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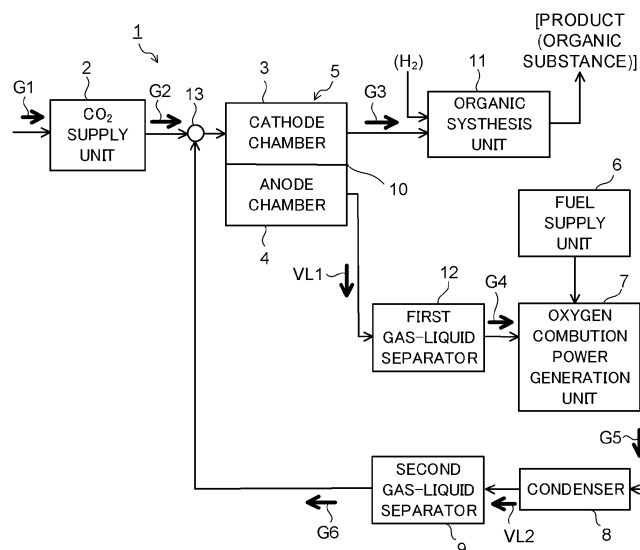
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(54) **CARBON DIOXIDE CONVERSION DEVICE**

(57) A CO₂ conversion device of the embodiment includes: a CO₂ supply unit supplying CO₂; a CO₂ electrolysis unit including a cathode chamber supplied with CO₂ from the CO₂ supply unit and reducing and converting CO₂ into CO and an anode chamber oxidizing substances to be oxidized to produce oxides; a fuel supply unit supplying fuel; an oxygen combustion power generation unit to which O₂-CO₂-containing gas discharged from

from the anode chamber of the carbon dioxide electrolysis unit is supplied, the fuel is supplied from the fuel supply unit, and that combusts the O₂-CO₂-containing gas; a condenser cooling and condensing water vapor-CO₂-containing gas discharged from the oxygen combustion power generation unit; and a gas-liquid separator separating a water-CO₂ two-phase fluid discharged from the condenser into water and CO₂.

FIG. 1



Description

FIELD

5 **[0001]** Embodiments disclosed herein relate generally to a carbon dioxide conversion device.

BACKGROUND

10 **[0002]** Carbon dioxide (CO₂) generated by combustion of fossil fuels such as natural gas, coal, and petroleum is considered a major cause of global warming due to the greenhouse effect, and there is a need to reduce the use of fossil fuels. CO₂ is removed from exhaust gas emitted from CO₂ generation sources, and chemical synthesis is performed using the CO₂ removed from the exhaust gas as feedstock. As part of this process, a carbon dioxide conversion device (electrolytic device) that electrolyzes CO₂ and water (H₂O) to produce carbon monoxide (CO) and oxygen (O₂) is being developed. In the carbon dioxide conversion device, the produced CO and approximately a similar amount of CO₂ are moved to an oxygen-producing side and released with the oxygen. As a result, an effective utilization rate of CO₂ supplied to the carbon dioxide conversion device is as low as 50% or less. Therefore, there is a need to reduce the release of CO₂ into the atmosphere and to increase the effective utilization rate of CO₂.

SUMMARY

20 **[0003]** A subject to be solved by the aspects of the invention is to provide a carbon dioxide conversion device that reduces release of CO₂ into the atmosphere and increases an effective utilization rate of CO₂.

[0004] According to the aspects of the present invention, there is provided a carbon dioxide conversion device which makes it possible to reduce release of CO₂ into the atmosphere and increase an effective utilization rate of CO₂.

BRIEF DESCRIPTION OF THE DRAWINGS

[0005]

30 FIG. 1 is a diagram illustrating a carbon dioxide conversion device of an embodiment.
FIG. 2 is a diagram illustrating gases produced by each unit and amounts of gases produced when hydrogen (H₂) is used as fuel in the carbon dioxide conversion device of the embodiment.
FIG. 3 is a diagram illustrating gases produced by each unit and amounts of gases produced when methane (CH₄) is used as fuel in the carbon dioxide conversion device of the embodiment.
35 FIG. 4 is a diagram illustrating gases produced by each unit and amounts of gases produced when carbon (C) is used as fuel in the carbon dioxide conversion device of the embodiment.
FIG. 5 is a diagram illustrating gases produced by each unit and amounts of gases produced when hydrogen (H₂) and carbon monoxide (CO) are used as fuel in the carbon dioxide conversion device of the embodiment.

DETAILED DESCRIPTION

40 **[0006]** The carbon dioxide conversion device of the embodiment includes: a carbon dioxide supply unit that supplies carbon dioxide; a carbon dioxide electrolysis unit that includes a cathode chamber where carbon dioxide is supplied from the carbon dioxide supply unit and reduces and converts the carbon dioxide into carbon monoxide and an anode chamber that oxidizes substances to be oxidized to produce oxides; a fuel supply unit that supplies fuel; an oxygen combustion power generation unit to which oxygen-carbon dioxide-containing gas discharged from the anode chamber of the carbon dioxide electrolysis unit is supplied, the fuel is supplied from the fuel supply unit, and that combusts the oxygen-carbon dioxide-containing gas; a condenser that cools and condenses water vapor-carbon dioxide-containing gas discharged from the oxygen combustion power generation unit; and a gas-liquid separator that separates a water-carbon dioxide two-phase fluid discharged from the condenser into water and carbon dioxide.

50 **[0007]** Carbon dioxide conversion devices in embodiments will be described hereinafter with reference to the drawings. Substantially the same components are denoted by the same reference signs and explanation thereof may be omitted in some cases in the embodiments described below. The drawings are schematic, and a relationship between a thickness and a planar size, thickness proportions of the respective portions, and the like are sometimes different from actual ones. The "~" symbol in the following explanation indicates a range between upper and lower limit values of the respective numerical values. In such cases, each numerical value range includes the upper and lower limit values.

[0008] FIG. 1 is a diagram illustrating a carbon dioxide conversion device of an embodiment. The carbon dioxide (CO₂) conversion device 1 illustrated in FIG. 1 includes a CO₂ supply unit 2 that supplies CO₂; a CO₂ electrolysis unit 5 that

includes a cathode chamber 3 reducing and converting CO₂ into carbon monoxide (CO) and an anode chamber 4 that oxidizes substances to be oxidized to produce oxides; a fuel supply unit 6 that supplies fuel; an oxygen combustion power generation unit 7 to which oxygen (O₂)-carbon dioxide (CO₂)-containing gas discharged from the anode chamber 4 of the CO₂ electrolysis unit 2 is supplied, the fuel is supplied from the fuel supply unit 6, and that combusts the O₂-CO₂-containing gas; a condenser 8 that cools and condenses water vapor (H₂O)-carbon dioxide (CO₂)-containing gas discharged from the oxygen combustion power generation unit 7; and a gas-liquid separator 9 that separates a water (H₂O)-carbon dioxide (CO₂) two-phase fluid discharged from the condenser 8 into water (H₂O) and carbon dioxide (CO₂).

[0009] As the CO₂ supply unit 2, a device that recovers and supplies CO₂ from carbon dioxide (CO₂)-containing gas or a CO₂ storage unit is used. For example, the CO₂ supply unit 2 is configured to separate and recover CO₂ from CO₂-containing emission gas (CO₂-containing gas) G1 emitted from thermal power plants, waste incineration plants, steel plants, and other plants, and supply CO₂ gas G2 with increased CO₂ concentration to the CO₂ electrolysis unit 5. In such CO₂ supply unit 2, for example, the following methods can be used: a chemical absorption method using a chemical absorbing liquid such as an amine aqueous solution, a physical absorption method using a physical absorbing liquid such as methanol or a polyethylene glycol solution, a solid absorption method using a solid absorbent such as an amine compound, a membrane separation method using a CO₂ separation membrane, a physical adsorption method using zeolite or other inorganic substances as an absorber, a PSA (pressure swing adsorption) method, a TSA (thermal swing adsorption) method, and other methods. For example, in the chemical absorption method and a device using the amine aqueous solution, the emission gas G1 is supplied to an absorption tower where the amine aqueous solution is sprayed, and the amine aqueous solution that has absorbed CO₂ is heated in a regeneration tower to recover CO₂ emitted from the amine aqueous solution. The CO₂ recovery methods and devices applied to the CO₂ supply unit 2 are not limited, and various methods and devices that can recover CO₂ from the emission gas G1 can be used.

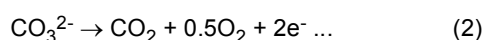
[0010] The CO₂ electrolysis unit 5 is a CO₂ electrolytic device with an electrolysis cell and includes a cathode chamber (reduction portion) 3 and an anode chamber (oxidation portion) 4. The cathode chamber 3 includes a reduction electrode (cathode) and the anode chamber 4 includes an oxidation electrode (anode), and an electrolytic solution is circulated or filled in at least the anode chamber 4. In the cathode chamber 3, CO₂ gas may be circulated or a CO₂ containing electrolytic solution may be circulated or filled. In the cathode chamber 3 or anode chamber 4, the electrolytic solution is a solution using water (H₂O), for example, an aqueous solution containing any electrolyte. Examples of the aqueous solutions containing electrolytes include aqueous solutions containing phosphate ions (PO₄²⁻), borate ions (BO₃³⁻), sodium ions (Na⁺), potassium ions (K⁺), calcium ions (Ca²⁺), lithium ions (Li⁺), cesium ions (Cs⁺), magnesium ions (Mg²⁺), chloride ions (Cl⁻), hydrogen carbonate ions (HCO₃⁻), carbonate ions (CO₃²⁻), hydroxide ions (OH⁻), and other ions. Concrete examples of the electrolytic solutions include alkaline aqueous solutions in which KOH, KHCO₃, K₂CO₃, and the like are dissolved.

[0011] The cathode chamber 3 is supplied with the CO₂ gas G2 from the CO₂ supply unit 2. The cathode chamber 3 has a gas flow path facing the non-illustrated reduction electrode, and the CO₂ gas is supplied to the gas flow path. The anode chamber 4 has a liquid flow path facing the non-illustrated oxidation electrode, and the electrolytic solution is supplied to the liquid flow path. A non-illustrated power supply is connected to the reduction and oxidation electrodes. The cathode chamber 3 and anode chamber 4 are separated from each other by a diaphragm 10 capable of moving ions such as hydrogen ions (H⁺), hydroxide ions (OH⁻), carbonate ions (CO₃²⁻), hydrogen carbonate ions (HCO₃⁻), and other ions, for example, an ion exchange membrane. The CO₂ electrolysis unit 5 may be a single electrolysis cell, have a structure in which the single electrolysis cells are connected in a plane direction, or a stack structure in which a plurality of electrolysis cells are stacked and integrated.

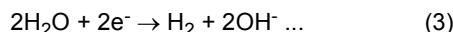
[0012] In the cathode chamber 3 and anode chamber 4 of the CO₂ electrolysis unit 5, the following reactions occur. In the cathode chamber 3, an electrolytic reaction and reduction reaction of CO₂ occur as shown in formula (1) below. In the cathode chamber 3, the reduction reaction of CO₂ produces CO and carbonate ions (CO₃²⁻).



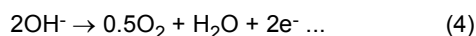
[0013] The carbonate ions (CO₃²⁻) produced in the cathode chamber 3 move to the anode chamber 4 through the diaphragm 10. In the anode chamber 4, as shown in formula (2) below, an oxidation reaction of the carbonate ions (CO₃²⁻) produced in the cathode chamber 3 and moved through the diaphragm 10 occurs, resulting in production of CO₂ and O₂.



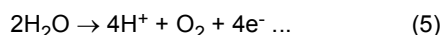
[0014] Furthermore, in the cathode chamber 3, the electrolytic reaction (reduction reaction) of H₂O in the electrolytic solution occurs simultaneously with the electrolytic reaction (reduction reaction) of CO₂, producing hydrogen (H₂) and hydroxide ions (OH⁻), as shown in formula (3) below.



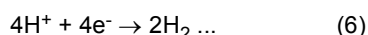
[0015] The hydroxide ions (OH^-) produced in the cathode chamber 3 move to the anode chamber 4 through the diaphragm 10. Water (H_2O) and oxygen (O_2) are then produced in the anode chamber 4, as shown in formula (4) below.



[0016] In the anode chamber 4, water (H_2O) in the electrolytic solution is electrolyzed to produce oxygen (O_2) and hydrogen ions (H^+), as shown in formula (5) below.



[0017] The produced hydrogen ions (H^+) move to the cathode chamber 3 through the diaphragm 10. In the cathode chamber 3 where hydrogen ions (H^+) reach and electrons (e^-) reach through external circuits, hydrogen is generated by the reaction shown in formula (6) below.



[0018] In the cathode chamber 3, CO is produced by the reduction reaction of CO_2 as shown in formula (1), and H_2 is produced by the electrolytic reaction of H_2O as shown in formula (3) and the reaction shown in formula (6). CO and H_2 produced in the cathode chamber 3 are discharged from the cathode chamber 3 together with unreacted CO_2 . Mixed gas G3 containing CO and H_2 , and CO_2 discharged from the cathode chamber 3 is supplied to an organic synthesis unit 11, for example, as part of source gas for an organic synthesis reaction. Hydrogen (H_2) is supplied to the organic synthesis unit 11 as needed, in addition to the mixed gas G3 containing CO and H_2 , as part of the source gas for the organic synthesis reaction.

[0019] In the organic synthesis unit 11, the organic synthesis reactions are performed, for example, using the Fischer-Tropsch synthesis reaction to synthesize hydrocarbons, alcohols, and other organic substances, for example. Concrete examples of the organic substances synthesized in the organic synthesis unit 11 include carbon-containing liquid fuels, or the like. The products (organic substances) of the organic synthesis unit 11 are discharged from the organic synthesis unit 11 and sent, for example, to a separately installed tank or other storage facility (not illustrated).

[0020] Meanwhile, in the anode chamber 4 of the CO_2 electrolysis unit 5, oxygen (O_2) and carbon dioxide (CO_2) are produced by oxidation of carbonate ions (CO_3^{2-}) and hydroxide ions (OH^-), as shown in formulas (2) and (4) above. The gas containing O_2 and CO_2 (O_2 - CO_2 -containing gas) produced in the anode chamber 4 is discharged from the anode chamber 4 together with the electrolytic solution. An electrolytic solution VL1 containing the O_2 - CO_2 -containing gas is sent to a first gas-liquid separation unit 12, which is connected to a discharge pipe of the anode chamber 4, and O_2 - CO_2 -containing gas G4 is separated from the electrolytic solution. The separated electrolytic solution is returned to the anode chamber 4, though not illustrated. Since a CO_2 concentration of the separated O_2 - CO_2 -containing gas G4 is as high as 60 volume% or more, effective utilization of CO_2 would be hindered when the gas G4 is directly released into the atmosphere. Furthermore, since the O_2 - CO_2 -containing gas G4 contains a relatively large amount of O_2 , operation and function of the CO_2 electrolysis unit 5 will be degraded when it is sent directly to the cathode chamber 3. The O_2 - CO_2 -containing gas G4 discharged from the anode chamber 4 of the CO_2 electrolysis unit 5 and separated and recovered in the first gas-liquid separation unit 12 is sent to the oxygen combustion power generation unit 7.

[0021] Closed-cycle gas turbines, turbine combined cycle power generation, Graz cycle gas turbines, supercritical CO_2 turbines, oxy-fuel combustion devices, and the like can be used as the oxygen combustion power generation unit 7. These can efficiently combust and consume O_2 in the O_2 - CO_2 -containing gas using hydrogen or the like as fuel, and can also efficiently operate gas turbines or the like based on combustion, so that electric power can be generated using O_2 in the O_2 - CO_2 -containing gas as an oxidant. The electric power generated by the oxygen combustion power generation unit 7 can be used as part of operating power of the CO_2 electrolysis unit 5.

[0022] To combust and consume the O_2 - CO_2 -containing gas in a combustion reaction in the oxygen combustion power generation unit 7, fuel is supplied to the oxygen combustion power generation unit 7 from the fuel supply unit 6 in addition to the O_2 - CO_2 -containing gas G4. The fuel supplied from the fuel supply unit 6 is not limited, and may be any of hydrogen (H_2), carbon (C), carbon monoxide (CO), and lower hydrocarbons such as methane (CH_4). The fuel supply unit 6 may be an oxygen-blown gasification furnace or the like. The fuel supplied from the fuel supply unit 6 is preferably supplied in an equivalent ratio to O_2 in the O_2 - CO_2 containing gas. In the oxygen combustion power generation unit 7, O_2 is converted into H_2O or CO_2 . For example, when hydrogen (H_2) is supplied as the fuel, H_2O is produced. When carbon (C) or carbon monoxide (CO) is supplied as the fuel, CO_2 is produced. The same is true when methane (CH_4) or other fuels are supplied, and H_2O and CO_2 are produced. The produced CO_2 is recovered simultaneously with CO_2 in the

O₂-CO₂-containing gas.

[0023] When the closed-cycle gas turbines, turbine combined cycle power generation, Graz cycle gas turbines, supercritical CO₂ turbines, oxy-fuel combustion devices, and the like are used as the oxygen combustion power generation unit 7, simply supplying fuel for combustion will result in higher temperatures in the oxygen combustion power generation unit 7. In contrast, using the produced H₂O (for example, water vapor) or CO₂ as a circulating medium can suppress the temperature rise of the oxygen combustion power generation unit 7. For example, H₂O and CO₂ are used as the circulating media in the closed-cycle gas turbines, turbine combined cycle power generation, and Graz cycle gas turbines. In the supercritical CO₂ turbines and oxy-fuel combustion devices, CO₂ is used as the circulating medium. These allow the oxygen combustion power generation unit 7 to operate safely and efficiently.

[0024] H₂O (water vapor)-CO₂-containing gas G5 is discharged from the oxygen combustion power generation unit 7. The H₂O (water vapor)-CO₂-containing gas G5 is sent to the condenser 8 to be cooled and water vapor (H₂O) is condensed to be converted into water. A water (H₂O)-carbon dioxide (CO₂) two-phase fluid VL2 containing water from the condensation of water vapor (H₂O) is discharged from the condenser 8. The H₂O-CO₂ two-phase fluid VL2 is sent to a second gas-liquid separator 9. In the second gas-liquid separator 9, water in the H₂O-CO₂ two-phase fluid VL2 is separated. At least some of the separated water (H₂O) may be used as part of the electrolytic solution supplied to the anode chamber 4 of the CO₂ electrolysis unit, or may be returned to the oxygen combustion power generation unit 7 as the circulating medium. CO₂ gas G6 separated in the second gas-liquid separator 9 is sent to a gas mixer 13. The gas mixer 13 is connected to the cathode chamber 3 of the CO₂ electrolysis unit 5. In the gas mixer 13, the CO₂ gas G6 separated in the second gas-liquid separator 9 is mixed with the CO₂ gas G2 and supplied to the cathode chamber 3. At least one of the water and CO₂ discharged from the second gas-liquid separator 9 may be used as the circulating medium in the oxygen combustion power generation unit 7.

[0025] As described above, the CO₂ gas that is produced as a by-product in the anode chamber 4 and separated in the second gas-liquid separator 9 can be returned to the cathode chamber 3 of the CO₂ electrolysis unit 5, with operation failure and functional degradation and the like in the cathode chamber 3 under control, by combusting the O₂-CO₂-containing gas G4 discharged from the anode chamber 4 of the CO₂ electrolysis unit 5 in the oxygen combustion power generation unit 7 to convert O₂ in the O₂-CO₂-containing gas G4 into H₂O and CO₂. Furthermore, O₂ in the O₂-CO₂-containing gas G4 can be regenerated as CO₂. These features make it possible to reduce release of CO₂ into the atmosphere in the carbon dioxide conversion device 1, increase an effective utilization rate of CO₂, and improve device utilization efficiency.

[0026] FIG. 2 to FIG. 5 each illustrate a relationship between a type of fuel used in the carbon dioxide conversion device 1 of the embodiment and an amount of gas produced in each unit. FIG. 2 is a diagram illustrating the amount of gas produced in each unit when hydrogen (H₂) is used as fuel in the carbon dioxide conversion device 1 of the embodiment. FIG. 3 is a diagram illustrating the amount of gas produced in each unit when methane (CH₄) is used as fuel in the carbon dioxide conversion device 1 of the embodiment. FIG. 4 is a diagram illustrating the amount of gas produced in each unit when carbon (C) is used as fuel in the carbon dioxide conversion device 1 of the embodiment. FIG. 5 is a diagram illustrating the amount of gas produced in each unit when hydrogen (H₂) and carbon monoxide (CO) are used as fuel in the carbon dioxide conversion device 1 of the embodiment. In these figures, the numeric values in each unit indicate the volume of the target gas. In cases of using either fuel, CO₂ can be effectively utilized.

[0027] Some of the water discharged from the second gas-liquid separator 9 may be supplied to the CO₂ electrolysis unit 5. Although not illustrated, some of the water and CO₂ recovered in the second gas-liquid separator 9 can be used as the circulating medium in the oxygen combustion power generation unit 7. Although not illustrated, there are pumps, compressors, blowers, control devices, and other devices to supply fluid between each piece of equipment. To avoid residual oxygen in CO₂ released from the second gas-liquid separator 9, a fuel/oxygen ratio in the oxygen combustion power generation unit 7 is preferably set slightly larger than the equivalent ratio to prevent residual oxygen. Alternatively, a catalytic combustor may be installed upstream or downstream of the second gas-liquid separator 9 to react the residual oxygen with hydrogen or the like. Pt and Pd are used as combustion catalysts.

[0028] The configurations in the embodiments can be applied in combination and partially replaced. While certain embodiments of the present invention have been described herein, these embodiments have been presented by way of example only, and are not intended to limit the scope of the inventions. The embodiments described herein may be embodied in a variety of other forms; furthermore, various omissions, substitutions, and changes may be made without departing from the spirit of the inventions. The embodiments and modifications fall within the scope and spirit of the inventions and fall within the scope of the inventions as set forth in claims and their equivalents.

Claims

1. A carbon dioxide conversion device, comprising:

a carbon dioxide supply unit that supplies carbon dioxide;
a carbon dioxide electrolysis unit that includes a cathode chamber where carbon dioxide is supplied from the carbon dioxide supply unit and reduces and converts the carbon dioxide into carbon monoxide and an anode chamber that oxidizes substances to be oxidized to produce oxides;
5 a fuel supply unit that supplies fuel;
an oxygen combustion power generation unit to which oxygen-carbon dioxide-containing gas discharged from the anode chamber of the carbon dioxide electrolysis unit is supplied, the fuel is supplied from the fuel supply unit, and that combusts the oxygen-carbon dioxide-containing gas;
10 a condenser that cools and condenses water vapor-carbon dioxide-containing gas discharged from the oxygen combustion power generation unit; and
a gas-liquid separator that separates a water-carbon dioxide two-phase fluid discharged from the condenser into water and carbon dioxide.

2. The device according to claim 1, wherein
15 the gas-liquid separator is configured to supply carbon dioxide separated from the water-carbon dioxide two-phase fluid to the cathode chamber of the carbon dioxide electrolysis unit.
3. The device according to claim 1, wherein
20 the gas-liquid separator is configured to supply at least some of the separated water to the anode chamber of the carbon dioxide electrolysis unit as part of an electrolytic solution.
4. The device according to any one of claim 1 to claim 3, wherein
25 the oxygen combustion power generation unit includes a closed-cycle gas turbine, turbine combined cycle power generation, Graz cycle gas turbine, supercritical CO₂ turbine, or oxy-fuel combustion device.
5. The device according to claim 4, wherein
at least one of water and carbon dioxide produced by combustion and carbon dioxide in the oxygen-carbon dioxide-containing gas is supplied as a circulating medium for the oxygen combustion power generation unit.
- 30 6. The device according to claim 4, wherein
at least one of water and carbon dioxide discharged from the gas-liquid separator is supplied as a circulating medium for the oxygen combustion power generation unit.

FIG. 1

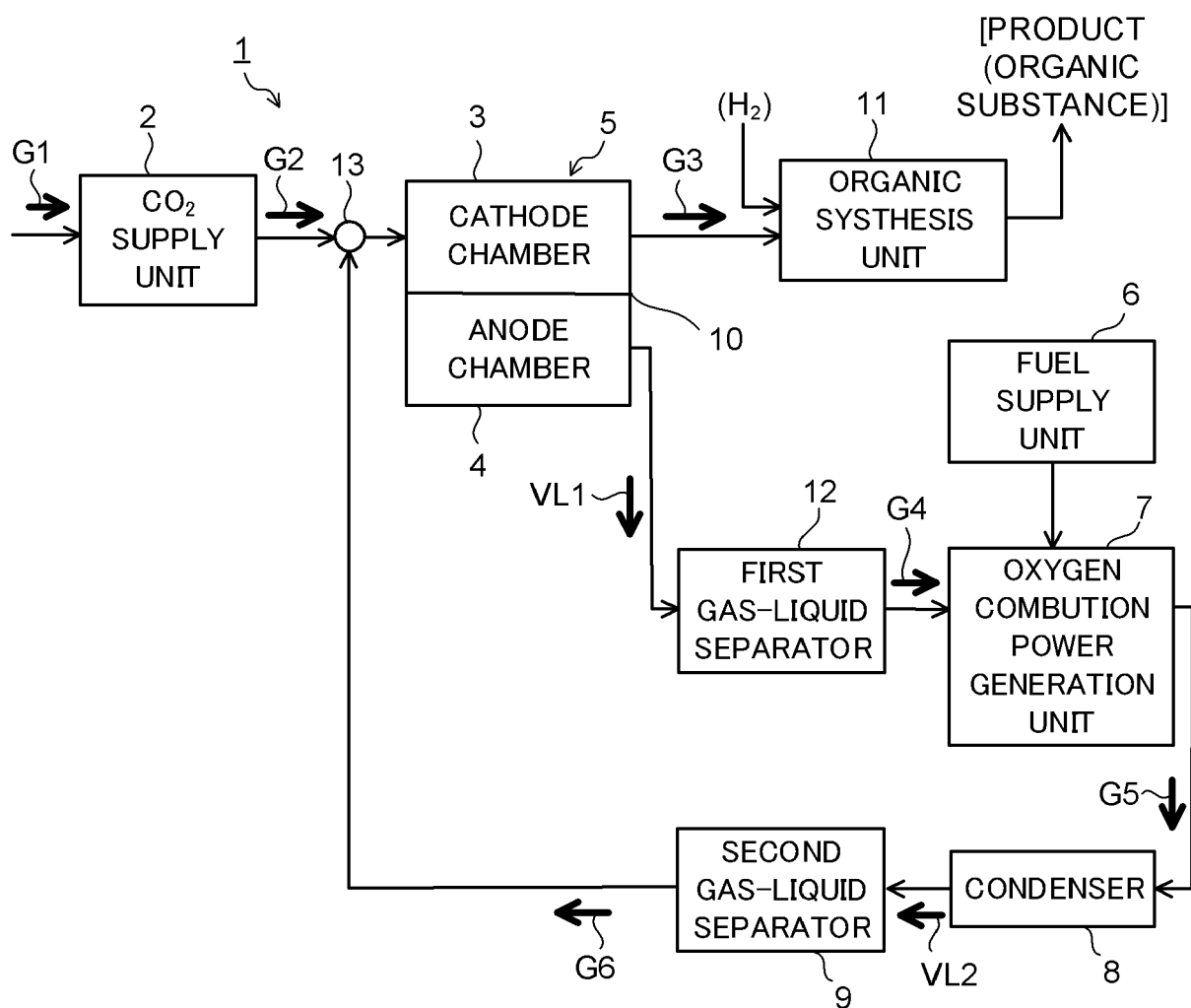


FIG. 2

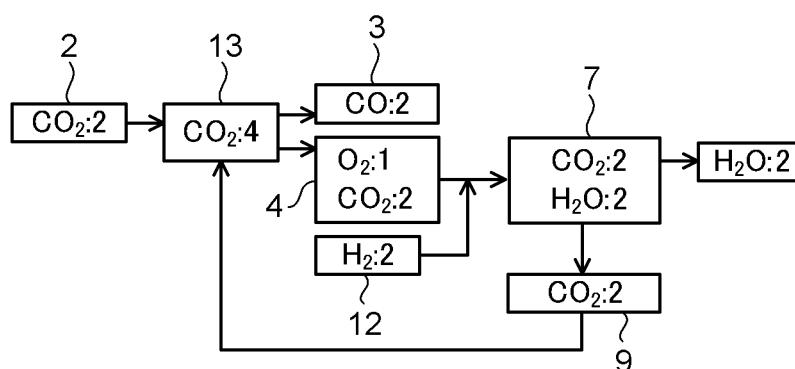


FIG. 3

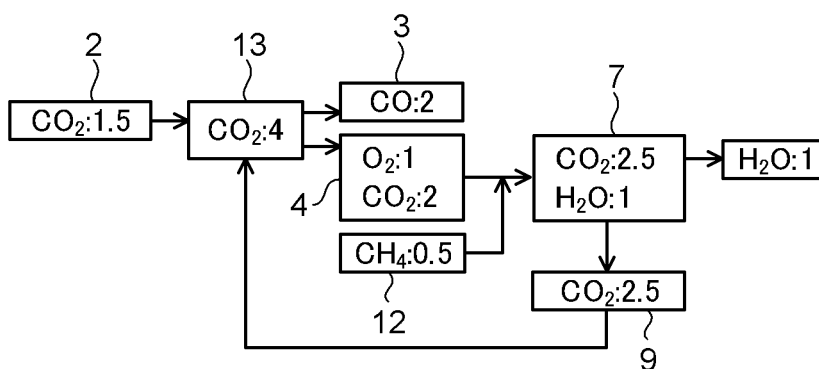


FIG. 4

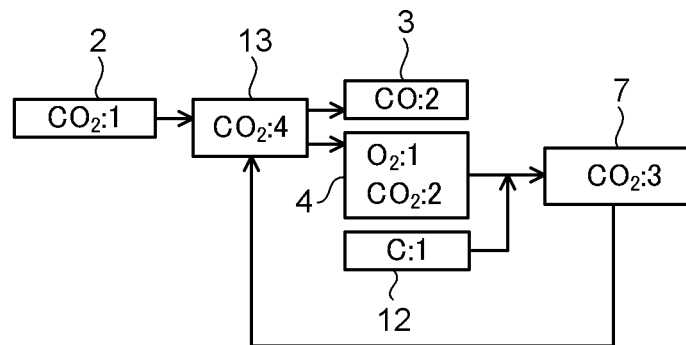
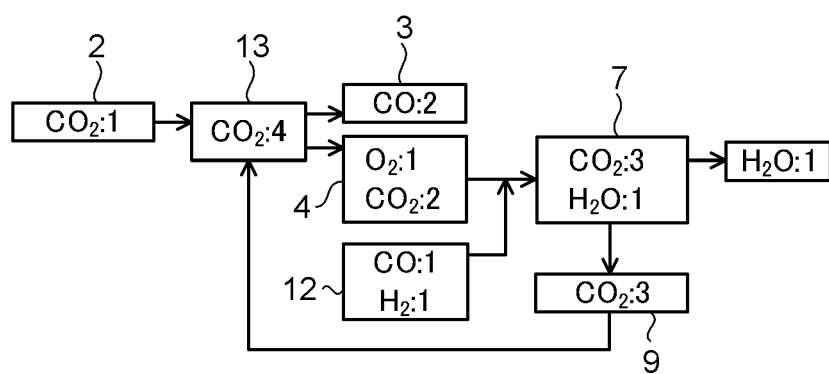


FIG. 5





EUROPEAN SEARCH REPORT

Application Number

EP 24 15 3442

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EPO FORM 1503 03.82 (P04C01)

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (IPC)
X	EP 3 670 705 A1 (AIR LIQUIDE [FR]) 24 June 2020 (2020-06-24) * paragraphs [0047], [0048], [0049], [0085], [0089], [0101], [0105], [0108]; figures 1-3 * -----	1-6	INV. C25B1/04 C25B15/08 F23C9/06 F23L7/00 C25B1/23
X	US 2023/020698 A1 (HANNEMANN FRANK [DE] ET AL) 19 January 2023 (2023-01-19) * paragraph [0043] - paragraph [0051]; figures 2,3 * -----	1,2,4,6	
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			C25B F23C F23L
The present search report has been drawn up for all claims			
Place of search The Hague		Date of completion of the search 25 July 2024	Examiner Harder, Sebastian
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EP 24 15 3442

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