application. This has the advantage that the respective ionomer may at least partially penetrate the respective layer and the availability thereof may be improved. Furthermore, this
- ⁴⁰ allows a very specific dosing of the respective ionomer, which is particularly advantageous since their loading in the respective layer is a very sensitive parameter. For example, an excess amount of the respective ionomer may block the active catalyst sites by creating a thick

⁴⁵ diffusion layer whereas a suboptimal amount may limit the extents of the necessary three phase boundaries.
[0018] Accordingly, the weight percent of anion exchange ionomer is preferably between 0.01 wt. % and 20 wt. % and the weight percent of cation exchange
⁵⁰ ionomer in the respective layer is preferably between 0.01 wt. % and 20 wt. %. In particular, the weight percent of anion exchange ionomer may be between 1 wt. % and 15 wt. %.

[0019] The above percentage ranges for the anion exchange ionomer and the cation exchange ionomer, in particular the specific weight percent ranges of the cation exchange ionomer, have been found to be particularly advantageous in terms of controlling the local pH,

defining the overall hydrophobicity, and the possibility of blocking potentially salt-forming components. Thereby, flooding of the gas diffusion layer could be advantageously avoided and the CO₂ reduction reactions resulting e.g. in ethylene could be favored. Accordingly, the above preferred percentages enabled a significant improvement in view of the selectivity and durability or stability of the CO₂ reduction provided by the multilayer electrode.

[0020] To avoid that the application of the anion exchange ionomers results in a blocking of the pores of the gas diffusion layer, the anion exchange ionomer is preferably arranged at a side of the catalyst layer opposing the gas diffusion layer. In other words, the anion exchange ionomers are preferably located at a side adjacent to the conductive layer. To facilitate such arrangement, the anion exchange ionomers are preferably applied by ink drop application, e.g. as layer-by-layer preparation. However, alternative or additional methods may be implemented, such as drop casting, air brushing, spray coating, decal process, dry or wet calendaring, physical vapor deposition, or chemical vapor deposition. [0021] Preferably, the conductive layer comprises a graphite layer comprising a predefined portion of the cation exchange ionomer.

[0022] The implementation of a graphite layer, which is preferably formed as an outer layer of the gas diffusion electrode, provides improved conductivity and corresponding electron transfer. This is due to the typically hexagonal structure of graphite, wherein a free valence electron is available to facilitate electron transfer. By including a predefined portion of the cation exchange ionomer in the graphite layer a balance is hence provided between, on the one hand, the improved conductivity towards the catalyst layer and the local pH control, reduction of salt formation, and the formation and/or accessibility of the three-phase boundary layer, on the other hand.

[0023] The hexagonal structure of the graphite is furthermore advantageous to control the density of said layer. Accordingly, the graphite layer preferably has a density of between 1 mg/cm² and 20 mg/cm², preferably between 5 mg/cm² and 10 mg/cm² Thereby, it has been found that an optimal tradeoff may be achieved, in particular with the more specific density range, with regard to the required structural stability, the porosity for the cation exchange ionomer, and the conductivity.

[0024] In addition, the conductive layer may also comprise a carbon nanoparticle layer comprising a predefined portion of the cation exchange ionomer and being arranged between the graphite layer and the catalyst layer.

[0025] The implementation of an additional carbon nanoparticle layer, e.g. as the outer or outermost layer of the gas diffusion layer, has the advantage that an improved surface area for the cation exchange ionomer may be provided. The carbon nanoparticles may further stabilize the copper so as to inhibit or prevent it from

bleaching under electroreductive conditions. As for the graphite layer, the predefined portion of the cation exchange ionomer may be between 0.01 - 20 wt. % to further support the selective CO_2 reduction reaction and improve the stability of the bipolar gas diffusion electrode.

[0026] The density of the carbon nanoparticle layer is preferably between 0.05 mg/cm² and 2 mg/cm². Compared with the graphite layer, such relative lower density

10 has been found to be advantageous in view of the function of the cation exchange ionomer and/or to enable a predefined level of swelling due to the corresponding hydrophobicity of the cation exchange ionomer.

[0027] In particular, the weight percent of the cation
exchange ionomer may be between 0.01 wt. % and 20 wt.
%, e.g. between 1 wt. % and 10 wt. %, wherein each of the preferred graphite and carbon nanoparticle layers may comprise essentially the same weight percent of the cation exchange ionomer. Such weight percent may
result in an overall amount of cation exchange ionomer essentially corresponding to the overall amount of anion exchange ionomer present in the catalyst layer. In this regard, the dimensioning and/or weight of the catalyst layer and the graphite and carbon nanoparticle layers

[0028] The conductive layer and/or the catalyst layer may also comprise a hydrophobic agent. Such hydrophobic agent may be present in addition to the respective cation exchange ionomer and anion exchange ionomer

and may e.g. provide an additional hydrophobicity to achieve an overall predefined hydrophobicity without significantly affecting the bipolarity of the gas diffusion electrode. Preferred hydrophobic agents may comprise or essentially consist of hydrophobic polymers, such as
 polytetrafluoroethylene (PTFE), polyvinylidene fluoride

(PVDF), polypropylene (PP), or polyethylene (PE). **[0029]** Furthermore, the multilayer bipolar gas diffusion electrode may comprise a protective layer and/or current collector, which is arranged adjacent to the con-

⁴⁰ ductive layer at a side opposing the catalyst layer and/or at least partially surrounding the conductive layer.
 [0030] Said additional layer may e.g. be formed of a metal or metal and polymer mix and is preferably formed

- as a grid or cage structure. Thereby, it may be facilitated
 that the gas diffusion electrode is secured in place and/or any currents applied or being transferred to the gas diffusion electrode may be effectively collected. The protective layer and/or current collector is hence preferably arranged so as to be in direct contact with an electrolyte present e.g. in an electrolyte flow chamber, when the bipolar gas diffusion electrode is implemented in an electrochemical cell for CO₂ electrolysis or reduction.
- [0031] According to a further aspect of the invention, an electrochemical cell for CO₂ electrolysis is suggested, comprising a multilayer electrode according to the invention. The electrochemical cell preferably comprises an anode catalyst layer, a separator adjacent to the anode

catalyst layer and an electrolyte flow chamber arranged between the multilayer electrode and the separator.

[0032] The electrochemical cell may comprise a gas feed chamber upstream of and adjacent to the gas diffusion layer for wet or humidified CO_2 to permeate or to be conveyed to the gas diffusion layer in a predefined and controlled manner. Gaseous CO_2 reduction reaction products may also be enriched or be transferred to said gas feed chamber. At the other side of the gas diffusion electrode, electrolyte may be provided in an electrolyte flow chamber. Via said flow chamber it is also enabled that liquid CO_2 reduction reaction products may be collected.

[0033] Preferred electrolyte solutions include potassium hydroxide, potassium (bi)carbonate, cesium bicarbonate, potassium sulfate, calcium bicarbonate, sodium bicarbonate, lithium bicarbonate, ionic liquids, or solid electrolytes.

[0034] At the anode side, i.e. at the anode catalyst layer, which is separated by a separator, e.g. a cation or anion exchange membrane, water is oxidized to oxygen and hydrogen/hydronium ions or hydroxide ions, depending on the employed membrane.

[0035] By means of the implementation of the bipolar gas diffusion electrode according to the invention, the electrochemical cell hence enables a CO_2 reduction reaction to selectively and stably produce CO_2 reduction reaction products, in particular of ethylene, ethanol, and propanol, as will be shown in view of the examples and Figures in the following.

Brief description of the drawings

[0036] The present disclosure will be more readily appreciated by reference to the following detailed description when being considered in connection with the accompanying drawings in which:

Figure 1 is a schematic view of a multilayer bipolar gas diffusion electrode according to the invention;

Figure 2 is a schematic view of an embodiment of an electrochemical cell according to the invention;

Figure 3 shows results of faradaic efficiencies for different products obtained with the bipolar gas diffusion electrode over time;

Figure 4 shows more detailed faradaic efficiencies for different products obtained with the bipolar gas diffusion electrode after 24 hours; and

Figure 5 shows results of faradaic efficiencies for different products obtained with the bipolar gas diffusion electrode over time with alternative weight percentages of the anion exchange ionomer and the cation exchange ionomer compared with the embodiment according to Figure 3.

Detailed description of preferred embodiments

[0037] In the following, the invention will be explained in more detail with reference to the accompanying figures. In the Figures, like elements are denoted by identical reference numerals and repeated description there-

of may be omitted in order to avoid redundancies. [0038] In Figure 1 a schematic view of a multilayer

bipolar gas diffusion electrode 10 according to the invention is depicted. As shown, the multilayer electrode 10

¹⁰ tion is depicted. As shown, the multilayer electrode 10 comprises a gas diffusion layer 12 having a predefined pore size adapted for diffusion of CO_2 , preferably humidified CO_2 . The gas diffusion layer 12 is arranged directly adjacent to a cathode catalyst layer 14, which forms the

primary reactive layer for the reduction of CO₂ at a three-phase boundary. At the side of the cathode catalyst layer
14 opposing the gas diffusion layer 12, i.e. not being adjacent to the gas diffusion layer 12, a conductive layer
16 is present, wherein the gas diffusion layer 12, the
cathode catalyst layer 14, and the conductive layer 16

together form the gas diffusion electrode 10.
[0039] Also depicted at the side of the cathode catalyst layer 14 opposing the gas diffusion layer 12 is a layer comprising anion exchange ionomers 18. While said
²⁵ anion exchange ionomers are depicted as a separate layer, which may e.g. be formed using an ink drop application, it is to be understood that said anion exchange ionomers 18 may also be distributed throughout the cathode catalyst layer, e.g. should the cathode catalyst

³⁰ layer 14 exhibit a corresponding porosity. However, these anion exchange ionomers 18 are preferably present only at the side of the cathode catalyst layer 14 being adjacent to the conductive layer 16 and/or are not present at the side of the cathode catalyst layer 14 being adjacent to the

³⁵ gas diffusion layer 12. Thereby, it can be ensured that the pores of the gas diffusion layer 12 are not or at least not significantly affected by the application and presence of the anion exchange ionomers 18.

[0040] The conductive layer 16 according to the present example is formed of or comprises two layers, a graphite layer 20 and an optional carbon nanoparticle layer 22, as indicated with the dashed lines. Each of said layers 20, 22 comprises a predefined portion of cation exchange ionomers 24. The provision of the anion ex-

⁴⁵ change ionomers 18 in the cathode catalyst layer 14 and of the cation exchange ionomers 24 in the conductive layer 16 provides that the multilayer gas diffusion electrode 10 is bipolar. As described above, such configuration has the advantage that a significant improvement in
⁵⁰ the selectivity of the CO₂ reduction products and the stability of the corresponding reactions and the cathode catalyst layer 14 may be achieved, as will be shown in further detail in view of Figures 3, 4, and 5 below.

[0041] In the embodiment depicted in Figure 1 a protective layer and/or current collecting layer 26 is proved adjacent to the outer layer of the multilayer electrode 10, i.e. adjacent to the conductive layer. Such layer 26, which is optional as indicated with the dashed lines, may e.g. be

formed as a metal grid or cage-like structure, which preferably protects the conductive layer 16 and functions as an efficient means for current collection.

[0042] Although a spacing is present between some of the adjacent layers, such spacing is merely for illustrative purposes to identify the respective layers and it is to be understood that the layers in an actual, practical are in direct contact with each other.

[0043] In Figure 2 a schematic view of an embodiment of an electrochemical cell 28 is depicted, wherein the multilayer electrode 10 according to the invention has been implemented. Accordingly, the multilayer electrode 10 forms a cathode, wherein humidified CO₂ may be conveyed to its gas diffusion layer 12 by means of an adjacent gas feed chamber 30. At the opposing side of the multilayer electrode 10, an electrolyte flow chamber 32 is present, which provides a liquid barrier between the multilayer electrode 10 and an anode catalyst layer 34 and an adjacent anode gap 38. Between the multilayer electrode 10 and the anode catalyst layer 34, a separator 36, such as a cation exchange membrane, an anion exchange membrane or a diaphragm membrane is furthermore present, which is in direct contact with the electrolyte present in the electrolyte flow chamber 32 and is arranged adjacent to the anode catalyst layer 34. Adjacent to the anode gap 38 on a side opposing the anode catalyst layer 34, an anolyte flow chamber (not shown) may be present, which is flowing by the anode catalyst 38.

[0044] Upon applying a voltage, CO_2 is reduced at the multilayer electrode 10, wherein gaseous CO_2 reduction products may be collected at the side of the gas feed chamber 30 and liquid CO_2 reduction products may be collected at the side of the electrolyte flow chamber.

[0045] Figure 3 shows the electrochemical performance of a copper-based electrode according to the invention at a current density of 100 mA/cm² in a CO₂ electrolysis flow cell with a 10 cm² active cell area. For these results, a configuration of the multilayer electrode 10 has been used, wherein a conductive layer 16 including a carbon nanoparticle layer 22 and a graphite layer 20 comprising the cation exchange ionomer 24 has been implemented. As shown by the triangular data points, which represent the production of ethylene (C_2H_4) , the inventive configuration of the cathode electrode provides a significant improvement in the CO2 reduction by ensuring, for the first time in this R&D field, a stable reaction longer than 720 hours while a large proportion of e.g. value-added ethylene may be selectively provided. In particular, compared with carbon monoxide (indicated by the dots) or undesired side-reaction products such as hydrogen (indicated by the squares), the relative amount of ethylene being produced remains stable, as indicated by the faradaic efficiency (FE) percentage, being between about 20 and 25 percent. Accordingly, this achieved performance takes the copper-based CO₂ electrolysis to another level of technology readiness by prolonging the stable operation duration to months scale from days scale.

[0046] Figure 4 shows more detailed faradaic efficiencies for different products obtained with the bipolar gas diffusion electrode after 24 hours, wherein the same configuration of the multilayer electrode 10 has been implemented as in the embodiment according to Figure 3. In Figure 4, the full product spectrum obtained in copper-based CO_2 electrochemical reduction at 300 mA/cm² is depicted in a stacked chart. From bottom to

10 top the cumulative percentages are shown for ethylene, acetate, ethanol, propanol, propionaldehyde, propionate, carbon monoxide, formate, and hydrogen. Accordingly, from ethylene to propionate, a faradaic efficiency of about 50 percent could be achieved for C_2 + products with

¹⁵ more than 30 percent being attributed to ethylene. Hence, both the durability and stability of the reduction reactions of CO_2 and the selectivity of said reactions to desired products have been significantly improved.

[0047] The results depicted in Figure 5 have been obtained using the same testing conditions as for Figure 3. However, compared with the embodiment used to obtain the results depicted in Figure 3 and within the preferred weight percentages of between 0.01 wt. % and 20 wt. %, a lower weight percentage of anion exchange and a higher weight percentage of cation exchange ionomer has been used in the corresponding layers. As shown, the faradaic efficiencies for different products indicate that a similar faradaic efficiency (FE)

percentage may be achieved for ethylene (indicated by
the triangles), i.e. between about 20 and 25 percent.
Accordingly, similar to the results indicated in Figure 3,
by optimizing the different ratios of the preferred weight
percentages, a stable reaction may be provided longer
than 720 hours while a large proportion of e.g. valueadded ethylene may be selectively produced.

[0048] It will be obvious for a person skilled in the art that these embodiments and items only depict examples of a plurality of possibilities. Hence, the embodiments shown here should not be understood to form a limitation
40 of these features and configurations. Any possible combination and configuration of the described features can be chosen according to the scope of the invention.

List of reference numerals

[0049]

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- 10 Multilayer electrode
- 12 Gas diffusion layer
- 14 Cathode catalyst layer
- 16 Conductive layer
- 18 Anion exchange ionomer
- 20 Graphite layer
- 22 Carbon nanoparticle layer
- 24 Cation exchange ionomer
- 26 Protective layer and/or current collecting layer
- 28 Electrochemical cell
- 30 Gas feed chamber

- 32 Electrolyte flow chamber
- 34 Anode catalyst layer
- 36 Separator
- 38 Anode gap

Claims

1. A multilayer electrode (10) for CO₂ electrolysis, comprising

a gas diffusion layer (12) with a predefined pore size adapted for CO_2 diffusion,

a catalyst layer (14) adjacent to the gas diffusion layer (12) and comprising a copper-based cathode catalyst, and

a conductive layer (16) adjacent to the catalyst layer (14), wherein the gas diffusion layer (12), the catalyst layer (14), and the conductive layer (16) together form a gas diffusion electrode, and wherein the catalyst layer (14) comprises a predefined amount of anion exchange ionomer (18) and the conductive layer (16) comprises at least one layer comprising a predefined amount of cation exchange ionomer (24) so as to form a bipolar gas diffusion electrode.

- The multilayer (10) electrode according to claim 1, wherein the weight percent of anion exchange ionomer (18) is between 0.01 wt. % and 20 wt. % and the weight percent of cation exchange ionomer (24) in the respective layer is between 0.01 wt. % and 20 wt. %.
- The multilayer electrode (10) according to claim 2, wherein the weight percent of anion exchange ionomer (18) is between 1 wt. % and 15 wt. %.
- The multilayer electrode (10) according to any of the preceding claims, wherein the anion exchange ionomer (18) is arranged at a side of the catalyst layer (14) opposing the gas diffusion layer (12).
- The multilayer electrode (10) according to any of the preceding claims, wherein the conductive layer (16) comprises a graphite layer (20) comprising a predefined portion of the cation exchange ionomer (24).
- The multilayer electrode (10) according to claim 5, wherein the graphite layer (20) has a density of between 1 mg/cm² and 20 mg/cm², preferably between 5 mg/cm² and 10 mg/cm².
- The multilayer electrode (10) according to claim 5 or 6, wherein the conductive layer (16) comprises a carbon nanoparticle layer (22) comprising a predefined portion of the cation exchange ionomer (24) and being arranged between the graphite layer (20) and the catalyst layer (14).

- The multilayer electrode (10) according to claim 7, wherein the carbon nanoparticle layer (22) has a density of between 0.05 mg/cm² and 2 mg/cm², preferably between 0.2 mg/cm² and 1 mg/cm².
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- The multilayer electrode (10) according to claim 7 or 8, wherein the weight percent of cation exchange ionomer (24) is essentially the same for the graphite layer (20) and the carbon nanoparticle layer (22).
- The multilayer electrode (10) according to any of claims 5 to 9, wherein the weight percent of cation exchange ionomer (24) is between 1 wt. % and 20 wt. %, preferably between 2.5 wt. % and 7.5 wt.
- **11.** The multilayer electrode (10) according to any of the preceding claims, wherein the conductive layer (16) and/or the catalyst layer (14) comprises a hydrophobic agent.
- **12.** The multilayer electrode (10) according to any of the preceding claims, further comprising a protective layer and/or current collector (26) being arranged adjacent to the conductive layer (16) at a side opposing the catalyst layer (14) and/or at least partially surrounding the conductive layer (16).
- **13.** An electrochemical cell (28) for CO₂ electrolysis, comprising a multilayer electrode (10) according to any of the preceding claims.
- 14. The electrochemical cell (28) according to claim 13, comprising an anode catalyst layer (34), a separator (36), preferably a cation exchange membrane, adjacent to the anode catalyst layer (34) and an electrolyte flow chamber (32) arranged between the multilayer electrode (10) and the separator (36).

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EUROPEAN SEARCH REPORT

Application Number

EP 23 19 2687

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