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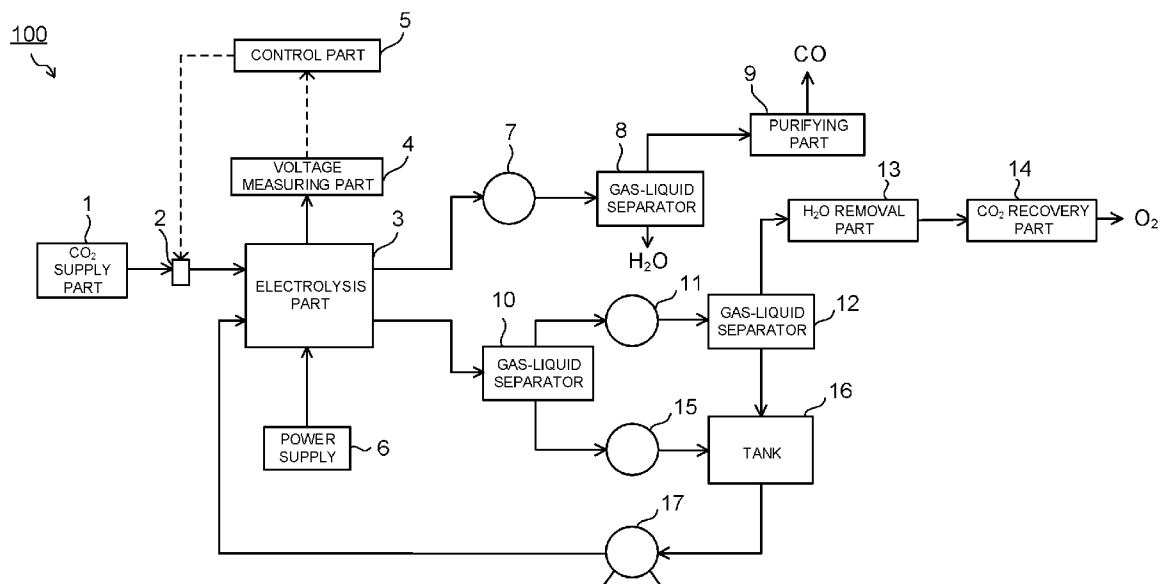
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(54) **CARBON DIOXIDE CONVERSION DEVICE AND CARBON DIOXIDE CONVERSION METHOD**

(57) A carbon dioxide conversion device includes: a carbon dioxide supply part configured to supply a supply gas containing carbon dioxide gas; a flow rate adjusting part that includes flow rate adjusting valves configured to distribute the supply gas to be supplied from the carbon dioxide supply part; an electrolysis part that includes electrolysis cell stacks configured to receive the supply

gas distributed through the flow rate adjusting valves; a voltage measuring part that includes voltmeters configured to measure voltages across the electrolysis cell stacks; and a control part configured to adjust opening degrees of the flow rate adjusting valves in response to voltage signals to be received from the voltmeters.

FIG. 1



Description

FIELD

5 **[0001]** Embodiments relate to a carbon dioxide conversion device and a carbon dioxide conversion method.

BACKGROUND

10 **[0002]** Global warming progresses have been increased the demand for reducing the use of the carbon dioxide (CO₂) sources such as natural gas, coal, and petroleum. In addition, development of chemical synthesis using carbon dioxide as a raw material is underway. As part of this process, there are carbon dioxide conversion devices that electrolyze carbon dioxide to produce carbon monoxide (CO)-containing gas. The carbon dioxide conversion device has electrolysis cell stacks, each electrolysis cell stack being formed by stacking electrolysis cells. In the electrolysis cell stack where the amount of carbon dioxide supplied is insufficient for a load current, the amount of hydrogen generated increases, and an
 15 electric load increases due to voltage increase. On the other hand, in the electrolysis cell stack where the amount of carbon dioxide supplied is excessive for the load current, the amount of unreacted carbon dioxide increases, and a carbon monoxide conversion rate decreases. To solve these problems, there is a need for a carbon dioxide conversion device and a carbon dioxide conversion method that equalizes the amount of carbon dioxide supplied to each electrolysis cell stack.
 20 **[0003]** However, the carbon dioxide conversion device under consideration distributes the supplied carbon dioxide to each electrolysis cell stack from a distribution tube installed upstream of each electrolysis cell stack, and a distribution ratio is determined on a case-by-case basis.

BRIEF DESCRIPTION OF THE DRAWINGS

25 **[0004]**

FIG. 1 is a schematic diagram illustrating an example configuration of a carbon dioxide conversion device of an embodiment.

FIG. 2 is a diagram to further explain the example configuration of some of components illustrated in FIG. 1.

30 FIG. 3 is a cross-sectional schematic diagram illustrating an example structure of an electrolysis cell stack CS.

FIG. 4 is a flowchart to explain an example of a method of adjusting an opening degree of each flow rate adjusting valve FC in an example of a carbon dioxide conversion method.

FIG. 5 is a diagram illustrating a relationship between a rate of increase of a flow rate of a supply gas and a voltage V_i .

35 FIG. 6 is a diagram illustrating a relationship between the rate of increase of the flow rate of the supply gas and a production amount of carbon monoxide.

FIG. 7 is a flowchart to explain another example of the method of adjusting the opening degree of each flow rate adjusting valve FC in the example of the carbon dioxide conversion method.

DETAILED DESCRIPTION

40 **[0005]** A carbon dioxide conversion device of an embodiment includes: a carbon dioxide supply part configured to supply a supply gas containing a carbon dioxide gas; a flow rate adjusting part that includes flow rate adjusting valves configured to distribute the supply gas to be supplied from the carbon dioxide supply part; an electrolysis part that includes electrolysis cell stacks configured to receive the supply gas distributed through the flow rate adjusting valves; a voltage
 45 measuring part that includes voltmeters configured to measure voltages across the electrolysis cell stacks; and a control part configured to adjust opening degrees of the flow rate adjusting valves in response to voltage signals to be received from the voltmeters.

[0006] 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
 50 some cases in the embodiments described below. The drawings are schematic, and a relationship between a thickness and a planar size, thickness proportions of respective portions, and the like are sometimes different from actual ones.

[0007] In this specification, "connecting" includes not only directly connecting but also indirectly connecting, unless otherwise specified. Furthermore, "connecting" herein includes not only physically connecting but also electrically connecting, unless otherwise specified.

55 **[0008]** An example configuration of a carbon dioxide conversion device of an embodiment is described. FIG. 1 is a schematic diagram illustrating the example configuration of the carbon dioxide conversion device of the embodiment. FIG. 1 illustrates a carbon dioxide conversion device 100.

[0009] The carbon dioxide conversion device 100 includes a carbon dioxide supply part (CO₂ supply part) 1, a flow rate

adjusting part 2, an electrolysis part 3, a voltage measuring part 4, a control part 5, a power supply 6, a cooler 7, a gas-liquid separator 8, a purifying part 9, a gas-liquid separator 10, a cooler 11, a gas-liquid separator 12, a water removal part (H₂O removal part) 13, a carbon dioxide recovery part (CO₂ recovery part) 14, a cooler 15, a tank 16, and a pump 17.

[0010] FIG. 2 is a diagram to further explain the example configuration of some of components illustrated in FIG. 1. FIG. 2 illustrates the carbon dioxide supply part 1, the flow rate adjusting part 2, the electrolysis part 3, the voltage measuring part 4, and the control part 5.

[0011] The carbon dioxide supply part 1 includes, for example, a carbon dioxide supply source SS. The carbon dioxide supply source SS can, for example, supply a supply gas containing carbon dioxide gas to the electrolysis part 3. The carbon dioxide supply part 1 is connected to the electrolysis part 3, for example, through the flow rate adjusting part 2. The carbon dioxide conversion device may have a humidifier to humidify the carbon dioxide gas from the carbon dioxide supply source SS and supply the gas humidified by the humidifier to the electrolysis part 3.

[0012] The carbon dioxide supply source SS can, for example, separate and recover carbon dioxide gas from an emission gas containing carbon dioxide gas (CO₂-containing gas) discharged from facilities such as thermal power plants, waste incineration plants, and steel plants, and supply the supply gas containing the separated carbon dioxide gas to the electrolysis part 3. In the carbon dioxide supply part 1, for example, a chemical absorption method using a chemical absorbent such as an amine solution, a physical absorption method using a physical absorbent 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 carbon dioxide separation membrane, a physical adsorption method using an inorganic substance such as zeolite as an adsorbent, a pressure swing adsorption (PSA) method, a thermal swing adsorption (TSA) method, and other methods can be used. For example, in the chemical absorption method and device using the amine solution, the emission gas is supplied to an absorption tower where the amine solution is sprayed, and the amine solution that has absorbed carbon dioxide is heated in a regeneration tower to recover carbon dioxide emitted from the amine solution. Various methods and devices capable of recovering carbon dioxide from the emission gas (CO₂-containing gas) can be applied to the carbon dioxide supply source SS.

[0013] The flow rate adjusting part 2 includes, for example, a flow rate adjusting valve FC. The flow rate adjusting valve FC can adjust a flow rate of the supply gas from the carbon dioxide supply source SS. The flow rate adjusting part 2 includes flow rate adjusting valves FC. The flow rate adjusting valves FC are provided, for example, in a middle of pipes that branch out on the way from the carbon dioxide supply part 1 to the electrolysis part 3 and can distribute the supply gas from the carbon dioxide supply part 1. Although FIG. 2 illustrates flow rate adjusting valves FC1, FC2, FC3, FC4, and FC5 as the flow rate adjusting valves FC, the number of the flow rate adjusting valves FC can be "n - 1" (n being a natural number of two or more) and is not limited to the number illustrated in FIG. 2.

[0014] The electrolysis part 3 includes, for example, an electrolysis cell stack CS. The electrolysis cell stack CS can receive, for example, the supply gas distributed from the flow rate adjusting part 2 and a supply liquid supplied from the tank 16 through the pump 17. The electrolysis cell stack CS can, for example, perform an electrolytic reaction using the supply gas and the supply liquid. The electrolysis part 3 includes electrolysis cell stacks CS. FIG. 2 illustrates electrolysis cell stacks CS1, CS2, CS3, CS4, CS5, and CS6 as the electrolysis cell stacks CS, but the number of the electrolysis cell stacks CS can be n (n being a natural number of two or more) and is not limited to the number illustrated in FIG. 2. The carbon dioxide conversion device may not have, for example, the flow rate adjusting valve FC corresponding to the n-th electrolysis cell stack CS. The n-th electrolysis cell stack CS may not be connected to any flow rate adjusting valve FC. The electrolysis cell stack CS1 is connected to the flow rate adjusting valve FC1. The electrolysis cell stack CS2 is connected to the flow rate adjusting valve FC2. The electrolysis cell stack CS3 is connected to the flow rate adjusting valve FC3. The electrolysis cell stack CS4 is connected to the flow rate adjusting valve FC4. The electrolysis cell stack CS5 is connected to the flow rate adjusting valve FC5. The electrolysis cell stack CS6 is connected to a pipe from the carbon dioxide supply part 1 to the electrolysis part 3 and is not connected to any flow rate adjusting valve FC.

[0015] FIG. 3 is a cross-sectional schematic diagram illustrating an example structure of the electrolysis cell stack CS. The electrolysis cell stack CS illustrated in FIG. 3 has a cathode 21, an anode 22, a membrane 23, and a flow path plate 24. FIG. 3 illustrates X, Y, and Z axes. The X, Y, and Z axes intersect each other perpendicularly. The Z axis is along a thickness direction of a carbon dioxide electrolysis cell 20. FIG. 3 illustrates a portion of an X-Z cross-section including the X and Z axes. FIG. 3 illustrates the cathodes 21, the anodes 22, the membranes 23, and the flow path plates 24.

[0016] The cathode 21, anode 22, and membrane 23 are stacked in the Z-axis direction, for example, to form the carbon dioxide electrolysis cell 20.

[0017] An example of the electrolysis cell stack CS includes the carbon dioxide electrolysis cell stack. The electrolysis cell stack CS has, for example, the carbon dioxide electrolysis cells 20. The carbon dioxide electrolysis cells 20 are stacked across the flow path plate 24. The carbon dioxide electrolysis cells 20 may, for example, be disposed between supporting members not illustrated. The number of stacks of the carbon dioxide electrolysis cells 20 is not limited, but is, for example, eight or more.

[0018] The cathode 21 is connected to the power supply 6, for example, through a cathode current collector provided at an end portion of the electrolysis cell stack CS. The cathode 21 can, for example, reduce carbon dioxide to produce carbon

compounds such as carbon monoxide. The cathode 21 further has a catalyst that, for example, promotes the electrolytic reaction. The catalyst is supported on a surface of a conductive substrate, for example, such as metal. The catalyst may form a catalyst layer provided on the surface of the conductive substrate.

[0019] The anode 22 is connected to the power supply 6, for example, through an anode current collector provided at the end portion of the electrolysis cell stack CS. The anode 22 can, for example, oxidize water to produce oxygen gas. The anode 22 further has a catalyst that, for example, promotes the electrolytic reaction. The catalyst is supported on a surface of a conductive substrate, for example, such as metal. The catalyst may form a catalyst layer on the surface of the conductive substrate.

[0020] The membrane 23 is disposed between the cathode 21 and anode 22, for example. The membrane 23 may be provided in contact with each of the cathode 21 and anode 22. The membrane 23 allows, for example, distribution of ions such as hydrogen ions (H^+), hydroxide ions (OH^-), carbonate ions (CO_3^{2-}), and hydrogen carbonate ions (HCO_3^-). The membrane 23 has, for example, at least an ion exchange membrane or a porous membrane.

[0021] The flow path plate 24 is provided to separate two carbon dioxide electrolysis cells 20. The flow path plate 24 can, for example, isolate an atmosphere between adjacent carbon dioxide electrolysis cells 20. The flow path plate 24 can be formed, for example, using a material such as metal, carbon material, or conductive ceramic.

[0022] The flow path plate 24 has flow paths 24a and flow paths 24b. Respective shapes of the flow paths 24a and 24b are not limited to those illustrated in FIG. 3. The respective shapes of the flow paths 24a and 24b may be serpentine or strip-shaped with respect to surfaces of the flow path plates 24 facing the cathode 21 and anode 22, respectively.

[0023] The flow path 24a is provided on the cathode 21 side surface of the flow path plate 24 and faces the cathode 21. The flow path 24a can, for example, distribute a fluid containing the supply gas or reduction products such as carbon compounds. The flow path 24a can, for example, form a cathode chamber. The flow path 24a has, for example, an inlet connected to the flow rate adjusting valve FC and an outlet from which a fluid containing carbon compounds such as carbon monoxide (cathode fluid) is discharged.

[0024] The flow path 24b is provided on the anode 22 side surface (opposite side of flow path 24a) of the flow path plate 24 and faces the anode 22. The flow path 24b can, for example, distribute a fluid containing the supply liquid, water and oxygen gas, or oxidation products such as carbon dioxide gas. The flow path 24b can, for example, form an anode chamber. The flow path 24b has, for example, an inlet connected to the pump 17 and an outlet from which a fluid containing oxygen (anode fluid) is discharged.

[0025] The supply liquid contains, for example, an electrolytic solution. Examples of the electrolytic solutions include a solution using water (H_2O), for example, an aqueous solution containing any electrolyte. Examples of the aqueous solutions containing electrolytes include aqueous solutions containing ions such as phosphate ions (PO_4^{2-}), borate ions (BO_3^{3-}), sodium ions (Na^+), potassium ions (K^+), calcium ions (Ca^{2+}), lithium ions (Li^+), cesium ions (Cs^+), magnesium ions (Mg^{2+}), chloride ions (Cl^-), hydrogen carbonate ions (HCO_3^-), carbonate ions (CO_3^{2-}), and hydroxide ions (OH^-). Examples of the electrolytic solutions include electrolytic solutions such as alkaline aqueous solutions in which compounds such as KOH, $KHCO_3$, and K_2CO_3 are dissolved.

[0026] The voltage measuring part 4 includes, for example, a voltmeter VM. The voltmeter VM can, for example, measure a voltage to be applied to the electrolysis cell stack CS during electrolysis operation. The voltage measuring part 4 can transmit a data signal indicating the voltage to be measured by the voltmeter VM to the control part 5. The voltage measuring part 4 includes voltmeters VM. FIG. 2 illustrates voltmeters VM1, VM2, VM3, VM4, VM5, and VM6 as the voltmeters VM, but the number of the voltmeters VM can be n (n is a natural number of 2 or more) and is not limited to the number illustrated in FIG. 2. The voltmeter VM may be provided for each electrolysis cell stack CS, for example.

[0027] The control part 5 has, for example, a controller CT. The controller CT can receive the data signals (voltage signals) indicating the measured voltages from the voltmeters VM. The controller CT can control opening degrees of the flow rate adjusting valves FC using the data of the measured voltages contained in the voltage signals. The controller CT can, for example, generate and output a control signal to control the opening degree of the flow rate adjusting valve FC according to the measured voltage. A dotted arrow illustrated in FIG. 2 indicates the control signal. The controller CT can, for example, generate control signals. Each control signal can control the opening degree of the flow rate adjusting valve FC corresponding to the voltage to be measured by each voltmeter VM. The controller CT is connected to each of the flow rate adjusting valves FC, for example, by a wired or wireless connection. The controller CT may be connected to at least one of the carbon dioxide supply source SS and the electrolysis cell stack CS by a wired or wireless connection and may control operation of at least one of the carbon dioxide supply source SS and the electrolysis cell stack CS.

[0028] The controller CT has hardware with an arithmetic device such as a processor, for example. Each operation may be stored as an operation program in a computer-readable recording medium, such as memory, and each operation may be executed by reading the operation program stored in the recording medium by the hardware as appropriate.

[0029] Next, the remaining components of the carbon dioxide conversion device 100 are further described with reference to FIG. 1.

[0030] The power supply 6 can, for example, supply load currents to the electrolysis cell stacks CS. The power supply 6 can, for example, be electrically connected to each of the cathode 21 and anode 22 of the electrolysis cell stack CS, and

can supply electric current to the electrolysis cell stack CS through the cathode current collector and anode current collector.

[0031] The cooler 7 can, for example, cool the cathode fluid discharged from the flow path 24a to a predetermined temperature. The cooler 7 may, for example, be connected to each of the flow paths 24a, or may be connected to a connection point of the flow paths 24a when the flow paths 24a are connected to each other to form a single cathode discharge flow path.

[0032] The gas-liquid separator 8 can, for example, process the cathode fluid to separate water from the cathode fluid. The gas-liquid separator 8 is connected, for example, to a subsequent stage of the cooler 7.

[0033] The purifying part 9 can, for example, separate carbon compounds such as carbon monoxide from the cathode fluid by purifying the cathode fluid processed by the gas-liquid separator 8. The purifying part 9 is connected, for example, to a subsequent stage of the gas-liquid separator 8.

[0034] The purifying part 9 can produce the cathode fluid using methods such as a pressure swing adsorption (CO-PSA) method using adsorbents such as, for example, a copper ion-zeolite adsorbent, a copper ion activated carbon adsorbent, a copper chloride-aluminum-crosslinked polystyrene solid adsorbent, a copper chloride-aluminum-activated carbon adsorbent, a cuprammonium cleaning method, a copper aluminum chloride complex solution absorption method, a copper chloride-aluminum-polystyrene polymer complex solution absorption method, a low-temperature method, and other methods. The purified cathode fluid is contained in a storage container, such as, for example, a not-illustrated tank, or sent to an organic-substance synthesis reactor, such as a methane synthesis reactor, an alcohol synthesizer, or a Fischer-Tropsch reactor, where, for example, carbon monoxide and hydrogen are reacted. These organic-substance synthesis reactors may be used in place of the purifying part 9, which applies the methods such as CO-PSA. After the cathode fluid is purified, purified residual gas is discharged from the purifying part 9. The purified residual gas may contain residual carbon monoxide, hydrogen, carbon dioxide, and other gases.

[0035] The gas-liquid separator 10 can process the anode fluid discharged from the flow path 24b to separate gas such as oxygen and carbon dioxide from the anode fluid. The gas-liquid separator 10 may be connected to each of the flow paths 24b, for example, or to a connection point of the flow paths 24b when the flow paths 24b are connected to each other to form a single anode discharge flow path.

[0036] The cooler 11 can, for example, cool a fluid containing gas such as oxygen and carbon dioxide separated from the anode fluid by the gas-liquid separator 10 to a predetermined temperature. The cooler 11 is connected, for example, to a subsequent stage of the gas-liquid separator 10.

[0037] The gas-liquid separator 12 can, for example, process the fluid cooled by the cooler 11 to separate oxygen gas from the cooled fluid. The fluid processed by the gas-liquid separator 12 may be supplied to the tank 16.

[0038] The water removal part 13 can, for example, process the fluid containing oxygen gas and carbon dioxide separated by the gas-liquid separator 12 to remove water remaining in the fluid. The water removal part 13 is connected, for example, to a subsequent stage of the gas-liquid separator 12. The water removal part 13 has, for example, a water vapor-removing polymer membrane, or a water vapor adsorbent such as zeolite, silica gel, mesoporous silica, or activated carbon.

[0039] The carbon dioxide recovery part 14 can, for example, process the fluid processed by the water removal part 13 to remove carbon dioxide contained in the fluid. This allows carbon dioxide to be recovered. The fluid processed by the carbon dioxide recovery part 14 may contain oxygen. The carbon dioxide recovery part 14 is connected, for example, to a subsequent stage of the water removal part 13. The carbon dioxide recovery part 14 has, for example, a carbon dioxide adsorbent. Examples of the carbon dioxide adsorbents include primary or secondary amine carriers, FAU-type zeolites, GIS-type zeolites, using 1,4-benzenedicarboxylic acid as an organic ligand and isonicotinic acid as an auxiliary ligand, Co-MOF (metal-organic frameworks) obtained by reaction with cobalt nitrate, ZIF-69 (CAS No. 1018477-10-5, chemical formula: $C_{10}H_6N_5O_2ClZn$), Cubic $[Zn_4O(piperazine\ dicarbamate)_3]$ and other materials can be used to separate and recover carbon dioxide using the pressure swing adsorption (PSA) method or the temperature swing adsorption (TSA) method, depending on the adsorbent.

[0040] The carbon dioxide recovery part 14 may use an organic semiconductor such as tetracyanoquinodimethane (CAS No. 1518-16-7, chemical formula: $(NC)_2CC_6H_4C(CN)_2$) pore material as an oxygen adsorbent and use the pressure swing adsorption (PSA) method or temperature swing adsorption (TSA) method to separate and recover oxygen to consequently recover carbon dioxide. The carbon dioxide recovery part 14 may use a Faradaic electro-swing reactive adsorption method while using poly-1,4-anthraquinone as a carbon dioxide adsorption electrode and polyvinylferrocene as a counter electrode.

[0041] The cooler 15 can, for example, cool the anode fluid processed by the gas-liquid separator 10 to a predetermined temperature. The cooler 15 is connected, for example, to a subsequent stage of the gas-liquid separator 10.

[0042] The tank 16 can, for example, contain the anode fluid cooled by the cooler 15. The anode fluid cooled by the cooler 15 contains, for example, water and the electrolytic solution. The tank 16 is connected, for example, to a subsequent stage of the cooler 15. The tank 16 may be connected to a makeup tank. By receiving water from the makeup tank, the anode fluid contained in the tank may be replenished with water.

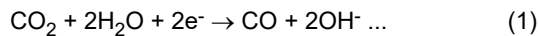
[0043] The pump 17 can, for example, supply the anode fluid contained in the tank 16 to each of the flow paths 24b of the electrolysis part 3 as a supply liquid. The pump 17 is provided, for example, in a middle of a circulation flow path connecting the tank 16 to the inlet of each flow path 24b.

[0044] The carbon dioxide conversion device illustrated in FIG. 1 may have blowers, pumps, compressors, and other devices as appropriate, and these devices may control the flow of gas, liquids, and other fluids.

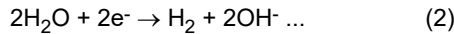
[0045] Next, an example of a carbon dioxide conversion method using the carbon dioxide conversion device having the configuration illustrated in FIG. 1 and FIG. 2 will be described. Here, an example of producing carbon monoxide as the carbon compound is described.

[0046] In the example of the carbon dioxide conversion method, the supply gas containing carbon dioxide gas distributed by the flow rate adjusting part 2 from the carbon dioxide supply part 1 is supplied to each flow path 24a through the inlet of each flow path 24a, the supply liquid containing the electrolytic solution is supplied to each flow path 24b through the inlet of each flow path 24b from the tank 16 using the pump 17, and electric current is supplied from the power supply 6 in each electrolysis cell stack CS. This allows the electrolytic reaction to take place at each of the cathode 21 and anode 22.

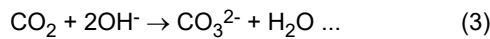
[0047] Carbon dioxide supplied to each flow path 24a is converted to carbon monoxide and hydroxide ions by the electrolytic reaction at the cathode 21. This reaction is expressed by Formula (1) below.



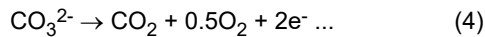
[0048] Water in the electrolytic solution is converted to hydrogen and hydroxide ions through the electrolytic reaction at the cathode 21. This reaction is expressed by Formula (2) below.



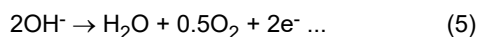
[0049] The hydroxide ions produced by the electrolytic reaction expressed by Formula (2) react with carbon dioxide to produce carbonate ions and water. This reaction is expressed by Formula (3) below.



[0050] The carbonate ions produced are converted to carbon dioxide and oxygen by the electrolytic reaction at the anode 22. This reaction is expressed by Formula (4) below.



[0051] Unreacted hydroxide ions that are not used in the reaction to produce carbonate ions, expressed by Formula (3), are converted to water and oxygen by the electrolytic reaction at the anode 22. This reaction is expressed by Formula (5) below.



[0052] Carbon monoxide and hydrogen produced at the cathode 21, along with unreacted carbon dioxide, are discharged from each flow path 24a through the outlet of each flow path 24a as the cathode fluid.

[0053] Carbon dioxide, oxygen, and water produced in the anode 22 are discharged from each flow path 24b through the outlet of each flow path 24b as the anode fluid along with the electrolytic solution.

[0054] Assuming that load currents through the electrolysis cell stacks CS are equal to each other, the flow rate FR_{CO_2} (mol/s) of the supply gas containing carbon dioxide gas supplied from the carbon dioxide supply part 1 and the load current I (A) through the electrolysis cell stack CS are controlled to satisfy the relationship expressed by Equality (6) below.

$$\text{FR}_{\text{CO}_2} = mnIU_{\text{CO}_2}/F \dots (6)$$

[0055] In Equality (6), "m" represents the number of stacked carbon dioxide electrolysis cells 20 in the electrolysis cell stack CS, "n" represents the number of the electrolysis cell stacks CS, " U_{CO_2} " represents a theoretical flow rate ratio of the supply gas containing carbon dioxide gas, and "F" represents a Faraday constant (96485 C/mol). According to the flow rate $\text{FR}_{\text{CO}_2}/n$ of the supply gas to one electrolysis cell stack CS, the opening degree of the corresponding flow rate adjusting valve FC is set to a set initial value. The flow rate FR_{CO_2} according to the sum of the load currents through the electrolysis cell stacks CS or the load current I with a theoretical current value according to the flow rate of the supply gas is given as a set value. For example, the supply gas with the flow rate according to the sum of the load currents through the electrolysis

cell stacks CS is distributed and supplied to the electrolysis cell stacks, or an equal load current with the theoretical current value according to the flow rate of the supply gas supplied from the carbon dioxide supply part 1 is supplied to the electrolysis cell stacks CS. The carbon dioxide supply part 1 has a function to maintain the set value of the flow rate FR_{CO_2} . The power supply 6 has a function to maintain the set value of the load current I. When the set value is changed, the change is made in a preset time and the opening degree of each flow rate adjusting valve FC is changed according to the new set value.

[0056] FIG. 4 is a flowchart to explain an example of a method of adjusting the opening degree of each flow rate adjusting valve FC in the example of the carbon dioxide conversion method. As illustrated in FIG. 4, a voltage V_i of each electrolysis cell stack CS is measured by the corresponding voltmeter VM to generate a voltage signal (data signal), and a mean voltage of the voltages V_i applied to the electrolysis cell stacks CS (mean stack voltage) V_{mean} is calculated from the data indicating the measured voltage V_i contained in the generated voltage signal (Step S1). For example, the voltage V_i of the electrolysis cell stack CS1 is measured by a voltmeter V1, the voltage V_i of the electrolysis cell stack CS2 is measured by a voltmeter V2, and the voltages V_i of the electrolysis cell stacks CS3, CS4, and CS5 are measured by voltmeters V3, V4, and V5, respectively. The voltage measuring part 4 may measure the voltage V_i across at least one electrolysis cell stack CS in sequence, or the voltages V_i across the electrolysis cell stacks CS simultaneously. The mean voltage V_{mean} may be calculated, for example, by the control part 5 upon receiving the voltage signal, or by an arithmetic device such as a personal computer connected to the control part 5. The mean voltage V_{mean} is determined by dividing the sum of the voltages V_i by the number of the electrolysis cell stacks CS and is expressed, for example, by Equality (7) below.

$$V_{mean} = \sum V_i / n \dots (7)$$

[0057] Next, "1" is entered ($i = 1$) in the numerical value i representing the target electrolysis cell stack CS in the control part 5 (Step S2). This indicates that the target electrolysis cell stack CS is the electrolysis cell stack CS1 in the first stack.

[0058] Next, the voltage V_i across the target electrolysis cell stack CS is compared with the mean voltage V_{mean} (Step S3). When $i=1$, the voltage V_i of the electrolysis cell stack CS1 is compared with the mean voltage V_{mean} . The above comparison is performed, for example, by calculating a difference ($V_i - V_{mean}$) between the voltage V_i and the mean voltage V_{mean} . The above calculation may be performed, for example, by the control part 5 or by an arithmetic device such as a personal computer connected to the control part 5.

[0059] When the value of the voltage V_i is greater (higher) than the value of the mean voltage V_{mean} (S3: $V_i > V_{mean}$), the opening degree of the flow rate adjusting valve FC corresponding to the target electrolysis cell stack CS is increased (Step S4-1). This allows the flow rate of the supply gas supplied to the target electrolysis cell stack CS to be increased. For example, when the voltage V_i across the electrolysis cell stack CS1 is greater than the mean voltage V_{mean} , the flow rate of the supply gas supplied to the electrolysis cell stack CS1 can be increased by increasing the opening degree of the flow rate adjusting valve FC1. The amount of increase of the supply gas is, for example, set appropriately so that the supply gas supplied to the target electrolysis cell stack CS is used without excess or deficiency.

[0060] When the value of the voltage V_i is smaller (lower) than the mean voltage V_{mean} (S3: $V_i < V_{mean}$), the opening degree of the flow rate adjusting valve FC corresponding to the target electrolysis cell stack CS is decreased (step S4-2). This allows the flow rate of the supply gas supplied to the target electrolysis cell stack CS to be decreased. For example, when the voltage V_i across the electrolysis cell stack CS1 is smaller than the mean voltage V_{mean} , the flow rate of the supply gas supplied to the electrolysis cell stack CS1 can be decreased by decreasing the opening degree of the flow rate adjusting valve FC1. The amount of decrease of the supply gas is, for example, set appropriately so that the supply gas supplied to the target electrolysis cell stack CS is used without excess or deficiency.

[0061] When the value of the voltage V_i is equal to the mean voltage V_{mean} (S3: $V_i = V_{mean}$), the opening degree of the flow rate adjusting valve FC corresponding to the target electrolysis cell stack CS is not changed (step S4-3). For example, when the voltage V_i of the electrolysis cell stack CS1 is equal to the mean voltage V_{mean} , the opening degree of the flow rate adjusting valve FC1 is maintained unchanged.

[0062] FIG. 5 is a diagram illustrating a relationship between the rate of increase in the flow rate of the supply gas and the voltage V_i . FIG. 5 illustrates the relationship between the rate of increase in the flow rate of the supply gas for each of one electrolysis cell stack CS_A and one other electrolysis cell stack CS_B and the voltage V_i . The voltage V_i is a normalized value, and the value of the voltage V_i based on the specification of the electrolysis cell stack CS is set as 1.000.

[0063] As illustrated in FIG. 5, the electrolysis cell stack CS_A , in which the voltage V_i increases as the flow rate of the supply gas increases, preferably decreases the opening degree of the corresponding flow rate adjusting valve FC to decrease the flow rate of the supply gas until the value of the voltage V_i reaches a reference value. The reference value is, for example, 1.000. As illustrated in FIG. 5, the electrolysis cell stack CS_B , in which the voltage V_i decreases as the flow rate of the supply gas increases, preferably decreases the opening degree of the corresponding flow rate adjusting valve FC to decrease the flow rate of the supply gas until the value of the voltage V_i reaches the reference value.

[0064] FIG. 6 is a diagram illustrating a relationship between the rate of increase in the flow rate of the supply gas and the

amount of carbon monoxide produced. FIG. 6 illustrates the relationship between the rate of increase in the flow rate of the supply gas for each of one electrolysis cell stack CS_A and one other electrolysis cell stack CS_B and the amount of carbon monoxide produced. The amount of carbon monoxide produced is a normalized value, and the value of carbon monoxide produced based on the specification of the electrolysis cell stack CS is set as 1.000.

[0065] As illustrated in FIG. 6, the electrolysis cell stack CS_A , in which the amount of carbon monoxide produced increases as the flow rate of the supply gas increases, preferably decreases the opening degree of the corresponding flow rate adjusting valve FC to decrease the flow rate of the supply gas until the amount of carbon monoxide produced reaches a reference value. The reference value is, for example, 1.000. As illustrated in FIG. 6, the electrolysis cell stack CS_B , in which the amount of carbon monoxide produced decreases as the flow rate of the supply gas increases, preferably decreases the opening degree of the corresponding flow rate adjusting valve FC to decrease the flow rate of the supply gas until the amount of carbon monoxide produced reaches the reference value.

[0066] Next, "1" is added to the numerical value i ($i = i + 1$) in the control part 5 (Step S5). This indicates that the target electrolysis cell stack CS transfers to the electrolysis cell stack CS in the next stack.

[0067] Next, it is determined whether $i = n$, that is, whether the target electrolysis cell stack CS is the n -th electrolysis cell stack CS (Step S6). This determination may be made, for example, by the control part 5 or by an arithmetic device such as a personal computer connected to the control part 5.

[0068] When $i \neq n$ (S6: NO), return to Step S3 and the same steps as for the electrolysis cell stack CS1 are performed for the next target electrolysis cell stack CS. For example, after the operations from Step S1 to Step S3 are performed in the electrolysis cell stack CS1, the operations from Step S1 to Step S3 are performed in the next electrolysis cell stack CS2. These operations are repeated until the " $n - 1$ "th electrolysis cell stack CS_{n-1} .

[0069] When $i = n$ (S6: YES), the process is complete because it indicates the completion of the adjustment. The above is an example of the carbon dioxide conversion method. The adjustment of the opening degree of each flow rate adjusting valve FC illustrated in FIG. 4 may be performed periodically at predetermined time intervals.

[0070] In the carbon dioxide conversion device of the embodiment, the flow rate of the supply gas supplied to the electrolysis cell stacks CS can be adjusted by adjusting the opening degree of the corresponding flow rate adjusting valve FC according to the voltage across the electrolysis cell stack CS. Therefore, the carbon dioxide gas can be supplied to the electrolysis cell stacks CS without excess or deficiency, thereby improving utilization efficiency of carbon dioxide in the electrolysis cell stacks CS.

[0071] The method of adjusting the opening degree of the flow rate adjusting valve FC is not limited to the method represented by the flowchart illustrated in FIG. 4. FIG. 7 is a flowchart to explain another example of the method of adjusting the opening degree of each flow rate adjusting valve FC in the example of the carbon dioxide conversion method. The flowchart illustrated in FIG. 7 differs from the flowchart illustrated in FIG. 4 in that it has Step S7 instead of Steps S3 and S4 (S4-1, S4-2, S4-3). Hereinafter, the parts of the flowchart that differ from the flowchart illustrated in FIG. 4 in another example of the method of adjusting the opening degree of each flow rate adjusting valve FC will be described, and for the other parts, the description of the flowchart illustrated in FIG. 4 can be used as appropriate.

[0072] In Step S7, when the flow rate adjusting valve FC has linear characteristics (CV value is proportional to a valve opening degree), the opening degree of the flow rate adjusting valve FC is adjusted in each electrolysis cell stack CS to be proportional to a ratio V_i/V_{mean} of the voltage V_i to the mean voltage V_{mean} (opening degree * V_i/V_{mean}). This operation is repeated until the " $n - 1$ "th electrolysis cell stack CS_{n-1} . The flow rate of the supply gas supplied to the electrolysis cell stacks CS can be thereby adjusted. The flow rate of the supply gas supplied to the electrolysis cell stack CS_n can be adjusted indirectly by adjusting the opening degrees of the flow rate adjusting valves FC corresponding to all other electrolysis cell stacks CS. Thus, carbon dioxide gas can be supplied to the electrolysis cell stacks CS without excess or deficiency, thereby improving the utilization efficiency of carbon dioxide in the electrolysis cell stacks CS.

[0073] Information indicating the relationship between the load current I and the opening degree corresponding to each flow rate adjusting valve FC may be updated. The update may take place, for example, periodically or after the flow rate has stabilized, or at a stage when the difference from the information stored in the control part 5 exceeds a set value.

[0074] The carbon dioxide conversion device may further have an alarm. The alarm can generate an alarm after the voltage comparison (Step S3) and before adjusting the opening degree of the flow rate adjusting valve FC when the difference between the voltage V_i of the target electrolysis cell stack CS and the mean voltage V_{mean} is outside an acceptable range. In response to the alarm, the operation of the target electrolysis cell stack CS may be stopped. The electrolysis cell stack CS can be stopped in operation by, for example, stopping the supply of the supply gas and the supply liquid, and the supply of electric current. The alarm may, for example, be connected to the control part 5 and controlled by the control part 5.

[0075] In the embodiment, the example in which the number of flow rate adjusting valves FC is " $n - 1$ " (n is a natural number greater than or equal to 2) and the number of electrolysis cell stacks CS is " n " is described. However, the number of each of the flow rate adjusting valves FC and electrolysis cell stacks CS provided in the carbon dioxide conversion device is not limited thereto. For example, the carbon dioxide conversion device may be equipped with any number of flow rate adjusting valves FC and any number of electrolysis cell stacks CS, such as having " n " or more flow rate adjusting valves FC

and "n + 1" or more electrolysis cell stacks CS.

[0076] 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 part configured to supply a supply gas containing a carbon dioxide gas;
a flow rate adjusting part that includes flow rate adjusting valves configured to distribute the supply gas to be supplied from the carbon dioxide supply part;
an electrolysis part that includes electrolysis cell stacks configured to receive the supply gas distributed through the flow rate adjusting valves;
a voltage measuring part that includes voltmeters configured to measure voltages across the electrolysis cell stacks; and
a control part configured to adjust opening degrees of the flow rate adjusting valves in response to voltage signals to be received from the voltmeters.

2. The device according to claim 1, wherein

an equal load current is supplied to each of the electrolysis cell stacks, the equal load current having a theoretical current value according to the flow rate of the supply gas to be supplied from the carbon dioxide supply part.

3. The device according to claim 1, wherein

the supply gas is supplied from the carbon dioxide supply part, the supply gas having a flow rate according to the sum of the load currents through the electrolysis cell stacks.

4. The device according to any one of claims 1 to 3, wherein

the control part is configured to calculate a mean voltage of the voltages,
when one of the voltages is lower than the mean voltage, an opening degree of one of the flow rate adjusting valves corresponding to the one of the voltages is decreased, and
when one of the voltages is higher than the mean voltage, the opening degree of one of the flow rate adjusting valves corresponding to the one of the voltages is increased.

5. The device according to any one of claims 1 to 3, wherein

the control part is configured to calculate a mean voltage of the voltages and adjust an opening degree of one of the flow rate adjusting valves corresponding to be proportional to a ratio of one of the voltages to the mean voltage.

6. A carbon dioxide conversion method, comprising:

distributing a supply gas containing carbon dioxide gas through flow rate adjusting valves to supply to electrolysis cell stacks, and supplying load currents to the electrolysis cell stacks; and
measuring voltages across the electrolysis cell stacks and adjusting opening degrees of the flow rate adjusting valves according to the voltages.

7. The method according to claim 6, further comprising:

supplying an equal load current to each of the electrolysis cell stacks, the equal load current having a theoretical current value according to a flow rate of the supply gas.

8. The method according to claim 6, further comprising:

distributing the supply gas to supply to the electrolysis cell stacks, the supply gas having a flow rate according to the sum of the load currents through the electrolysis cell stacks.

9. The method according to any one of claims 6 to 8, further comprising:

calculating a mean voltage of the voltages;

decreasing an opening degree of one of the flow rate adjusting valves corresponding to one of the voltages when
the one of the voltages is lower than the mean voltage; and

increasing the opening degree of one of the flow rate adjusting valves corresponding to one of the voltages when
the one of the voltages is higher than the mean voltage.

10. The method according to any one of claims 6 to 8, further comprising:

calculating a mean voltage of the voltages; and

adjusting an opening degree of one of the flow rate adjusting valves corresponding to be proportional to a ratio of
one of the voltages to the mean voltage.

FIG. 1

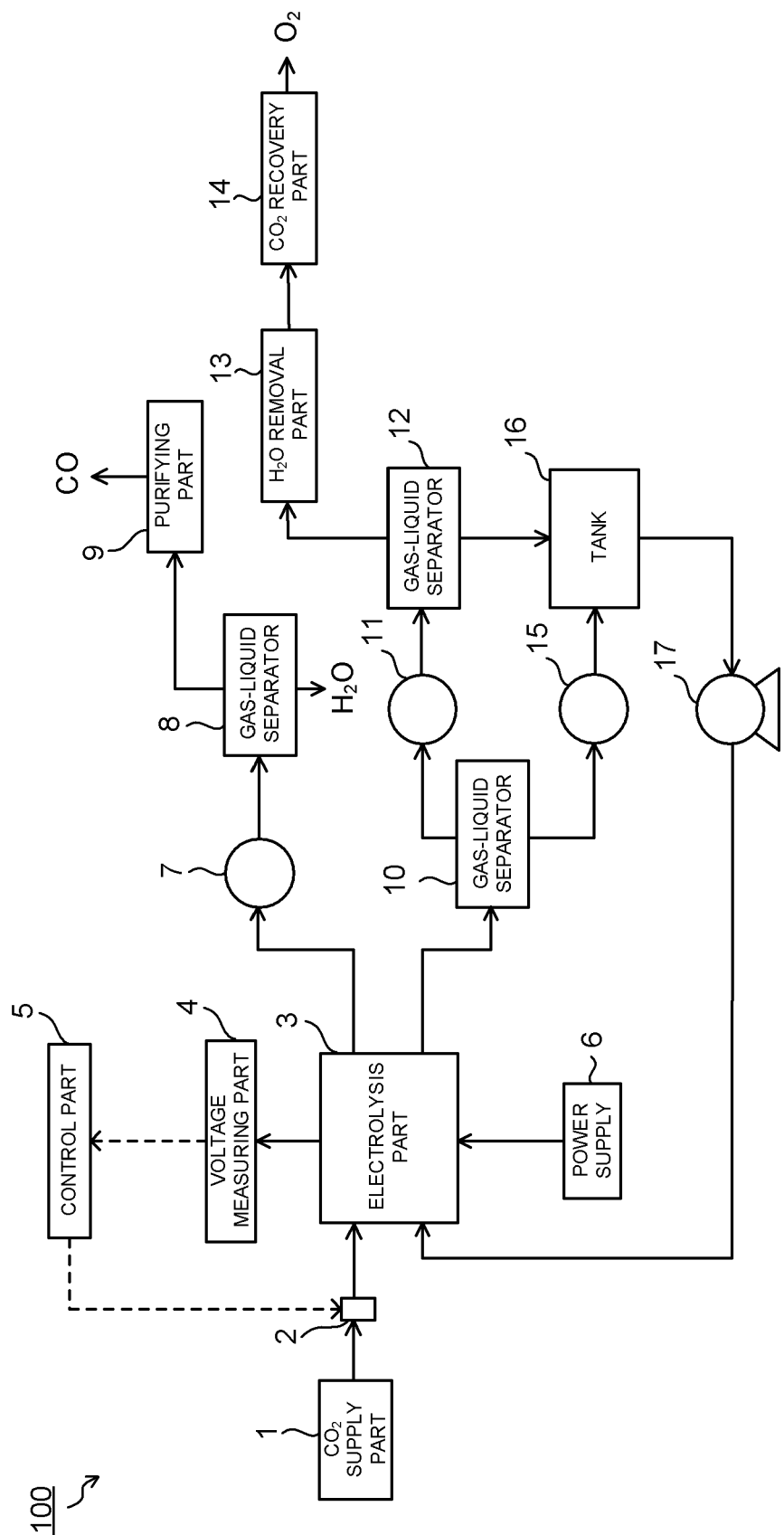


FIG. 2

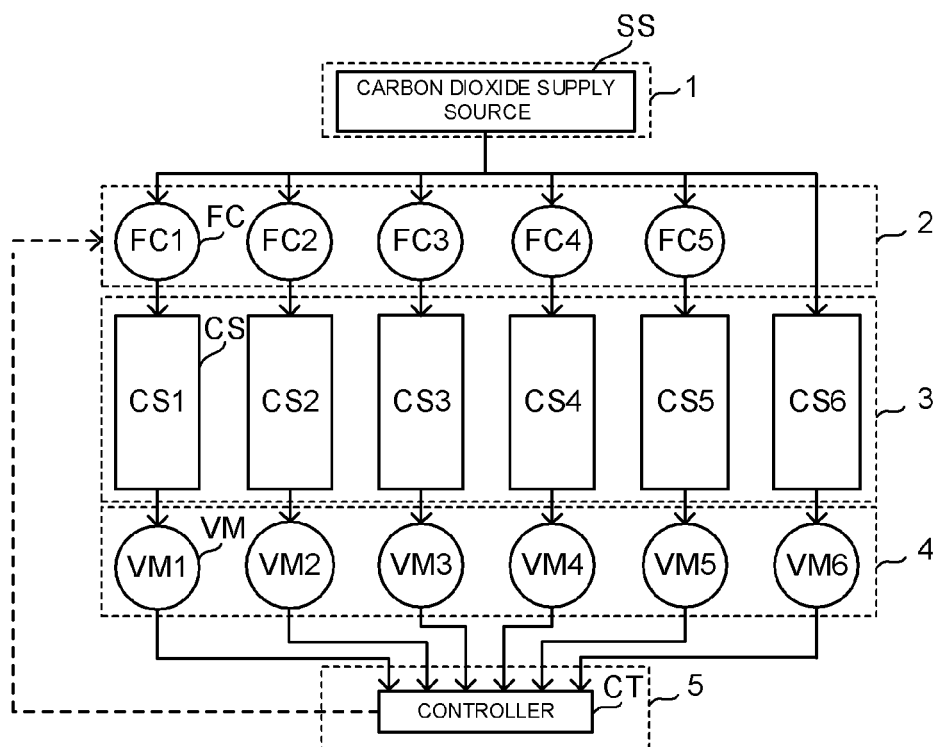


FIG. 3

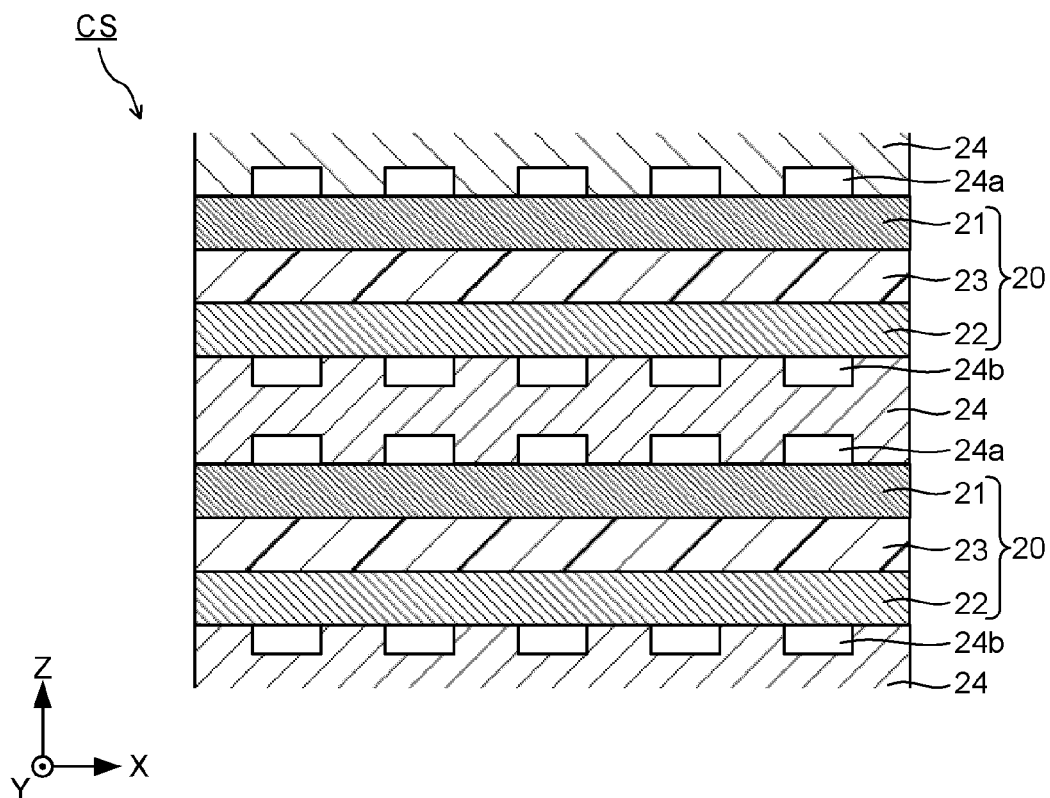


FIG. 4

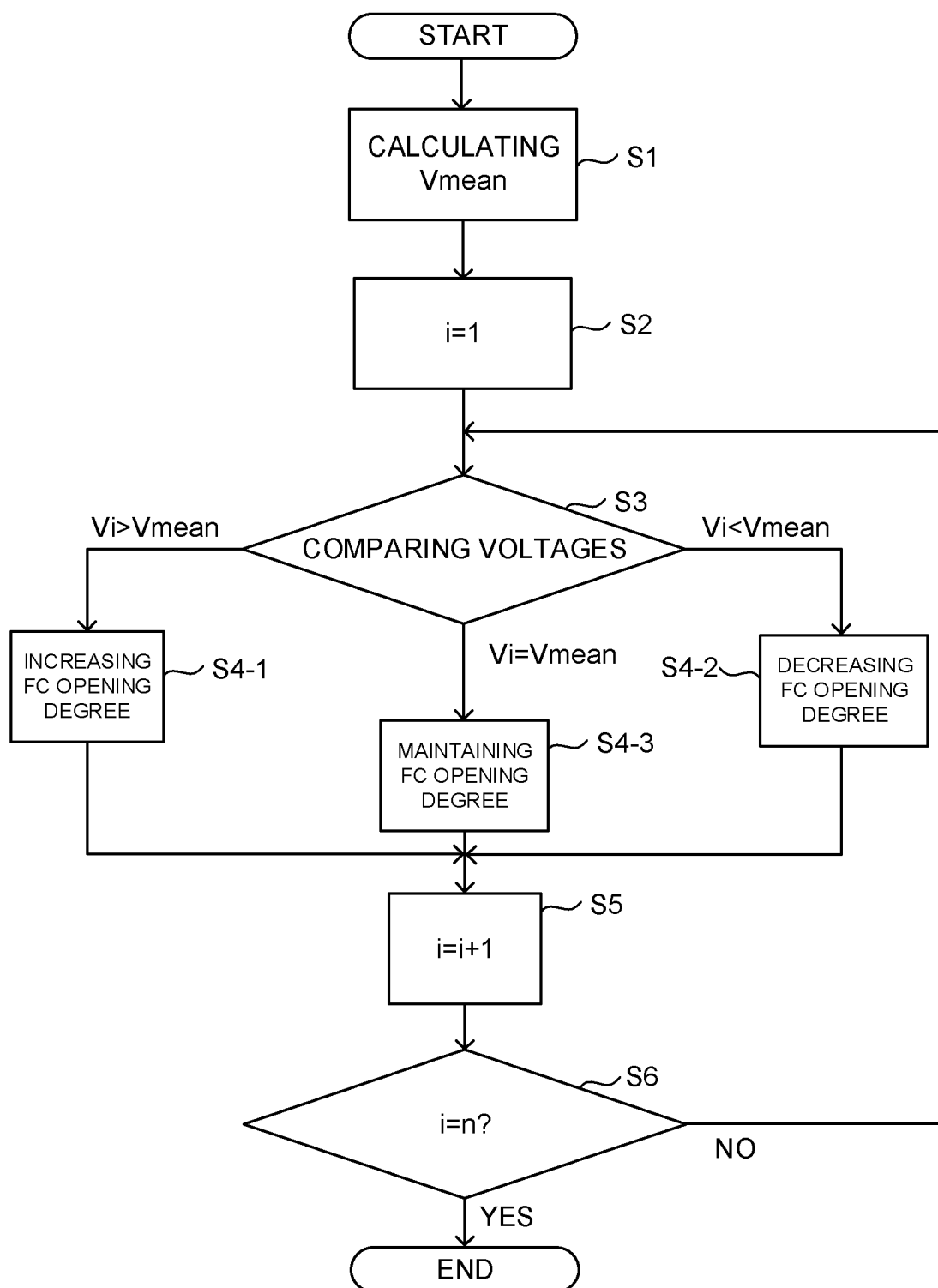


FIG. 5

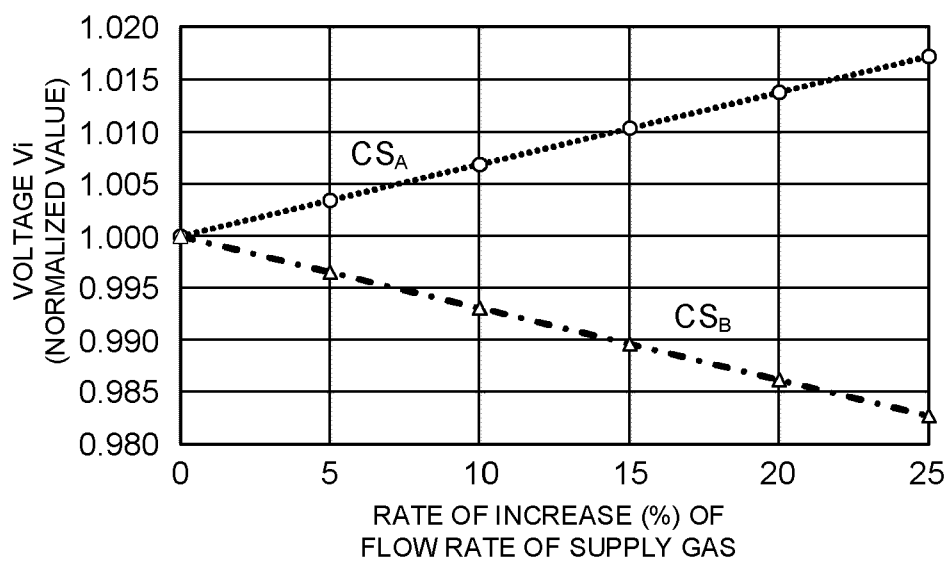


FIG. 6

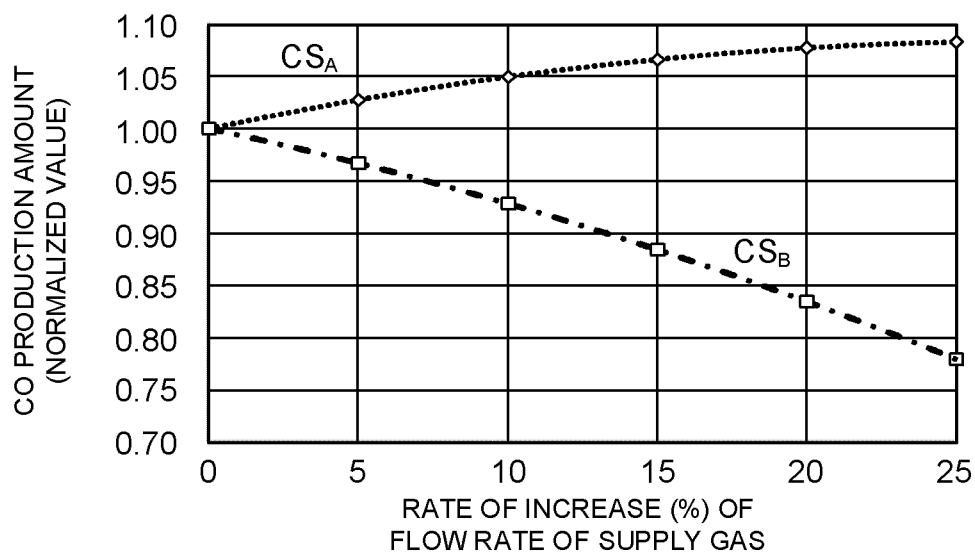
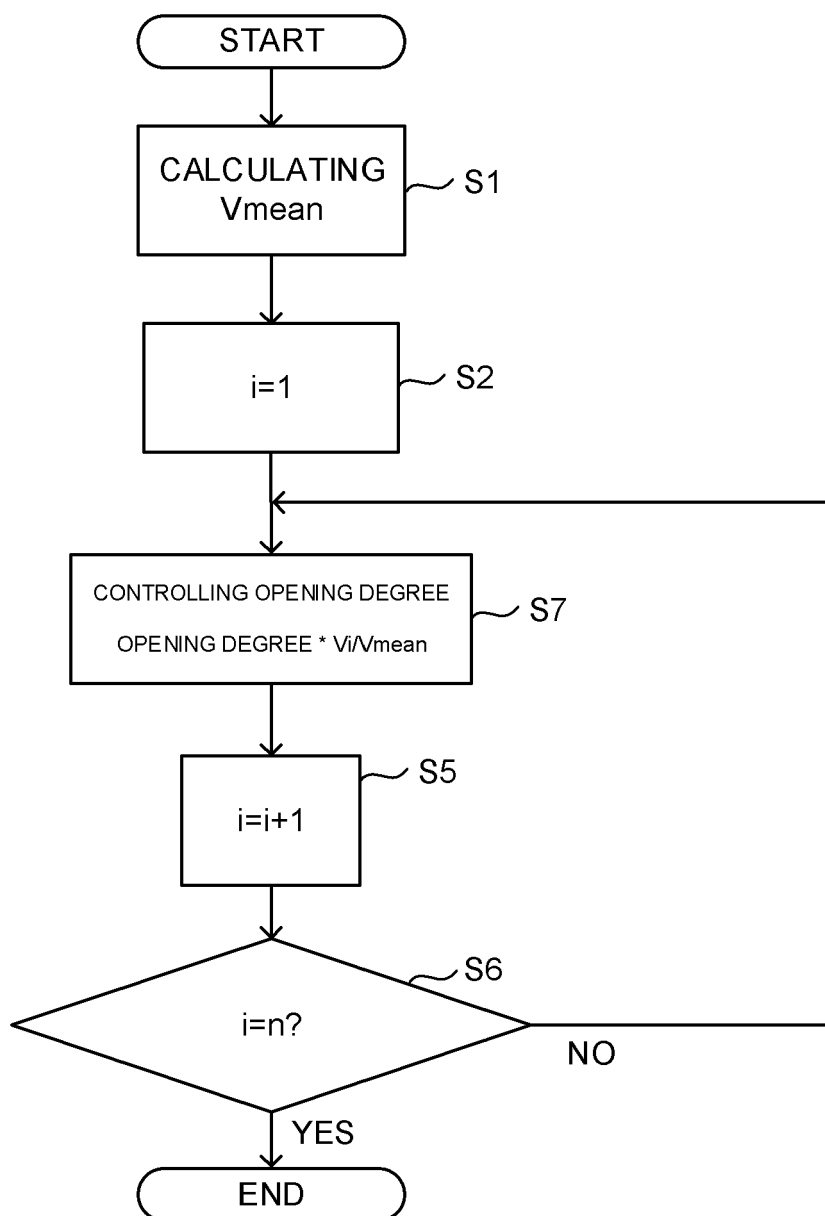


FIG. 7





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The present search report has been drawn up for all claims			
Place of search		Date of completion of the search	Examiner
Munich		24 January 2025	Perednis, Dainius
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