DESCRIPTION

METHOD AND SYSTEM FOR PRODUCING HYDROGEN AND/OR AMMONIA

FIELD

[0001] The present disclosure relates to a method and to a system for producing hydrogen and/or ammonia.

BACKGROUND

[0002] More than 90% of the world's ammonia is currently produced using the Haber-Bosch process. In this process, hydrogen and nitrogen are converted to ammonia at high temperatures and pressures in the presence of an iron catalyst. This process is extremely energy-intensive, with a typical energy consumption of 28 to 49 GJ per ton of ammonia.

[0003] To provide hydrogen, various known hydrocarbon-containing feedstocks can be converted using suitable processes such as steam reforming, partial oxidation, autothermal reforming or a combination of these processes. In all these processes, carbon dioxide is produced in at least one flue gas, which in conventional processes is at least partially released into the atmosphere.

[0004] Recently, the production of so-called blue hydrogen has become increasingly important. The aim is to avoid the release of carbon dioxide into the atmosphere as much as possible, through appropriate process steps. Typically, the resulting carbon dioxide is separated from a flue gas or a mixture of precursor products of the aforementioned processes by means of chemical and/or physical scrubbing. After its separation, the carbon dioxide can be

compressed, purified and/or liquefied before, for example, being placed in a final repository. This is also referred to as sequestration.

[0005] There is a need for methods for producing hydrogen and/or ammonia that at least partially overcome the disadvantages of known methods.

OVERVIEW

[0006] Against this background, methods and systems for producing hydrogen and/or ammonia, having the features of the independent claims, are proposed. Each of the embodiments is the subject matter of the dependent claims and of the description below.

[0007] The present disclosure relates to the production of ammonia, but also to the production of hydrogen alone without further conversion or with conversion to compounds other than ammonia.

[0008] The proposed method for producing hydrogen and/or ammonia comprises, but is not limited to, the following steps: Providing a first synthesis gas stream containing hydrogen, carbon monoxide, and carbon dioxide; providing a second synthesis gas stream which is enriched in hydrogen and carbon dioxide and depleted of carbon monoxide compared to the first synthesis gas stream, using the first synthesis gas stream or a part thereof; providing a first hydrogen stream using hydrogen from the second synthesis gas stream and by means of a first pressure swing adsorption unit; providing a carbon dioxide stream using carbon dioxide from the second synthesis gas stream and by means of a carbon dioxide separation unit; and processing a first residual gas stream or a part thereof remaining downstream of where the first hydrogen stream and the carbon dioxide stream are provided, in order to obtain a second hydrogen stream and a second residual gas stream.

[0009] To further refine and improve such a method, as disclosed for example in patent application EP 3 954 650 A1, it is proposed to return the second residual gas stream to a position directly upstream of where the carbon dioxide stream is provided.

[0010] In this way, carbon dioxide contained in the second residual gas stream is converted into a pure carbon dioxide stream, which is then disposed of, for example, by sequestration.

[0011] The proposed method makes it possible to increase the hydrogen yield and the degree of carbon dioxide separation compared to the prior art without further separation steps.

[0012] Advantageously, the first residual gas stream is processed by means of a second pressure swing adsorption unit to obtain the second residual gas stream.

[0013] Preferably, the first synthesis gas stream is provided from a feed gas stream containing one or more hydrocarbons, using autothermal reforming or partial oxidation. The feed gas stream can be fed directly to autothermal reforming or partial oxidation without prior reforming. However, it should not be ruled out to convert the feed gas stream in an adiabatic reforming unit into a partially reformed stream, which is then further processed in autothermal reforming or partial oxidation.

[0014] A preferred embodiment of the method according to the invention provides that the second hydrogen stream or a part thereof is thermally and/or materially recovered in the process itself or outside of it. In particular, if the second hydrogen stream is obtained by means of pressure swing adsorption, its thermal recovery is uncritical due to its largely carbon dioxide-free composition. Its material recovery can be achieved in particular by feeding it

back upstream of the first pressure swing adsorption unit, so that the contained hydrogen is largely transferred into the first hydrogen stream.

[0015] It is expedient to arrange a drying unit upstream of the carbon dioxide separation unit to prevent water from being introduced into the carbon dioxide processing, which is operated particularly cryogenically, where it would lead to blockages.

[0016] In one proposed embodiment, the method according to the invention comprises providing the second synthesis gas stream and converting carbon monoxide to carbon dioxide and hydrogen using a water-gas shift reaction, in particular using an isothermal, high-temperature, medium-temperature and/or low-temperature shift reaction. In this way, the hydrogen yield can be increased and a carbon dioxide concentration suitable for separation can be achieved.

[0017] In one proposed embodiment, the method comprises providing the first hydrogen stream downstream of where the carbon dioxide stream is provided.

[0018] The proposed system for producing hydrogen and/or ammonia comprises a device for providing a first synthesis gas stream containing hydrogen, carbon monoxide, and carbon dioxide; a conversion device for providing a second synthesis gas stream which is enriched in hydrogen and carbon dioxide and depleted of carbon monoxide compared to the first synthesis gas stream, using the first synthesis gas stream or a part thereof; a first pressure swing adsorption unit for providing a first hydrogen stream using hydrogen from the second synthesis gas stream; a carbon dioxide separation unit for providing a carbon dioxide stream using carbon dioxide from the second synthesis gas stream; and a separation device for separating a first residual gas stream or a part thereof remaining downstream of where the first hydrogen stream and the carbon dioxide stream are provided to obtain a second hydrogen stream and a second residual gas stream.

[0019] According to the invention, the device comprises a feedback device via which the second residual gas stream can be fed back to a position directly upstream of where the carbon dioxide stream is provided.

[0020] For further features and advantages of a corresponding system and embodiments thereof, reference is expressly made to the above explanations relating to the method proposed according to the invention and its embodiments, since they apply in the same way here.

DRAWINGS

[0021] Fig. 1 schematically shows a method disclosed in patent application EP 3 954 650 A1, while Fig. 2 also schematically illustrates an embodiment of this method according to the invention. In the two figures, identical system components and streams are identified by the same reference signs.

[0022] In the method of Fig. 1, a hydrocarbon-containing feed gas stream FG is fed to an endothermic reforming unit 200 and there is converted to a partially reformed stream SG1, which is further reformed in an autothermal reforming unit 201 to the first synthesis gas stream SG3. The heat generated in the autothermal reforming unit 201 is used to heat the endothermic reforming unit 200, as illustrated by the heat flow 202. In particular, the first synthesis gas stream SG3 can be used directly for heating in the endothermic reforming unit 200, i.e. the heat contained in the first synthesis gas stream SG3 can be used for heating in the endothermic reforming unit 200 without further transfer to a thermal exchange medium.

[0023] The first synthesis gas stream SG3 is fed to a conversion unit 203, in which carbon monoxide contained in the first synthesis gas stream SG3 is reacted with water vapor to form carbon dioxide and hydrogen. A second synthesis gas stream SG4 obtained in this process and enriched with hydrogen compared to the first SG3, is subsequently fed to a first separation unit 204

designed as a pressure swing adsorption unit in order to separate hydrogen HG1 from the second synthesis gas stream SG4 with high purity. Likewise, a first residual gas stream RG1, which is depleted of hydrogen compared to the second synthesis gas stream SG4, is withdrawn from the first separation unit 204 and fed to a separation unit 205. In the separation unit 205, carbon dioxide is separated in high purity through multiple compression and cooling steps and withdrawn as a carbon dioxide stream CG1 from the separation unit 205. In the terminology used here, the first hydrogen stream HG1 is provided using hydrogen from the second synthesis gas stream SG4, and a carbon dioxide stream CG1 is provided using carbon dioxide from the second synthesis gas stream SG4. The carbon dioxide stream CG1 still contains significant residual amounts of methane, which can optionally be removed by distillation of the carbon dioxide stream CG1 (not shown). A resulting pure carbon dioxide product is suitable for sequestration of the carbon dioxide or for further use, for example for the synthesis of methanol by reaction with hydrogen produced from electrolysis.

[0024] From the separation unit 205, a second residual gas stream RG2, which is highly depleted of carbon dioxide, can be extracted, from which a second hydrogen stream HG2, which is enriched with hydrogen compared to the second residual gas stream RG2, and a third residual gas stream RG3, which is depleted of hydrogen compared to the second residual gas stream RG2, can be generated in the second separation unit 206, which is designed as a membrane separation unit. In Fig. 1, the second hydrogen stream HG2 is supplied to the autothermal reforming unit 201 as fuel gas. Alternatively, it can also be fed to the first separation unit 204.

[0025] In the method designated as 10 in Fig. 2, a hydrocarbon-containing feed gas stream FG is fed to the adiabatic reforming unit 100 to produce the partially reformed stream SG1. No heat flow is conveyed from the autothermal reforming unit 201 to the adiabatic reforming unit 100. Also unlike the method of Fig. 1, the second separation unit 206' is designed as a pressure swing

adsorption unit, in which a third residual gas stream RG3 and a second hydrogen stream HG2 are obtained from the second residual gas stream RG2. While the second hydrogen stream HG2 is supplied to the autothermal reforming unit 201 as fuel gas or used in the first separation unit 204, the third residual gas stream RG3 is returned immediately upstream of the separation unit 205.

This allows the carbon dioxide separation rates typically required for producing blue hydrogen to be achieved with minimal equipment.

Claims

1. Method (10) for producing hydrogen and/or ammonia, comprising the following steps:

providing a first synthesis gas stream (SG3) containing hydrogen, carbon monoxide and carbon dioxide;

providing a second synthesis gas stream (SG4) enriched in hydrogen and carbon dioxide and depleted of carbon monoxide compared to the first synthesis gas stream (SG3), using the first synthesis gas stream (SG3) or a part thereof;

providing a first hydrogen stream (HG1) using hydrogen from the second synthesis gas stream (SG4) and by means of a first pressure swing adsorption unit (204);

providing a carbon dioxide stream (CG1) using carbon dioxide from the second synthesis gas stream (SG4) and by means of a carbon dioxide separation unit (205), and

processing a first residual gas stream (RG2) or a part thereof remaining downstream of where the first hydrogen stream (HG1) and the carbon dioxide stream (CG1) are provided, obtaining a second hydrogen stream (HG2) and a second residual gas stream (RG3), **characterized in that** the second residual gas stream (RG3) is returned to a position directly upstream of where the carbon dioxide stream (CG1) is provided.

2. Method (10) according to claim 1, characterized in that the first residual gas stream (RG2) is processed by means of a second pressure swing adsorption unit (206') to obtain the second residual gas stream (RG3).

3. Method (10) according to any of claims 1 or 2, characterized in that the first synthesis gas stream (SG3) is provided from a feed gas stream (FG) containing one or more hydrocarbons by means of autothermal reforming (201) or partial oxidation.

4. Method (10) according to claim 3, characterized in that the feed gas stream (FG) is supplied to the autothermal reforming (201) or to the partial oxidation without prior reforming.

5. Method (10) according to claim 3, characterized in that a product stream (SG1) for feeding into the autothermal reforming (201) or the partial oxidation is formed from the feed gas stream (FG) in an adiabatic reforming unit (200).

6. Method (10) according to any of claims 1 to 3, characterized in that the first hydrogen stream (HG1) is provided upstream of where the carbon dioxide stream is provided (CG1).

7. Method (10) according to any of the preceding claims, characterized in that providing the second synthesis gas stream (SG4) comprises converting carbon monoxide to carbon dioxide and hydrogen using a water-gas shift reaction.

8. Method (10) according to any of the preceding claims, characterized in that the second hydrogen stream (HG2) or a part thereof is supplied to a material and/or thermal recovery.

9. Method (10) according to claim 8, characterized in that the material and/or thermal recovery takes place within the process internally.

10. Method (10) according to any of claims 1 to 9, characterized in that a drying unit is arranged upstream of the carbon dioxide separation unit (205).

11. System for producing hydrogen and/or ammonia, comprising a device for providing a first synthesis gas stream (SG3) containing hydrogen, carbon monoxide, and carbon dioxide; a conversion device for providing a second synthesis gas stream (SG4) enriched in hydrogen and carbon dioxide and depleted of carbon monoxide compared to the first synthesis gas stream

(SG3), using the first synthesis gas stream (SG3) or a part thereof; a first pressure swing adsorption unit (204) for providing a first hydrogen stream (HG1) using hydrogen from the second synthesis gas stream (SG4); a carbon dioxide separation unit (205) for providing a carbon dioxide stream (CG1) using carbon dioxide from the second synthesis gas stream (SG4); and a separation device by means of which a first residual gas stream (RG2) or a part thereof remaining downstream of where the first hydrogen stream (HG1) and the carbon dioxide stream (CG1) are provided can be separated to obtain a second hydrogen stream (HG2) and a second residual gas stream (RG3), **characterized in that** it comprises a feedback device via which the second residual gas stream (RG3) can be fed back to a position directly upstream of where the carbon dioxide stream is provided (CG1).

12. The system according to claim 11, which is configured to carry out a method (10) according to any of claims 1 to 10.