

DEVICE AND METHOD FOR PRODUCING SYNTHETIC GAS

TECHNICAL FIELD OF THE INVENTION

The present invention relates to a device and method for producing synthetic gas. It applies, in particular, to the field of producing synthetic natural gas or "SNG" (Synthetic Natural Gas).

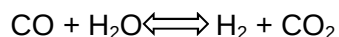
STATE OF THE ART

Biomethane, which is an "SNG" (Synthetic Natural Gas), can be produced by utilizing Power-to-SNG (abbreviated to "P2G") methods, designed to convert water, CO₂ and electrical energy into a combustible gas of a quality similar to that of natural gas.

To achieve a satisfactory economic return, P2G is based on using electrical energy from a renewable source during periods when there is overproduction of this energy. Consequently, the methanation technology requires great operational flexibility in terms of power, or flow rate, and processing.

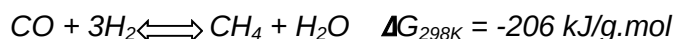
Several technological options exist within this sector.

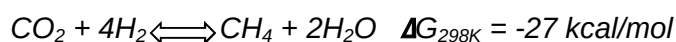
The first option consists of directly co-electrolyzing water, in the form of vapor, and anthropogenic CO₂ to produce CO and H₂. This conversion generally takes place at high temperature, and the gas produced is a syngas whose compositional characteristics are close to those from biomass gasification (CO, H₂, CO₂ and H₂O). The main difference lies in better control of the H₂/CO ratio without a gas-to-water reaction step, abbreviated to "WGS" (for Water Gas Shift) and referred to as the "Dussan reaction". The formula for this WGS reaction is



The second option consists of converting the water by alkaline or polymer electrolyte membrane electrolysis to produce H₂ at the cathode and O₂ at the anode. The H₂ produced in this way is then mixed with CO₂ to form syngas, which is the source of the synthetic natural gas.

The production of biomethane from the syngas is based on the catalytic methanation (or hydrogenation) reaction of the CO or CO₂, called the "Sabatier reaction". Methanation consists of converting the carbon monoxide or dioxide in the presence of hydrogen and a catalyst, generally based on nickel or any other more or less noble transition metal, to produce methane. It is governed by the following competitively balanced hydrogenation reactions:





To produce SNG from syngas, the first option utilizes a CO methanation reaction whereas the second option utilizes a CO₂ methanation reaction.

The methanation reaction is an exothermic reaction with a reduction in the number of moles; according to Le Chatelier's principle, the reaction is encouraged by increasing the pressure and discouraged by increasing the temperature.

These highly exothermic balanced reactions require good control of the cooling of the reactor in which they take place.

The heat generated during CO conversion is approximately 2.7 kWh during the production of 1 Nm³ of methane. Controlling the temperature inside the reactor, and therefore removing the heat produced by the reaction, is one of the keys to minimizing the deactivation of the catalyst, by sintering, and maximizing the methane conversion rate.

Profitable reuse of the heat produced during methanation, either within the actual unit or by selling heat, is one of the keys to the technical and economic balance of the SNG production method. The production of steam is a conventional way to obtain this reuse.

The methanation reactions, which have rapid kinetics at the temperatures utilized, are characterized by very high exothermicity.

To maximize the production of CH₄ by carbon dioxide hydrogenation, and minimize the excess CO₂, the H₂ and CO₂ should have a stoichiometric ratio of about 4:1. Even by respecting to this ratio, the reaction remains incomplete because of the chemical balances.

Among the methanation reactor technologies, some utilize a dense fluidized-bed reactor, the bed being formed by the methanation reaction catalyst. The heat produced by the reaction is therefore removed by exchangers immersed in the fluidized bed. However, because of the very high exothermicity of the reaction, the amount of heat to be removed, and therefore the exchange surface areas required, is very great. Therefore, the volume occupied by this exchanger leads to an overall over-dimensioning of the reactor size, and above all to making its design more complex.

The utilization of a fluidized bed is a simple solution for limiting the reaction temperature. Fluidization of the catalyst by the reactant mixture enables almost perfect homogenization of the temperatures at all points of the catalyst layer and the reactor can be assimilated to an isothermal reactor. The removal of the heat produced by the reaction is achieved by means of exchangers immersed within the fluidized bed. The thermal exchange coefficients between the fluidized layer and a wall immersed into the bed are very high (of the order of 400 to 600 watts per kelvin per square meter, expressed as W/K.m², comparable to those between a liquid and a wall) and make it possible to minimize the dimensions of the exchanger, and therefore the overall size of the reactor.

In the temperature range utilized in the fluidized-bed methanation reactors, the kinetics of the methanation reaction are very rapid and, as a result, the amount of catalyst required just for the chemical reaction is small. Consequently, the size of the reactor and the amount of catalyst used stem from the overall dimensions of the exchanger installed within the fluidized bed and from the required transfer surface area.

Because of the thermal exchange and the fluidization regime, a major drawback is attributable to this technology for high-pressure operation. The reduction in the gas volume due to increased pressure leads to a smaller cross-sectional area available for positioning the exchanger (for equivalent power). However, solutions of the person skilled in the art exist for overcoming the adjustment of the effective surface area or the fluidization regime. For example, the non-exhaustive solutions are:

- reducing the number of tubes and, as a result, increasing the height of the catalyst layer, with a height limit linked to the slugging phenomenon with reduced thermal exchange properties, slugging being the movement of solids in packets with, between two packets, a gaseous pocket that fills the entire cross-sectional area of a reactor, which has the effect of producing an alternation between solid and gaseous packets instead of a mixture of gas and solids, as expected; and

- modifying the physical characteristics of the catalyst (particle size, density of the support) to preserve an equivalent fluidization for a lower volumetric flow rate.

To optimize operational flexibility, the fluidized bed naturally allows greater flexibility in terms of reactor flow rate and therefore power with regard to sizing conditions.

The solutions currently proposed for this technological family are not really differentiated from each other by conversion efficiency, but mainly by the methodology utilized to cool the reactor.

The cooling principles in the context of CO₂ methanation are currently the same as for CO methanation when the conversion is carried out in an "isothermal" reactor. In addition, even with an adjustment of the H₂/CO₂ reagents to the stoichiometric ratio of 4:1, the SNG produced still contains H₂, CO₂ and water, which need to be removed in order to meet the specifications for injection into the natural gas grid. The overall schematic diagram for current P2G systems therefore comprises the following steps:

- electrolysis of the water;
- methanation of the CO₂ and hydrogen produced by the electrolysis; and
- adjustment to specifications, to separate the H₂O and residual H₂/CO₂.

The function of the adjustment to specifications step is to separate the constituents of the gas produced by methanation in order to obtain a biomethane meeting the specifications for injection into the natural gas grid. This separation therefore generates the sub-products

H₂O, CO₂ and H₂. It is usually carried out in separate equipment with sometimes very different operating conditions.

Before the SNG produced is processed and injected, a significant fraction of the residual H₂ from the methanation reaction has to be removed in order to meet specifications.

- 5 The technique used most frequently for this separation is membrane permeation, which can present a non-trivial level of complexity and costs, in terms of capital expenditure and operation, with a considerable impact on the value chain.

The composition of the raw SNG on output from the reactor is closely related to the operating conditions of the reactor, in terms of pressure, temperature, adiabatic or isothermal
10 operating mode of the reactor, these conditions governing the chemical balances of the methanation reactions. These reactions generally form water, and consequently this species needs to be separated. With regard to the other species (CO₂ and H₂), their respective concentrations can be modified by acting first on the operating mode of the reactor (adiabatic or isothermal) and second on the temperature or pressure. A high pressure and a low
15 temperature will therefore allow the concentrations of these compounds to be reduced considerably. When the operation is carried out in an “adiabatic” reactor, a series of steps is also necessary to achieve an equivalent conversion quality to the isothermal reactor. In any event, for an operating pressure less than 40 bars, the composition of the gas produced is generally incompatible with regard to the specifications for the injection or use of natural gas,
20 and improvement steps are necessary to remove the CO₂ and/or the residual H₂. Therefore, the operating mode forms a block to simplifying the chain of methods.

Systems are known such as those described in document US 2013/0317126. In these systems, an adiabatic methanation reactor is utilized and a portion of the methanation products is recirculated in the input to said reactor.

- 25 This recirculation of methanation products is aimed, according to this document, at adjusting the temperature of the reagents input into the adiabatic methanation reactor so as to moderate the exothermicity of the adiabatic reaction occurring in the reactor.

In these solutions, the temperature reached on input to the methanation reactor is of the order of 310 to 330°C and the temperature on output from this reactor is of the order of
30 620°C, which results in a somewhat inefficient conversion limited by the thermodynamics of the reaction and therefore the presence of undesirable compounds, for example excess H₂, CO₂ in the flow output from the reactor. This presence of undesirable compounds, especially hydrogen, makes a hydrogen separation step necessary downstream from the reactor to meet, for example, the specifications for injection into the natural gas distribution or transport
35 grid.

The flow is said to meet injection specifications when the following characteristics of the flow:

- high heating value ("HHV");
- Wobbe index; and
- hydrogen content

are within predefined value ranges corresponding to the particular characteristics of the natural gas distribution or transport grid.

Systems such as those described in document US 3 967 936 are also known. In these systems, a series of adiabatic methanation reactors is utilized and a portion of the methanation products is recirculated in the input to each reactor of the series.

This recirculation of methanation products is aimed, according to this document, at adjusting the temperature of the reagents input into the adiabatic methanation reactor so as to moderate the exothermicity of the adiabatic reaction occurring in the reactor.

In the same way, these solutions require the separation, downstream from the reactor, of undesirable compounds such as hydrogen to meet, for example, the specifications for injection into the natural gas distribution or transport grid.

Systems are known such as those described in document US 2009/0247653. In these systems, a series of three adiabatic methanation reactors is utilized, the last reactor in the series being designed to produce additional synthetic methane. In these systems, a portion of the methanation products CO and H₂ is recirculated after the second reactor towards the input flow of the first reactor in the series so as to adjust the CO and H₂ ratio and adjust the temperature of the input flow of the first reactor in order to moderate the exothermicity of the adiabatic reaction occurring in the first reactor.

However, these solutions also require the separation, downstream from the reactor, of undesirable compounds such as hydrogen.

The adiabatic reactors, while they do not assume cooling within the methanation reactor, have several drawbacks:

- a plurality of reactors in series is necessary to obtain a satisfactory methanation yield; and
- in the case of CO methanation, a specific composition of the synthetic gas, by adding a so-called gas-to-water catalyst step, is necessary to obtain a satisfactory stoichiometry.

On the other hand, the design of adiabatic reactors is simpler, since they basically consist of a chamber having to resist generally high pressures (>30 bars) to achieve a satisfactory conversion.

In the case of methanation in an isothermal reactor, the operating pressure does not need to be as high (<20 bars), but requires the arrangement of surfaces immersed in the catalyst layer, which generates a complex design and additional cost linked to the cooling system.

Consequently, current systems do not make it possible to control the reaction temperature in an isothermal methanation reactor.

In conclusion, the current techniques of the state of the art do not enable satisfactory control of the exothermicity of the isothermal methanation reaction and do not give the device satisfactory flexibility with regard to power fluctuations linked directly to the overproduction of electricity. In addition, these current techniques require a systematic separation of H₂ and/or CO₂ to meet the specifications for the injection or use as a substitute for natural gas in the grid.

Therefore, the current systems do not make the adjustment to specifications for injection into the natural gas distribution or transport grid possible without a hydrogen separation step, downstream from the methanation step, taking place.

SUBJECT OF THE INVENTION

The present invention aims to remedy all or part of these drawbacks.

To this end, according to a first aspect, the present invention envisages a synthetic gas production device, which comprises:

- a water electrolysis means for producing oxygen and hydrogen;
- a carbon dioxide supply channel;
- a means for injecting hydrogen into a supply channel supplied with carbon dioxide, this carbon dioxide and this hydrogen forming a syngas;
- an isothermal methanation reactor comprising:
 - an inlet intended for the syngas and connected to the syngas supply channel, and
 - an outlet for synthetic natural gas;
- a water separation means comprising:
 - an inlet for synthetic natural gas and
 - an outlet for dehydrated synthetic natural gas; and
- a bypass for a portion of the dehydrated synthetic natural gas from the outlet of the water separation means to the syngas supply channel in order to provide a mixture of the syngas and the bypassed synthetic natural gas to the reactor.

As the water separation means cools the synthetic natural gas, supplying a portion of this synthetic natural gas on input to the reactor allows the syngas to be cooled and means the reactor does not require a heat exchanger. The design of such a reactor, especially in terms of sizing, is made even simpler. In addition, the supply of dehydrated synthetic natural gas in the supply channel improves the Wobbe index and HHV of the methanation reaction products through the favorable modification of the reaction balances. Therefore, the device that is the subject of the present invention allows a reduced sizing of the reactor and an

adjustment to specifications for the gas grid requiring little or no processing downstream from the methanation reaction.

In addition, the utilization of an isothermal reactor makes it possible to have a single methanation step to obtain an efficient synthetic natural gas conversion and obtain a gas of a quality close to the specifications for injection into the natural gas distribution or transport grid.

In some embodiments, the device that is the subject of the present invention comprises a bypass channel, for a portion of the hot methanation reaction products, comprising:

- an inlet positioned between the outlet from the reactor and the water separation means; and
- an outlet positioned upstream from the inlet to the reactor.

These embodiments make it possible to regulate the flow rate input to the reactor so as to give the device great flexibility, regardless of the quantity of electrical energy produced upstream.

In some embodiments, the device that is the subject of the present invention comprises:

- a means for measuring the flow rate of hydrogen produced by electrolysis; and
- a means for regulating the flow rate of carbon dioxide in the carbon supply channel as a function of the hydrogen flow rate measured.

These embodiments make it possible to regulate the composition of syngas input to the methanation reactor.

In some embodiments, the device that is the subject of the present invention comprises:

- downstream from the reactor, a sensor of a temperature of the synthetic natural gas; and
- upstream from the input to the reactor, a means for preheating the mixture as a function of the temperature measured.

These embodiments make it possible to control the temperature of the reactor and ensure the isothermality of this reactor.

In some embodiments, the device that is the subject of the present invention comprises:

- a sensor of a physical magnitude representative of a fuel heating value or a composition of the dehydrated synthetic natural gas; and
- a recirculator of natural gas input into the bypass, controlled as a function of a value of the physical magnitude measured.

In some embodiments, the device that is the subject of the present invention comprises:

- a means for measuring the flow rate of the syngas downstream from the mixing location; and
- a recirculator of the hot natural gas input into the bypass channel, controlled as a function of the syngas flow rate measured.

These embodiments make it possible to regulate the flow rate input to the reactor as a function of the syngas flow rate measured.

In some embodiments, the water separation means is configured to cool the synthetic natural gases to a temperature between -5°C and +60°C.

These embodiments make it possible to separate almost all of the water contained in the SNG on output from the methanation reactor.

In some embodiments, the separation means is configured to cool the synthetic natural gases to a temperature below the dew point temperature of the water at the operating conditions of the reactor in question.

In some embodiments, the isothermal reactor is a fluidized-bed reactor.

In some embodiments, the device that is the subject of the present invention comprises at least one heat exchange surface positioned in the fluidized bed.

These embodiments make it possible to regulate the temperature inside the methanation reactor.

These embodiments allow the temperature in the catalyst layer of the isothermal reactor to be made uniform simply.

According to a second aspect, the present invention envisages a method for producing synthetic gas, which comprises:

- a water electrolysis step to produce oxygen and hydrogen;
- a step of supplying carbon dioxide;
- a step of injecting hydrogen, output from the electrolysis step, into a supply channel supplied with carbon dioxide, output from the carbon dioxide supply step, this carbon dioxide and this hydrogen forming a syngas;
- a methanation reaction step, comprising:
 - a step of inputting syngas into an isothermal methanation reactor by means of the syngas supply channel; and
 - a step of outputting synthetic natural gas;
- a step of separating water, comprising:
 - a step of inputting synthetic natural gas and
 - a step of outputting dehydrated synthetic natural gas; and

- a step of bypassing a portion of the dehydrated synthetic natural gas output from the water separation step to the syngas supply channel in order to provide a mixture of the syngas and the bypassed synthetic natural gas to the reactor.

The method that is the subject of the present invention corresponding to the device that is the subject of the present invention, the particular features, advantages and aims of this method are similar to those of the device that is the subject of the present invention. These features, advantages and aims are not repeated here.

In some embodiments, the method that is the subject of the present invention comprises a step of bypassing a portion of the hot methanation reaction products, to upstream of the methanation step.

BRIEF DESCRIPTION OF THE FIGURES

Other advantages, aims and particular features of the invention will become apparent from the non-limiting description that follows of at least one particular embodiment of the device and method for producing synthetic gas that are the subjects of the present invention, with reference to drawings included in an appendix, wherein:

- figure 1 represents, schematically, a particular embodiment of the device that is the subject of the present invention;
- figure 2 represents, schematically and in the form of a logical diagram, a particular series of steps of the method that is the subject of the present invention;
- figure 3 represents, in the form of a curve, the Wobbe index of synthetic gas obtained by the embodiment of the device figure 1 and the method that are the subjects of the present invention;
- figure 4 represents, in the form of a curve, the HHV of synthetic gas obtained by the embodiment of the device figure 1 and method that are the subjects of the present invention;
- figure 5 represents, in the form of a curve, the reduction of the hydrogen molar flow in the natural gases on output from the methanation reactor during the utilization of the embodiment of the device figure 1 and method that are the subjects of the present invention;
- figure 6 represents, in the form of a curve, the exothermicity of the methanation reaction during the utilization of the embodiment of the device figure 1 and method that are the subjects of the present invention; and
- figure 7 represents, schematically, an example of system utilized in the state of the art.

DESCRIPTION OF EXAMPLES OF REALIZATION OF THE INVENTION

The present description is given as a non-limiting example, each characteristic of an embodiment being able to be combined with any other characteristic of any other embodiment in an advantageous way.

It is now noted that the figures are not to scale.

5 Figure 7 shows a schematic view of an example of system 80 utilized in the state of the art.

In these systems 1600, methanation reagents enter into a methanation reactor 1605, which can be part of a series (not shown) of such reactors.

10 On output from the methanation step, the water is separated from the methanation products by a means 1625 for separating this water, such as a heat exchanger for example.

The dehydrated synthetic natural gas is then processed by a means 1645 for separating carbon dioxide.

15 Lastly, the synthetic natural gas is processed by a means 1655 for separating hydrogen so that this synthetic natural gas meets the specifications for injection into the natural gas distribution grid.

The steps of separating water, carbon dioxide and hydrogen can be carried out in any order.

20 Figure 1, which is not to scale, shows a schematic view of an embodiment of the device 1000 that is the subject of the present invention. This synthetic gas production device 1000 comprises:

- a water electrolysis means 1105 for producing oxygen and hydrogen;
- a carbon dioxide supply channel 1110;
- a means 1115 for injecting hydrogen into a supply channel 1015 supplied with carbon dioxide, this carbon dioxide and this hydrogen forming a syngas;
- 25 - an isothermal methanation reactor 1005 comprising:
 - an inlet 1010 intended for the syngas and connected to the syngas supply channel 1015, and
 - an outlet 1020 for synthetic natural gas;
- a water separation means 1025 comprising:
 - 30 - an inlet 1030 for synthetic natural gas,
 - an outlet 1035 for dehydrated synthetic natural gas; and
 - an outlet 1027 for the water separated from the synthetic natural gas; and
- a bypass 1040 for a portion of the dehydrated synthetic natural gas from the outlet of the water separation means to the syngas supply channel in order to provide a mixture of the bypassed syngas and synthetic natural gas to the reactor.
- 35

The reactor 1005 is, preferably, an isothermal fluidized-bed methanation reactor operating at a predefined temperature. Fluidization of the catalyst by the reactant mixture

enables almost perfect homogenization of the temperatures at all points of the catalyst layer and the reactor 1005 can be assimilated to an isothermal reactor. In some variants, this reactor 1005 can be a boiling water reactor, known to the person skilled in the art under the abbreviation "BWR" (for Boiling Water Reactor). In other variants, this reactor 1005 can be a
5 wall-cooled reactor or an exchanger reactor.

In some embodiments, the reactor 1005 comprises at least one heat exchange surface 1006 positioned in the fluidized bed of the isothermal reactor 1005.

This surface 1006 is, for example, a tube configured to form a loop for circulating a fluid from the outside the reactor 1005 to the inside of this reactor 1005, the fluid being
10 cooled outside the reactor 1005.

This fluid is, for example, superheated or saturated water vapor.

This reactor 1005 is configured to carry out the methanation of the carbon dioxide.

This reactor 1005 comprises the inlet 1010 for syngas which is, for example, an aperture of the reactor 1005 equipped with a connector (not shown) compatible with the
15 syngas supply channel 1015.

The synthetic natural gases leave the reactor 1005 through the outlet 1020 from the reactor. This outlet 1020 is, for example, an aperture connected to a connector (not shown) making it possible to connect a sealed channel for transporting synthetic natural gas.

The water separation means 1025 is, for example, a heat exchanger for cooling the
20 synthetic natural gases to a temperature below the dew point temperature of the water. This temperature is preferably between -5°C and $+60^{\circ}\text{C}$. Preferentially, this temperature is between 5°C and 40°C .

In some preferred embodiments, the water separation means 1025 is configured to cool the synthetic natural gases to a temperature below the dew point temperature of the
25 water at the operating conditions of the reactor 1005 in question.

The water separated in this way is collected by an outlet 1027 for water and can be used by the water electrolysis means 1105.

The water separation means 1025 comprises the inlet 1030 for synthetic natural gas. This inlet 1030 is, for example, an aperture associated with a connector (not shown) to be
30 connected to a sealed channel for transporting synthetic natural gas output from the reactor 1005.

The water separation means 1025 comprises the outlet 1035 for dehydrated synthetic natural gas. This outlet 1035 is, for example, an aperture associated with a connector (not shown) to be connected to a sealed channel (not referenced) transporting dehydrated
35 synthetic natural gas.

The bypass 1040 is, for example, a sealed channel connected to the transport channel for dehydrated synthetic natural gas in order to capture a portion of the flow passing through this transport channel.

This bypass 1040 injects the dehydrated synthetic natural gases into the syngas supply channel 1015.

In this way, the syngas and the dehydrated synthetic natural gas, cooled by the water separation process, form a mixture which, in the reactor 1005, reduces the exothermicity of the methanation reaction and in this way improves the specifications of the synthetic natural gas output from the reactor 1005.

In particular, the mixture produced makes it possible to avoid the downstream separation of H_2 in the SNG.

The supply channel 1015 is sealed and receives, for example, CO_2 and H_2 output from a water electrolysis means.

The electrolysis means 1105 is, for example, an electrolyzer for water, configured to separate water molecules into oxygen molecules and hydrogen molecules.

The carbon dioxide in the supply channel 1110 is processed by compression and by treating impurities upstream from the device 1000.

The injection means 1115 is, for examples, a valve for injecting hydrogen into the supply channel 1015.

In some preferred embodiments, such as that shown in figure 1, the device 1000 comprises:

- a means 1120 for measuring the flow rate of hydrogen produced by electrolysis; and
- a means 1125 for processing carbon dioxide in the carbon supply channel 1015 as a function of the hydrogen flow rate measured.

The carbon dioxide processing means 1125 is, for example, an electronic control circuit that multiplies the measured hydrogen flow rate by a determined setpoint coefficient 1126. The result of this multiplication is communicated to a carbon dioxide flow rate regulator valve, this result corresponding to a flow rate setpoint.

For an optimum production of CH_4 , the H_2/CO_2 ratio must be adjusted to close to stoichiometry, ie between 3.9 and 4.1, so as to limit the excess reagents. This ratio is controlled by regulating the CO_2 flow rate as a function of the flow rate measurement of the H_2 flow and a stoichiometric factor.

In some preferred embodiments, such as that shown in figure 1, the device 1000 comprises:

- downstream from the reactor 1005 and upstream from the water separation means 1025, a sensor 1062 of a temperature of the synthetic natural gas; and

- upstream from the input to the reactor, a means 1060 for preheating the mixture as a function of the temperature measured.

The preheating means 1060 preferably heats the mixture to a temperature higher than the dew point temperature of the mixture, to prevent any pre-condensation of the mixture, and lower than the operating temperature of the reactor, to allow the reactor 1005 to be cooled.

In some preferred embodiments, such as that shown in figure 1, the mixture preheating means 1060 heats this mixture to a temperature between 50°C and 300°C. Preferably, the mixture preheating means 1060 heats this mixture to a temperature higher than 50°C and less than the operating temperature of the reactor 1005.

This preheating means 1060 is, for example, a heat exchanger for heating the syngas.

The temperature of the preheating means 1060 varies as a function of the captured temperature and a predefined temperature setpoint. If the captured temperature is higher than the temperature setpoint, the power of the preheating means 1060 is reduced. Conversely, the power of the preheating means 1060 is increased when the captured temperature is below the temperature setpoint.

In some preferred embodiments, such as that shown in figure 1, the device 1000 comprises a bypass channel 1070, for a portion of the hot methanation reaction products, comprising:

- an inlet 1075 positioned between the outlet from the reactor 1005 and the water separation means 1025; and
- an outlet 1080 positioned upstream from the inlet 1010 to the reactor 1005.

The bypass channel 1070 is, for example, a sealed channel. The inlet 1075 is, for example, an aperture emerging at the interior of the synthetic natural gas transport channel, upstream from the water separation means 1025. The outlet 1080 is, for example, an aperture for injecting synthetic natural gas into the mixture, downstream from the preheating means 1060.

The synthetic natural gases, being hot, make it possible to maintain the flow rate in the reactor 1005.

The flow rate of the syngas is entirely a function of the quantities of electricity available. In order to maintain conversion stability during the methanation operation, the hydrodynamic conditions must be kept as constant as possible. However, if the available electrical power is insufficient and, as a result, the syngas flow rate is reduced, it is necessary to maintain a constant overall flow rate input to the reactor 1005 or opt for a very flexible technology. Even in the case of the fluidized bed able to operate in a flow rate range of one to six, flow rates that are too low can cause degradation of the cooling, and therefore

of the conversion. To overcome this difficulty, in the event of a significant drop in the syngas flow rate, the flow rate on output from the preheating means 1060 is supplemented by a hot recirculation coming directly from the outlet 1020 of the reactor 1005 by means of the bypass channel 1070. The fact of using a hot recirculation fluid does not cause a thermal imbalance of the reactor 1005 but allows the device 1000 to be made very flexible.

In some preferred embodiments, such as that shown in figure 1, the device 1000 comprises:

- a means 1085 for measuring the flow rate of the syngas downstream from the mixing location and upstream from the reactor 1005; and
- a recirculator 1090 of the natural gas input into the bypass channel 1070, controlled as a function of the flow rate measured.

The flow rate measurement means 1085 can be any type known to the person skilled in the art that is suitable for measuring the flow rate of gases, such as an anemometer, Coriolis effect flowmeter, vortex effect flowmeter, or electromagnetic flowmeter, for example.

The recirculator 1090 is similar to the recirculator 1055 in structural terms. This recirculator 1090 is controlled as a function of the flow rate measured by the flow rate measurement means 1085 and a flow rate setpoint value 1087. If the measured flow rate is below a predefined flow rate setpoint 1087, the recirculator 1090 is actuated so as to make up the difference between the measured flow rate and the flow rate setpoint 1087 by an equivalent flow rate of synthetic natural gas.

In some preferred embodiments, such as that shown in figure 1, the device 1000 comprises:

- a sensor 1050 of a physical magnitude representative of a fuel heating value or a composition of the dehydrated synthetic natural gas; and
- a recirculator 1055 of hot natural gas input into the bypass, controlled as a function of a value of the physical magnitude measured.

The sensor 1050 is, for example, an electronic circuit for estimating the HHV associated with a gas chromatograph determining a composition of the SNG in the bypass 1040 or in the outlet 1035.

The recirculator 1055 aims to offset the load, or pressure, losses successively in the preheating means 1060, the reactor 1005, the water separation means 1025 particularly for the device 1000 described with reference to figure 1, and all the connecting channels of these various items of equipment.

Such a load loss is estimated to be between 200 and 800 mbars, for example. The recirculator 1055 is, for example, a fan, compressor or ejector.

In the embodiment of figure 1, the device 1000 comprises a means 1085 for measuring the flow rate of the syngas downstream from the mixing location, the recirculator

1090 of the natural gas input into the bypass channel 1070 being controlled as a function of the syngas flow rate measured.

Thanks to the characteristics described with reference to figure 1, at an operating pressure close to ten bars, and for a given flow rate, this embodiment allows the successive
 5 H₂ and CO₂ separation steps to be eliminated.

The utilization of the device 1000 that is the subject of the present invention makes it possible to obtain synthetic gas close to the specifications of the gas grid requiring few additional operations.

In addition, the device 1000 can also be utilized for a pressure range of between one
 10 bar and one hundred bars, and a range of predefined temperatures of between 150°C and 700°C.

Figure 2 shows, in the form of a logical diagram of steps, a particular embodiment of the method 1600 that is the subject of the present invention. This synthetic gas production method 1600 comprises:

- 15 - a water electrolysis step 1640 to produce oxygen and hydrogen;
- a step 1645 of supplying carbon dioxide;
- a step 1650 of injecting hydrogen, output from the electrolysis step 1640, into a supply channel supplied with carbon dioxide, output from the carbon dioxide supply step 1645, this carbon dioxide and this hydrogen forming a syngas;
- 20 - a methanation reaction step 1605, comprising:
 - a step 1610 of inputting syngas into an isothermal methanation reactor by means of the syngas supply channel; and
 - a step 1615 of outputting synthetic natural gas;
- a step 1620 of separating water, comprising:
 - 25 - a step 1625 of inputting synthetic natural gas and
 - a step 1630 of outputting dehydrated synthetic natural gas;
- a step 1635 of bypassing a portion of the dehydrated synthetic natural gas output from the water separation step to the syngas supply channel in order to provide a mixture of the syngas and the bypassed synthetic natural gas to the reactor; and
- 30 - preferably, a step 1655 of bypassing a portion of the hot methanation reaction products, output from the reaction step 1605, to upstream of the methanation step.

This method 1600 is utilized, for example, by a device 1000 that is the subject of the present invention and described with reference to figure 1.

Figures 3 to 6 show the result of simulations carried out for the device 1000 that is the
 35 subject of the present invention.

Figure 3 shows, on the x-axis, the Wobbe index of the synthetic natural gas produced by the device 1000, as a function of the recirculation rate, on the y-axis, and of the nature of

the recirculated synthetic natural gas on input to the methanation reactor 1005. "Recirculation rate" means the quantity of gas output from the separation means 1025 reinjected into the channel 1015 divided by the total quantity of gas output from the separation means 1025.

It is noted, in particular, that the recirculation of humid synthetic natural gas 1205 has no effect on the Wobbe index of the synthetic natural gas produced by the device.

It is also noted that the recirculation of dehydrated synthetic natural gas 1210 improves the Wobbe index of the synthetic natural gas produced by the device.

Figure 4 shows, on the x-axis, the HHV of the synthetic natural gas produced by the device 1000, as a function of the recirculation rate, on the y-axis, and of the nature of the recirculated synthetic natural gas on input to the methanation reactor 1005.

It is noted, in particular, that the recirculation of humid synthetic natural gas 1305 has no effect on the HHV of the synthetic natural gas produced by the device.

It is also noted that the recirculation of dehydrated synthetic natural gas 1310 improves the HHV of the synthetic natural gas produced by the device.

Figure 5 shows, on the x-axis, the relative variation in the H_2 molar flow on output from device 1000, as a function of the recirculation rate, on the y-axis, and of the nature of the recirculated synthetic natural gas on input to the methanation reactor 1005 compared to the H_2 molar flow in a device without recirculation.

It is noted, in particular, that the recirculation of humid synthetic natural gas 1405 has no effect on the H_2 molar flow on output from the device.

It is also noted that the recirculation of dehydrated synthetic natural gas 1410 causes a reduction in the H_2 molar flow on output from the device.

Figure 6 makes it possible to see the change in the reactor's normalized exothermicity, ie the heat to be removed compared to a case without recirculation, as a function of the recirculation rate for the reference configuration and the device 1000 described above.

Figure 6 shows, on the x-axis, the exothermicity of the methanation reaction of the device 1000 as a function of the recirculation rate, on the y-axis, and of the nature of the recirculated synthetic natural gas on input to the methanation reactor 1005.

It is noted, in particular, that the recirculation of humid synthetic natural gas 1505 reduces the exothermicity of the methanation reaction.

It is also noted that the recirculation of dehydrated synthetic natural gas 1510 also reduces the exothermicity of the methanation reactor.

It appears that increasing the recirculation rate leads to a linear reduction in the exothermicity of the reactor. Here, the recirculated SNG plays the role of heat accumulator, which is more marked in the presence of H_2O because of a higher heat capacity. Obtaining a

recirculation level corresponding to an ideal operating temperature of the device 1000 makes it possible to be liberated from the internal exchanger of the reactor and the H_2 separation for adjustment to specifications. Further, the reactor is allothermal and therefore requires a heat supply to maintain the reactions.

- 5 The change in the molar fractions of the H_2 , CO_2 and CH_4 species as a function of the recirculation rate for the configuration simulated is in the direction of improving the SNG quality with the dehydrated SNG recirculation device 1000.

CLAIMS

1. Synthetic gas production device (1000) characterized in that it comprises:

- a water electrolysis means (1105) for producing oxygen and hydrogen;
- a carbon dioxide supply channel (1110);
- a means (1115) for injecting hydrogen into a supply channel (1015) supplied with carbon dioxide, this carbon dioxide and this hydrogen forming a syngas;
- an isothermal methanation reactor (1005) comprising:
 - an inlet (1010) intended for the syngas and connected to the syngas supply channel (1015) and
 - an outlet (1020) for synthetic natural gas;
- a water separation means (1025) comprising:
 - an inlet (1030) for synthetic natural gas and
 - an outlet (1035) for dehydrated synthetic natural gas; and
- a bypass (1040) for a portion of the dehydrated synthetic natural gas from the outlet of the water separation means to the syngas supply channel in order to provide a mixture of the bypassed syngas and synthetic natural gas to the reactor.

2. Device (1000) according to claim 1, which comprises:

- a means (1120) for measuring the flow rate of hydrogen produced by electrolysis; and
- a means (1125) for processing carbon dioxide in the carbon supply channel (1015) as a function of the hydrogen flow rate measured.

3. Device (1000) according to one of claims 1 to 2, which comprises:

- downstream from the reactor (1005) and upstream from the water separation means (1025), a sensor (1062) of a temperature of the synthetic natural gas; and
- upstream from the input to the reactor, a means (1060) for preheating the mixture as a function of the temperature measured.

4. Device (1000) according to one of claims 1 to 3, which comprises:

- a sensor (1050) of a physical magnitude representative of a fuel heating value or a composition of the dehydrated synthetic natural gas; and
- a recirculator (1055) of natural gas input into the bypass, controlled as a function of a value of the physical magnitude measured.

5. Device (1000) according to one of claims 1 to 4, which comprises a bypass channel (1070), for a portion of the hot methanation reaction products, comprising:

- an inlet (1075) positioned between the outlet from the reactor (1005) and the water separation means (1025); and

5 - an outlet (1080) positioned upstream from the inlet to the reactor.

6. Device (1000) according to one of claims 1 to 5, which comprises:

- a means (1085) for measuring the flow rate of the syngas downstream from the mixing location; and

10 - a recirculator (1090) of the hot natural gas input into the bypass channel (1070), controlled as a function of the syngas flow rate measured.

7. Device (1000) according to one of claims 1 to 6, wherein the separation means (1025) is configured to cool the synthetic natural gases to a temperature between -5°C and +60°C.

15

8. Device (1000) according to claim 7, wherein the water separation means (1025) is configured to cool the synthetic natural gases to a temperature below the dew point temperature of the water at the operating conditions of the reactor (1005) in question.

20 9. Device (1000) according to claims 7 and 8, wherein the water separation means (1025) is configured to cool the synthetic natural gases to a temperature below the dew point temperature at the operating conditions of the reactor (1005).

10. Device (1000) according to one of claims 1 to 9, wherein the isothermal reactor (1005) is a fluidized-bed reactor.

25

11. Device (1000) according to claim 10, which comprises at least one heat exchange surface (1006) positioned in the fluidized bed.

30 12. Method (1600) for producing synthetic gas, characterized in that it comprises:

- a water electrolysis step (1640) to produce oxygen and hydrogen;
- a step (1645) of supplying carbon dioxide;
- a step (1650) of injecting hydrogen, output from the electrolysis step, into a supply channel supplied with carbon dioxide, output from the carbon dioxide supply step, this carbon dioxide and this hydrogen forming a syngas;
- a methanation reaction step (1605), comprising:

35

- a step (1610) of inputting syngas into an isothermal methanation reactor by means of the syngas supply channel; and
- a step (1615) of outputting synthetic natural gas;
- a step (1620) of separating water, comprising:
 - a step (1625) of inputting synthetic natural gas and
 - a step (1630) of outputting dehydrated synthetic natural gas;
- a step (1635) of bypassing a portion of the dehydrated synthetic natural gas output from the water separation step to the syngas supply channel in order to provide a mixture of the syngas and the bypassed synthetic natural gas to the reactor.

10

13. Method (1600) according to claim 12, which comprises a step (1655) of bypassing a portion of the hot methanation reaction products, to upstream of the methanation step.