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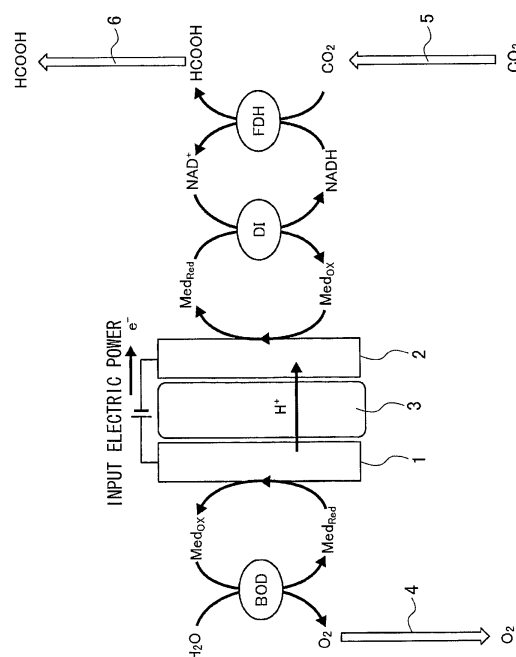
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(54) **DEVICE FOR IMMOBILIZING CARBON DIOXIDE**

(57) There is provided a carbon dioxide immobilization unit capable of easily immobilizing carbon dioxide in the form of an organic acid or a carbohydrate under a normal environment. An anode 1 and a cathode 2 both having a surface where an oxidoreductase is present are disposed to face each other with a proton conductor 3 in between. Then, when electric power is externally supplied to the carbon dioxide immobilization unit, in the anode 1, water is decomposed to produce protons, and in the cathode 2, an organic acid or a carbohydrate is produced from the protons produced in the anode 1 and carbon dioxide. At this time, while a carbon dioxide supply section 5 supplies a high concentration of carbon dioxide to the cathode 2, oxygen produced in the anode 1 and the organic acid or hydrocarbon produced in the cathode 2 are removed from a reaction system through an oxygen removal section 4 and a product recovery section 6, respectively.

[FIG. 1]



Description

TECHNICAL FIELD

[0001] The present invention relates to a carbon dioxide immobilization unit using an oxidoreductase. More specifically, the invention relates to a carbon dioxide immobilization unit producing an organic acid or a carbohydrate from carbon dioxide.

BACKGROUND ART

[0002] Biofuel cells using an oxidoreductase as a reaction catalyst have been attracting an attention as next-generation fuel cells with high capacity and high safety, since the biofuel cells effectively extract electrons from glucose, ethanol, and the like which are not usable in fuel cells using a typical industrial catalyst.

[0003] FIG. 5 is a diagram schematically illustrating an electric power generation principle of a biofuel cell using an enzyme. For example, in the case of a biofuel cell using glucose as a fuel as illustrated in FIG. 5, in an anode 101, glucose is decomposed by an enzyme immobilized on a surface thereof to extract electrons (e^-) and to produce protons (H^+). On the other hand, in a cathode 102, water (H_2O) is produced from the protons (H^+) transported from the anode 101 through a proton conductor 103, the electrons (e^-) transmitted through an external circuit, and oxygen (O_2) in, for example, air.

[0004] FIG. 6 is a diagram schematically illustrating an electric power generation principle of a methanol type biofuel cell. Moreover, as illustrated in FIG. 6, a biofuel cell using methanol as a fuel to generate electric power has been proposed in related art (for example, refer to PTL 1). In this biofuel cell, alcohol dehydrogenase (ADH), formaldehyde genase (FaIDH), and formate dehydrogenase (FateDH) are immobilized on the surface of the anode 101.

[0005] Then, in the anode 101, methanol (CH_3OH) is decomposed by these enzymes to extract electrons (e^-) and to produce protons (H^+), and then to produce carbon dioxide (CO_2). On the other hand, in the cathode 102, water (H_2O) is produced from the protons (H^+) transported from the anode 101 through the proton conductor 103, the electrons (e^-) transmitted through the external circuit, and oxygen (O_2) in, for example, air.

[0006] On the other hand, in related art, there are proposed methods of storing and producing hydrogen with use of a unit including a formic acid decomposition section and a formic acid production section (refer to PTLs 2 and 3). In this unit for formic acid production and decomposition, the formic acid production section produces formic acid through allowing hydrogen and carbon dioxide to react with each other by a catalyst for formic acid production, and then stores hydrogen in the form of formic acid.

[0007] Moreover, in the formic acid decomposition section, the formic acid produced in the formic acid produc-

tion section is decomposed into hydrogen and carbon dioxide by a catalyst for formic acid decomposition. Hydrogen produced by this decomposition reaction is used for an arbitrary purpose such as a fuel cell. On the other hand, carbon dioxide as a by-product is transmitted to the formic acid production section to be used for formic acid production.

[Citation list]

[Patent Literature]

[0008]

[PTL 1] Japanese Unexamined Patent Application Publication No. 2004-71559

[PTL 2] Japanese Unexamined Patent Application Publication No. 2009-78200

[PTL 3] Japanese Unexamined Patent Application Publication No. 2010-83730

SUMMARY OF INVENTION

[0009] However, in the techniques described in the above PTLs 2 and 3, hydrogen used as a reducing agent to produce formic acid from carbon dioxide is not stably present under a normal environment; therefore, there is an issue that extra energy is necessary to obtain hydrogen as a raw material. Thus, a user-friendly technique of immobilizing carbon dioxide in the form of formic acid or a carbon compound such as carbohydrate has not yet established.

[0010] Therefore, it is a main object of the present invention to provide a carbon dioxide immobilization unit capable of easily immobilizing carbon dioxide in the form of an organic acid or a carbohydrate under a normal environment.

[0011] A carbon dioxide immobilization unit according to the invention includes at least: a first electrode decomposing water to produce protons; a second electrode producing an organic acid or a carbohydrate from the protons produced in the first electrode and carbon dioxide; and a proton conductor transferring the protons produced in the first electrode to the second electrode, in which an oxidoreductase is present on a surface of the first electrode or a surface of the second electrode, or both.

Here and in the following description, a surface of an electrode includes an outer surface of the electrode and an inner surface of a gap in an inside of the electrode.

The carbon dioxide immobilization unit may further include a carbon dioxide supply section supplying carbon dioxide to the second electrode. In this case, the carbon dioxide supply section may supply a gas containing carbon dioxide in concentration of 0.028 to 100 vol% both inclusive.

Moreover, the carbon dioxide immobilization unit may include an oxygen removal section removing oxygen produced in the first electrode; and a product recovery sec-

tion extracting the organic acid or the carbohydrate produced in the second electrode.

Further, the first electrode may be a dipping type electrode which is directly in contact with a liquid phase or is in contact with the liquid phase with a separator in between, and the second electrode may be a semi-dipping type electrode which is directly in contact with the liquid phase or is in contact with the liquid phase with a separator in between, as well as is in contact with a vapor phase with a gas-liquid separator film in between.

Furthermore, the first electrode or the second electrode, or both may be formed of, for example, a conductive porous material.

[0012] According to the present invention, as the oxidoreductase is used, carbon dioxide is allowed to be easily immobilized in the form of an organic acid or a carbohydrate through only inputting electric power to the carbon dioxide immobilization unit without using hydrogen.

BRIEF DESCRIPTION OF THE DRAWINGS

[0013]

[FIG. 1] FIG. 1 is a diagram schematically illustrating a principle of a carbon dioxide immobilization unit according to an embodiment of the invention.

[FIG. 2] FIG. 2 is a diagram schematically illustrating an electrode configuration of an anode 1 illustrated in FIG. 1 which is of a dipping type.

[FIG. 3] FIG. 3 is a diagram schematically illustrating an electrode configuration of a cathode 2 illustrated in FIG. 1 which is of a semi-dipping type.

[FIG. 4] FIG. 4 is a diagram schematically illustrating a principle of a carbon dioxide immobilization unit according to a modification example of the above-described embodiment of the invention.

[FIG. 5] FIG. 5 is a diagram schematically illustrating an electric power generation principle of a biofuel cell using an enzyme.

[FIG. 6] FIG. 6 is a diagram schematically illustrating an electric power generation principle of a methanol type biofuel cell.

DESCRIPTION OF EMBODIMENTS

[0014] An embodiment of the present invention will be described in detail below referring to the accompanying drawings.

It is to be noted that the present invention is not limited to the following embodiment. Moreover, description will be given in the following order.

1. Embodiment

[0015] (An example of a carbon dioxide immobilization unit producing formic acid from carbon dioxide)

2. Modification Example

[0016] (An example of a carbon dioxide immobilization unit producing methanol from carbon dioxide)

(1. First Embodiment)

[Entire configuration of cell]

[0017] FIG. 1 is a diagram schematically illustrating a carbon dioxide immobilization unit according to an embodiment of the invention. Moreover, FIG. 2 is a diagram schematically illustrating an electrode configuration of an anode 1 (a first electrode) which is of a dipping type, and FIG. 3 is a diagram schematically illustrating an electrode configuration of a cathode 2 (a second electrode) which is of a semi-dipping type. As illustrated in FIG. 1, the carbon dioxide immobilization unit according to the embodiment includes the anode 1 and the cathode 2 which are disposed to face each other with a proton conductor 3 in between.

[0018] In the carbon dioxide immobilization unit, an oxidoreductase is present on a surface of the anode 1 or a surface of the cathode 2, or both, and an organic acid such as formic acid or a carbohydrate such as glucose is produced from carbon dioxide (CO₂) by reaction opposite to reaction in a biofuel cell in related art. Here and in the following description, a surface of an electrode includes an outer surface of the electrode and an inner surface of a gap in an inside of the electrode.

[Anode 1]

[0019] In the anode 1, water (H₂O) is oxidatively decomposed to produce oxygen (O₂) and to extract protons (H⁺) and electrons (e⁻). Therefore, the anode 1 adopts a dipping type electrode configuration in which the anode 1 is directly in contact with a liquid phase such as an electrolytic solution 13 including a buffer substance or is in contact with the liquid phase with a separator 14 made of nonwoven or the like in between as illustrated in FIG. 2. It is to be noted that, in the electrode configuration illustrated in FIG. 2, the electrolytic solution 13 serves as the proton conductor 3.

[0020] An electrode configuring the anode 1 is not specifically limited; however, for example, an electrode including, on a surface of an electrode 11 made of a conductive porous material, an enzyme immobilization layer 12 where an oxidoreductase or the like is immobilized may be used. As the conductive porous material used in this case, a known material may be used, and in particular, a carbon-based material such as porous carbon, carbon pellets, carbon felt, carbon paper, or a laminate of carbon fiber or carbon microparticles is suitable.

[0021] Moreover, examples of the oxidoreductase immobilized on the surface of the anode 1 include bilirubin oxidase (BOD), laccases, and ascorbate oxidase. Moreover, an electron mediator may be immobilized, together

with the above-described enzyme, on the surface of the anode 1 to decompose water by reaction of the enzyme and the electron mediator. In this case, as the electron mediator, a compound having a quinone skeleton is preferably used, and in particular, a compound having a naphthoquinone skeleton is suitable. More specifically, 2-amino-1,4-naphthoquinone (ANQ), 2-amino-3-methyl-1,4-naphthoquinone (AMNQ), 2-methyl-1,4-naphthoquinone (VK3), 2-amino-3-carboxy-1,4-naphthoquinone (ACNQ) or the like may be used.

[0022] It is to be noted that, as the compound having the quinone skeleton, in addition to the compound having the naphthoquinone skeleton, for example, anthraquinone or a derivative thereof may be used. Moreover, if necessary, one kind or two or more kinds of other compounds functioning as electron mediators may be immobilized together with the compound having the quinone skeleton. Further, the anode 1 is not limited to an electrode having a surface on which an oxidoreductase is immobilized, and, for example, an electrode to which a microorganism including an oxidoreductase and functioning as a reaction catalyst is attached may be used, as long as the oxidoreductase is present on a surface of the electrode.

[Cathode 2]

[0023] On the other hand, in the cathode 2, an organic acid such as formic acid or a carbohydrate such as glucose is produced from carbon dioxide (CO_2), and the protons (H^+) and the electrons (e^-) produced in the anode 1. Therefore, the cathode 2 adopts an air-exposure type electrode configuration in which an electrode is directly in contact with a vapor phase to allow carbon dioxide to be sufficiently supplied thereto, or a semi-dipping type electrode configuration, as illustrated in FIG. 3, in which an electrode is in contact with the vapor phase with a gas-liquid separation film 25 in between. In the case where the cathode 2 is the semi-dipping type electrode, the cathode 2 is also directly in contact with the liquid phase such as the electrolytic solution 13 including the buffer substance, or is also in contact with the separator 24 made of nonwoven in between, as illustrated in FIG. 3.

[0024] Moreover, as the cathode 2, for example, an electrode including an enzyme immobilization layer 22 on a surface of an electrode 21 made of a conductive porous material may be used. As the conductive porous material forming the cathode 2, a known material may be also used, and in particular, a carbon-based material such as porous carbon, carbon pellets, carbon felt, carbon paper, or a laminate of carbon fiber or carbon micro-particles is suitable.

[0025] On the other hand, the enzyme immobilized on the surface of the cathode 2 is allowed to be appropriately selected depending on a product, and, for example, when formic acid is produced, formate dehydrogenase (FDH) may be used. Moreover, when glucose is produced, glucose dehydrogenase (GDH) may be used.

[0026] In addition, electron transfer enzymes, ATP synthases, enzymes relating to saccharometabolism, for example, known enzymes such as hexokinase, glucose phosphate isomerase, phosphofructokinase, fructose biphosphate aldolase, triosephosphate isomerase, glyceraldehydephosphate dehydrogenase, phosphoglyceromutase, phosphopyruvate hydratase, pyruvate kinase, L-lactate dehydrogenase, D-lactate dehydrogenase, pyruvate dehydrogenase, citrate synthase, aconitase, isocitrate dehydrogenase, 2-oxoglutarate dehydrogenase, succinyl-CoA synthetase, succinate dehydrogenase, fumarase, and malonate dehydrogenase may be usable.

[0027] Moreover, coenzyme oxidase or an electron mediator is preferably immobilized, together with the enzyme such as FDH, on the surface of the cathode 2. Examples of a coenzyme used in this case include NADH and NADPH, and diaphorase reducing an oxidant thereof (such as NAD^+ or NADP^+). Moreover, examples of the electron mediator immobilized together with these enzymes include potassium hexacyanoferrate, potassium ferricyanide, and potassium octacyanotungstate.

[0028] It is to be noted that the cathode 2 is also not limited to an electrode having a surface on which an oxidoreductase is immobilized, and, for example, an electrode to which a microorganism including an oxidoreductase and functioning as a reaction catalyst is attached may be used, as long as the oxidoreductase is present on a surface of the electrode. Moreover, as illustrated in FIG. 3, in the electrode 21 configuring the cathode 2, a vapor-liquid coexistence layer 23 in which a vapor phase and a liquid phase coexist may be further formed on an outer side of the enzyme immobilization layer 22 where the enzyme or the like is immobilized.

[Proton conductor 3]

[0029] The proton conductor 3 may be a material not having electronic conductivity and capable of transporting protons (H^+), and an electrolytic solution including a buffer substance is typically used. In this case, for example, when a separator (for example, cellophane, nonwoven, or the like) impregnated with an electrolytic solution is sandwiched between electrodes, a short circuit is preventable while protons are conducted. Moreover, as the proton conductor 3, a separator having proton conductivity such as an ion-exchange resin film having a fluorine-containing carbon sulfonic acid group may be used.

[0030] On the other hand, it is preferable that the carbon dioxide immobilization unit according to the embodiment include a carbon dioxide supply section 5 supplying carbon dioxide or a gas containing carbon dioxide to the cathode 2. Moreover, it is more preferable that the carbon dioxide immobilization unit further include an oxygen removal section 4 removing oxygen produced in the anode 1, and a product recovery section 6 extracting the organic acid or the carbohydrate produced in the cathode 2.

[Oxygen removal section 4]

[0031] To accelerate the above-described anode reaction, it is preferable to remove oxygen (O_2) existing around the anode 1. A method of doing so, that is, the configuration of the oxygen removal section 4 is not specifically limited; however, for example, the oxygen removal section 4 may have a configuration in which a solution around the anode 1 is allowed to flow, and deoxidized water is supplied to emit a solution including oxygen. Thus, the oxygen concentration in the solution around the anode 1 is allowed to be reduced; therefore, a decline in anode reaction is preventable.

[Carbon dioxide supply section 5]

[0032] To accelerate cathode reaction, it is preferable to supply a sufficient amount of carbon dioxide (CO_2) to the cathode 2. Therefore, in the carbon dioxide immobilization unit according to the embodiment, carbon dioxide or a gas containing carbon dioxide is supplied from the carbon dioxide supply section 5 to the cathode 2 or its surrounding. The gas supplied from the carbon dioxide supply section 5 may be a gas containing carbon dioxide in concentration equivalent to or higher than the concentration of carbon dioxide in air, and, for example, in the case where the concentration of carbon dioxide in air is 0.028 vol%, a gas containing carbon dioxide in concentration of 0.028 to 100 vol% both inclusive may be supplied to the cathode 2. Thus, the concentration of carbon dioxide around the cathode 2 is allowed to be maintained at a high level, and reaction efficiency is allowed to be enhanced.

[0033] As the gas supplied from the carbon dioxide supply section 5 to the cathode 2, for example, exhaust from a thermal power station or a vehicle may be used, and dry ice, exhalation, and the like may be also used. A gas containing a high concentration of carbon dioxide is easily obtainable through using such a gas, and the organic acid or the carbohydrate is efficiently obtainable.

[Product recovery section 6]

[0034] To accelerate the above-described cathode reaction, it is preferable to remove a product (the organic acid or the carbohydrate) existing around the cathode 2. A method of doing so, that is, the configuration of the product recovery section 6 is not specifically limited; however, for example, a method of recovering a product contained in a solution around the cathode 2 through allowing the solution to flow and converting the product into a salt to precipitate the salt, or a method of recovering the product through allowing an absorbent such as activated carbon to absorb the product is applicable.

[0035] Thus, the concentration of the product in the solution around the cathode 2 is allowed to be reduced, thereby preventing a decline in cathode reaction. It is to be noted that, in the case where such a product recovery

section 6 is included, it is preferable that the cathode 2 be a semi-dipping type electrode. Therefore, as the solution around the cathode 2 flows, the product is allowed to be immediately removed from the cathode 2 and recovered.

[Operation]

[0036] Next, the operation of the carbon dioxide immobilization unit according to the embodiment will be described below. As illustrated in FIG. 1, when input electric power is externally supplied to the carbon dioxide immobilization unit according to the embodiment, the following reaction proceeds.

[0037] More specifically, in the anode 1, water (H_2O) is oxidized by the oxidoreductase in the enzyme immobilization layer 12 disposed on the surface to extract protons (H^+) and electrons (e^-). For example, the oxygen removal section 4 allows oxygen (O_2) produced by this reaction to exit from the carbon dioxide immobilization unit. On the other hand, the protons (H^+) are transferred to the cathode 2 through the proton conductor 3, and the electrons (e^-) are transmitted to the cathode 2 through an external circuit.

[0038] Moreover, in the cathode 2, an organic acid or a carbohydrate is produced from the protons (H^+) and the electrons (e^-) produced in the anode 1, and carbon dioxide (CO_2) which is supplied from, for example, the carbon dioxide supply section 5 and is present in a vapor phase or a liquid phase in contact with the cathode 2. It is to be noted that, for example, the product recovery section 6 allows the organic acid or the carbohydrate produced by this reaction to exit from the carbon dioxide immobilization unit.

[0039] Thus, in the carbon dioxide immobilization unit according to the embodiment, as the oxidoreductase is used, the organic acid or the carbohydrate is allowed to be easily produced through only inputting electric power to the carbon dioxide immobilization unit without using hydrogen. Moreover, in a field where carbon dioxide is emitted, the carbon dioxide immobilization unit is allowed to immobilize carbon dioxide more efficiently, and a useful carbon compound is obtainable.

[0040] For example, to immobilize carbon dioxide, a method of trapping carbon dioxide deep in the ground, or the like is considered in related art; however, an enormous amount of energy is necessary in the method or the like, and downsizing is difficult. On the other hand, the carbon dioxide immobilization unit according to the embodiment is allowed to immobilize carbon dioxide as a useful compound while using energy (electric power), and has a small and simple configuration; therefore, the carbon dioxide immobilization unit is applicable to a wide range of fields.

(2. Modification Example of First Embodiment)

[Entire configuration of cell]

[0041] In the above-described embodiment, the carbon dioxide immobilization unit producing formic acid from carbon dioxide is described; however, the present invention is not limited thereto, and the carbon dioxide immobilization unit is allowed to produce a carbohydrate such as methanol or glucose in addition to the organic acid such as formic acid.

[0042] FIG. 4 is a diagram schematically illustrating a principle of a carbon dioxide immobilization unit according to a modification example of the above-described embodiment. It is to be noted that, in FIG. 4, like components are denoted by like numerals as of the carbon dioxide immobilization unit according to the first embodiment illustrated in FIG. 1 and will not be further described. As illustrated in FIG. 4, the carbon dioxide immobilization unit according to the modification example also includes an anode 31 and a cathode 32 which are disposed to face each other with the proton conductor 3 in between.

[Cathode 32]

[0043] In the carbon dioxide immobilization unit according to the modification example, three kinds of NAD⁺-dependent dehydrogenases as dehydrogenase groups are immobilized in the cathode 32, and methanol (CH₃OH) is produced from CO₂ through a plurality of steps. More specifically, formic acid is produced from carbon dioxide by formate dehydrogenase (FateDH). Then, the formic acid is converted into formaldehyde by formaldehyde genase (FaLDH), and then methanol is produced by alcohol dehydrogenase (ADH).

[Operation]

[0044] When input electric power is supplied to the carbon dioxide immobilization unit according to the modification example, the following reaction proceeds. More specifically, in the anode 31, water (H₂O) is oxidized by the oxidoreductase existing on the enzyme immobilization layer 12 disposed on the surface to extract protons (H⁺) and electrons (e⁻). For example, the oxygen removal section 4 allows oxygen (O₂) produced by this reaction to exit from the carbon dioxide immobilization unit. On the other hand, the protons (H⁺) are transferred to the cathode 32 through the proton conductor 3, and the electrons (e⁻) are transmitted to the cathode 2 through an external circuit.

[0045] Moreover, in the cathode 32, formic acid, formaldehyde, and methanol are produced from the protons (H⁺) and the electrons (e⁻) produced in the anode 31, and carbon dioxide (CO₂) which is supplied from, for example, the carbon dioxide supply section 5 and is present in a vapor phase or a liquid phase in contact with the cathode 32. Then, if necessary, product recovery sections 36a to

36c allow formic acid, formaldehyde, and methanol produced in such a manner, respectively, to exit from the carbon dioxide immobilization unit.

[0046] In the carbon dioxide immobilization unit according to the modification example, as carbon dioxide is allowed to be reduced by multiple-step reaction, production of methanol which is commercially more beneficial than formic acid and has not been proposed in carbon dioxide immobilization units in related art is achievable. Moreover, in the carbon dioxide immobilization unit according to the modification example, three kinds of intermediate products including formic acid are allowed to be recovered, if necessary. It is to be noted that configurations and effects of the carbon dioxide immobilization unit according to the modification example other than those described above are similar to those in the above-described embodiment.

Claims

1. A carbon dioxide immobilization unit comprising at least:
 - a first electrode decomposing water to produce protons;
 - a second electrode producing an organic acid or a carbohydrate from the protons produced in the first electrode and carbon dioxide; and
 - a proton conductor transferring the protons produced in the first electrode to the second electrode, wherein an oxidoreductase is present on a surface of the first electrode or a surface of the second electrode, or both.
2. The carbon dioxide immobilization unit according to claim 1, further comprising a carbon dioxide supply section supplying carbon dioxide to the second electrode.
3. The carbon dioxide immobilization unit according to claim 2, wherein the carbon dioxide supply section supplies a gas containing carbon dioxide in concentration of 0.028 to 100 vol% both inclusive.
4. The carbon dioxide immobilization unit according to claim 3, comprising:
 - an oxygen removal section removing oxygen produced in the first electrode; and
 - a product recovery section extracting the organic acid or the carbohydrate produced in the second electrode.
5. The carbon dioxide immobilization unit according to claim 1, wherein

the first electrode is a dipping type electrode which is directly in contact with a liquid phase or is in contact with the liquid phase with a separator in between, and the second electrode is a semi-dipping type electrode which is directly in contact with the liquid phase or is in contact with the liquid phase with a separator in between, as well as is in contact with a vapor phase with a gas-liquid separator film in between.

6. The carbon dioxide immobilization unit according to claim 1, wherein the first electrode or the second electrode, or both are formed of a conductive porous material.

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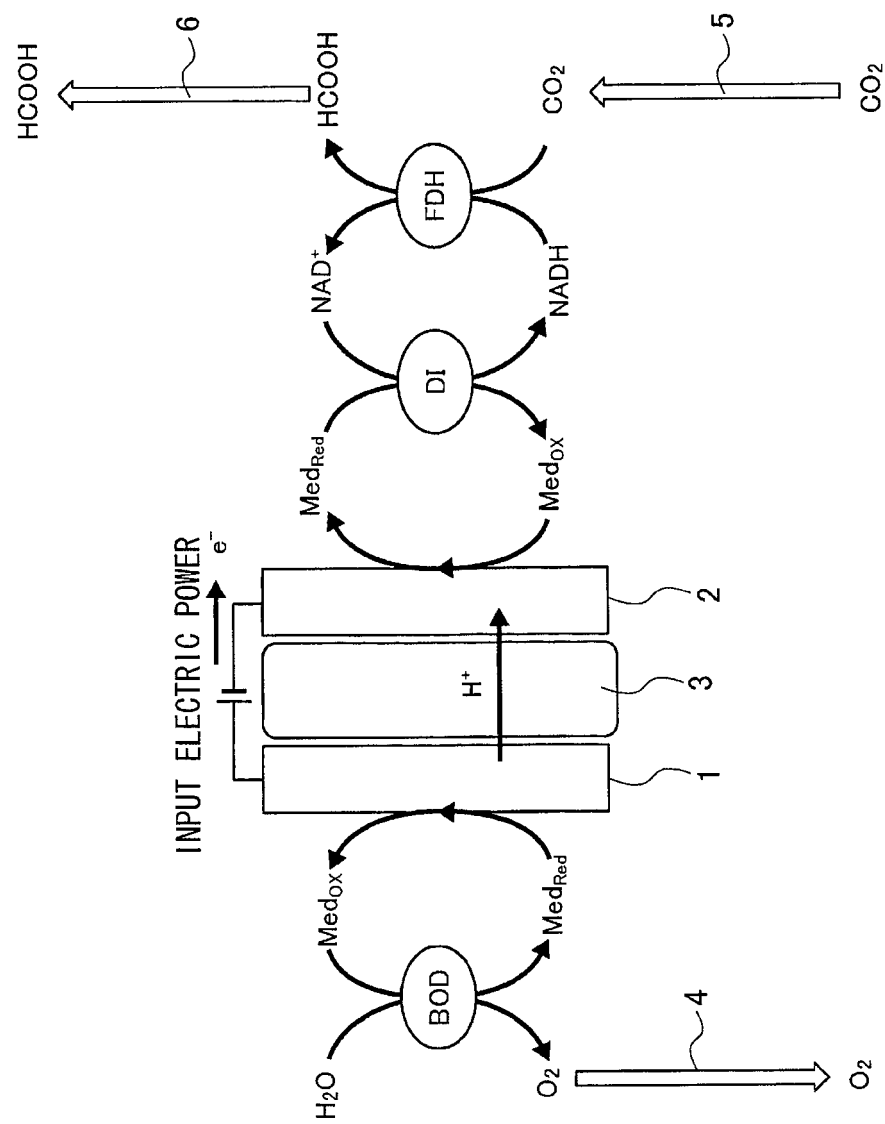
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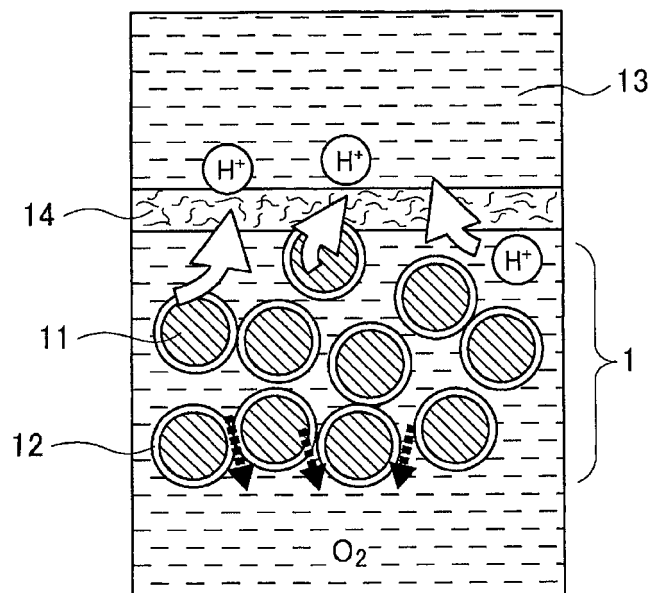
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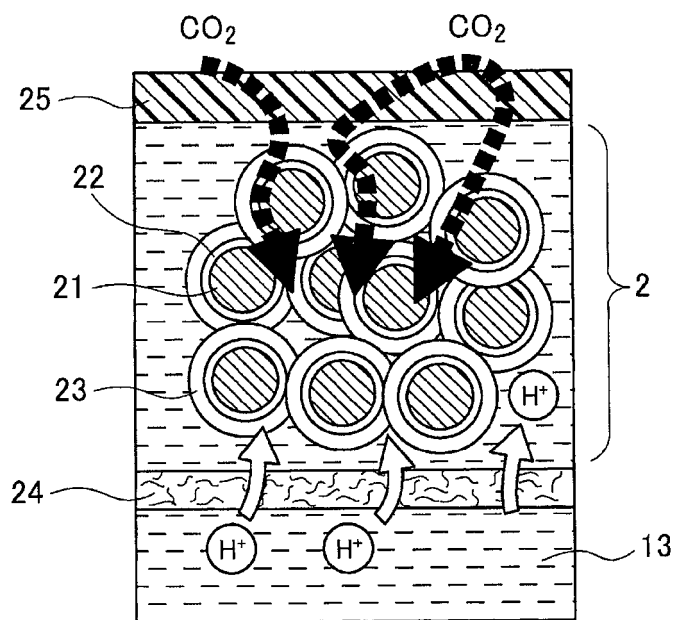
[FIG. 1]



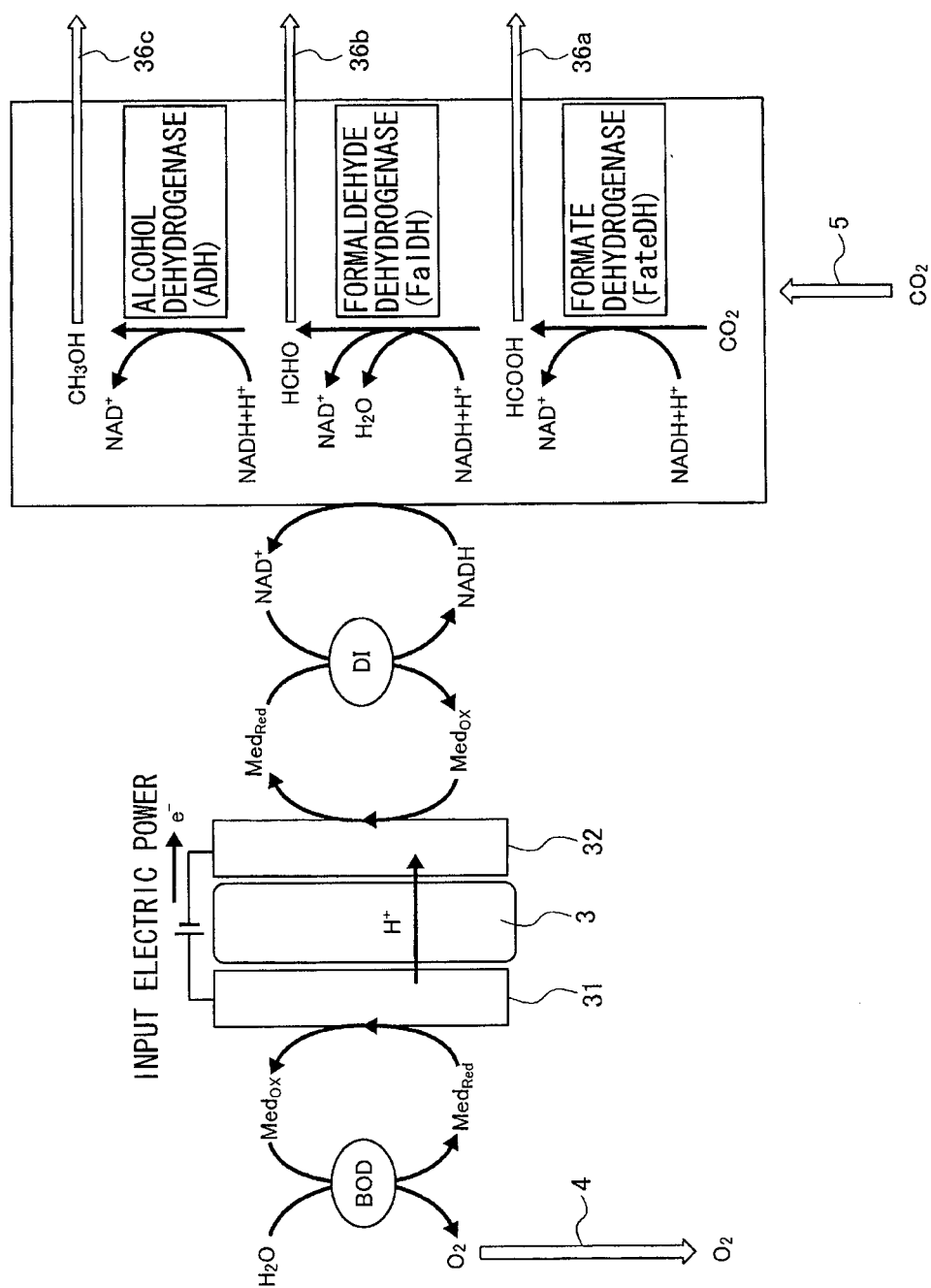
[FIG. 2]



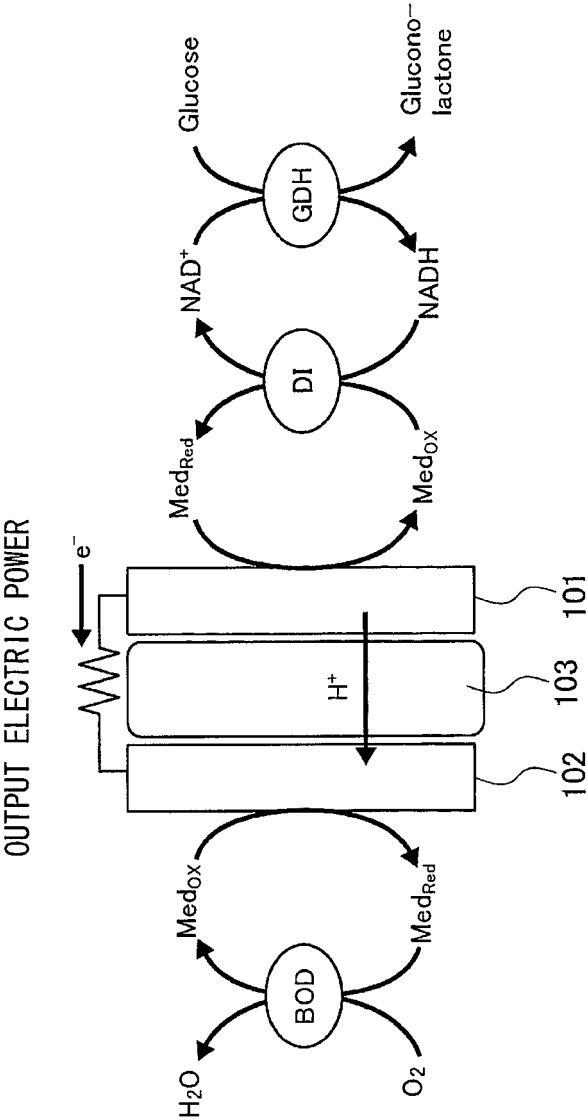
[FIG. 3]



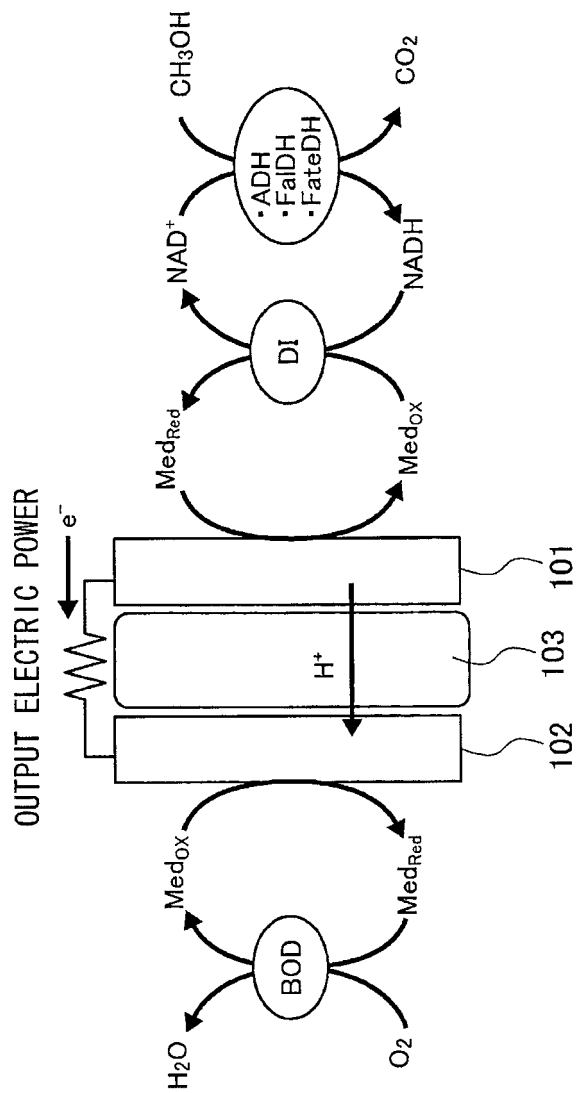
[FIG. 4]



[FIG. 5]



[FIG. 6]



INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP2011/065681

A. CLASSIFICATION OF SUBJECT MATTER <i>C25B3/04</i> (2006.01) i, <i>C01B31/20</i> (2006.01) i, <i>C25B11/03</i> (2006.01) i, <i>C25B11/06</i> (2006.01) i, <i>H01M8/16</i> (2006.01) i 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) <i>C25B3/04</i> , <i>C01B31/20</i> , <i>C25B11/03</i> , <i>C25B11/06</i> , <i>H01M8/16</i> Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched Jitsuyo Shinan Koho 1922-1996 Jitsuyo Shinan Toroku Koho 1996-2011 Kokai Jitsuyo Shinan Koho 1971-2011 Toroku Jitsuyo Shinan Koho 1994-2011 Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)		
C. DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	JP 8-246177 A (Director General, Agency of Industrial Science and Technology), 24 September 1996 (24.09.1996), paragraphs [0006] to [0011]; fig. 1 (Family: none)	1-6
Y	JP 2004-71559 A (Sony Corp.), 04 March 2004 (04.03.2004), paragraphs [0012], [0032]; fig. 1 & US 2005/0053825 A1 & EP 1531512 A1 & WO 2004/012293 A1 & CN 1579033 A & AU 2003248128 A	1-6
Y	JP 6-158374 A (Choichi FURUYA), 07 June 1994 (07.06.1994), paragraphs [0001] to [0008]; fig. 1 (Family: none)	5
☐ Further documents are listed in the continuation of Box C. ☐ See patent family annex.		
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Date of the actual completion of the international search 22 July, 2011 (22.07.11)		Date of mailing of the international search report 02 August, 2011 (02.08.11)
Name and mailing address of the ISA/ Japanese Patent Office		Authorized officer
Facsimile No.		Telephone No.

REFERENCES CITED IN THE DESCRIPTION

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