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# (54) PLANT AND METHOD FOR OBTAINING LIQUID METHANE FROM A METHANE-CONTAINING MIXTURE

(57) Plant for obtaining liquid methane from a methane-containing mixture, said plant comprising a separation zone (100) in which the methane, contained in a gas flow (F1) coming from a source of gas mixture is separated from the other gases (F2, F3) forming said gas flow (F1), wherein it comprises a liquefaction station (200, 300) configured to obtain liquefaction of at least one com-

ponent (F2, F3) of the gas flow (F1) obtained from said separation, wherein said liquefaction station (200, 300) operates at low temperature, and wherein said component (F3) to be liquefied comprises methane. The invention further provides a method for obtaining liquid methane by using said plant.

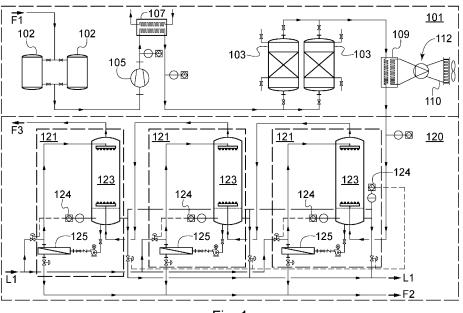


Fig. 1

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#### Technical field of the invention

**[0001]** The present invention relates to the field of gas separation, and more particularly, it relates to a plant and a method for obtaining liquid methane from a methanecontaining mixture.

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**[0002]** The present invention finds application mainly, though not exclusively, in obtaining liquid methane downstream of biogas production facilities using fermentation of organic waste from vegetal or animal residues, for example originating from waste disposal.

#### Prior Art

[0003] In the field of organic waste disposal, several technologies and installations are known adapted to produce biogas, i.e. gases obtained through degradation of various organic substances contained in agricultural or animal residues. Residues useful for biogas production may have several origins: agroindustrial waste, food industry waste (waste flours or expired products), livestock industry waste (effluents from animals or carcasses); crops can also be used specially devised for being harvested and chopped in order to produce a "biomass" from which biogas is obtained in appropriate facilities. Biomasses commonly used for this purpose are, for example, corn, sugar sorghum, wheat, giant cane, beets. The degradation process preferably takes place in the absence of oxygen and at a controlled temperature thereby producing carbon dioxide, hydrogen and methane. By virtue of these conditions favorable to degradation, some types of bacteria degrade the organic matter and generate biogas and digestate, i.e. natural liquid fertilizer. Biogas is then converted into electric power by means of a generator, whereas the digestate is used as fertilizer for land plots for farming.

**[0004]** At present, biogas produced with known facilities and technologies is usually employed for producing electric power. Instead, the obtainment of liquid methane, or biomethane from biogas produced by said facilities has to face a series of problems.

**[0005]** Considering that obtaining biomethane from biogas essentially comprises a gas separation process, a first problem lies in that the facilities for producing biogas have to be equipped with appropriate systems for separating methane from the other gases forming the gas mixture produced by the facility.

**[0006]** After the step of separating methane from the other gases forming the biogas, another drawback relates to the methane liquefaction step. At present systems are known that are arranged to carry out methane liquefaction by means of heat exchange with large quantities of cryogenic fluids such as, for example, nitrogen. Such systems are particularly disadvantageous both for reasons of operating costs, due to the high quantities of nitrogen required, and for reasons of environmental pro-

tection, because of the known polluting effects of nitrogen released into the atmosphere.

**[0007]** A further drawback of known systems based on the use of cryogenic fluids results from the fact that they are suitable especially for use in treating large flows of biogas and are therefore unsuitable to operate with reduced flows of incoming biogas for obtaining liquid methane. Particularly, the known systems are not suitable to treat biogas flows with flow rates below 1000 m<sup>3</sup>/h.

**[0008]** An object of the present invention is therefore to overcome the drawbacks of prior art, by providing a plant and corresponding method for obtaining liquid methane from a methane-containing mixture, such as, for example, a biogas coming from degradation of organic residues.

**[0009]** A further object of the invention is to provide a plant and corresponding method for obtaining methane that are capable of operating with low energy consumption and reduced environmental impact.

**[0010]** Another object of the invention is to provide a plant and corresponding method for obtaining liquid methane that can be easily stored and transported and is ready to be used as a fuel.

**[0011]** A still further object of the invention is to provide a plant and corresponding method for obtaining liquid methane that can be made industrially in a cost-effective manner.

**[0012]** These and other objects are achieved with the plant and corresponding method for obtaining liquid methane as claimed in the appended claims.

#### Summary of the invention

**[0013]** The invention relates to a plant and corresponding method for obtaining liquid methane from a methanecontaining mixture.

**[0014]** The plant for obtaining liquid methane according to the invention comprises a set of stations mutually interlinked in series, wherein a methane-containing mixture is subjected to a set of treatments aimed at obtaining liquid methane, which can then be used as a fuel.

[0015] The plant for obtaining methane according to the present invention is arranged to be associated with a source of gas mixture comprising a set of gas components, among which at least methane. Preferably, the source of gas mixture comprises a plant for biogas production in which a fermentation system, or digester, for producing biogas by degradation of organic residues is housed. The biogas coming from the fermentation system, or digester, usually comprises, besides methane, also carbon dioxide, sulphur dioxide and other components, among which ammonia and trace oxygen, hydrogen and nitrogen in traces and also various types of dusts.

**[0016]** The plant according to the invention is preferably a small-sized plant, or micro-plant, capable of operating with extremely reduced incoming biogas flows in order to obtain liquid methane. Specifically, the plant ac-

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cording to the invention is capable of liquefying methane flows smaller than 300  $\text{m}^3\text{/h}$ , more preferably with methane flows between 125  $\text{m}^3\text{/h}$  and 250  $\text{m}^3\text{/h}$ .

**[0017]** In the plant according to the invention, there is defined a separation zone comprising a first pre-treatment station for pre-treating the mixture containing methane and coming from the source of gas mixture, said pre-treatment station being arranged to purify said gas flow, and a second treatment station arranged to separate the components of the gas flow.

**[0018]** Advantageously, the plant for obtaining liquid methane according to the invention further comprises a liquefaction station located downstream of said treatment station and configured to carry out liquefaction of the least one gas flow component obtained as a result of separation, wherein the liquefaction station is configured to operate at low temperature, and wherein the component to be liquefied comprises a methane-containing gas flow for obtaining liquid methane.

[0019] Advantageously, the second treatment station is configured to separate the components  $\mathrm{CO}_2$  and methane  $\mathrm{CH}_4$  contained in said mixture exiting the first pretreatment station. By virtue of the fact that the separation zone is adapted to separate the various components of the mixture, a gas flow containing a high concentration of methane is obtained from said separation. Advantageously, the gas flow containing methane is then subjected to a liquefaction process for obtaining liquid methane, which can be easily stored and transported and is intended for subsequent use as a fuel.

**[0020]** According to the invention, the second treatment station comprises a plurality of separation units comprising spraying towers associated with a circuit arranged to deliver an incoming liquid to said towers and to appropriate extraction membranes allowing separation of carbon dioxide, in order to obtain the gas flow containing a high concentration of methane. Preferably, said incoming liquid supplied to the spraying towers is water. Due to the fact that the plant comprises a plurality of spraying towers associated with appropriate extraction membranes, separation of carbon dioxide takes place by means of a first step of absorption with water and a subsequent step of extraction of carbon dioxide by means of the extraction membranes.

[0021] Preferably, the spraying towers and appropriate extraction membranes are arranged in series, in a chain-like manner. Advantageously, thanks to the configuration according to which the spraying towers and the extraction membranes are arranged in series, in a chain-like manner, it is possible to obtain separation of carbon dioxide with a gradually decreasing percentages, whereby the last spraying tower will effect absorption of a carbon dioxide percentage much smaller than the one absorbed and extracted by means of the first spraying tower of the chain. Furthermore, thanks to the chain-like configuration of the spraying towers and the extraction membranes, the load losses upon passage of the gas flow from a spraying tower to a subsequent one are minimal, and

thus a remarkable energy saving is obtained.

**[0022]** According to the invention, upstream of the separation zone there are advantageously provided a volumetric compressor and a corresponding heat exchanger, which allow the methane-containing mixture to reach optimal conditions, i.e. certain pressure and temperature levels, for starting the separation process carried out in said separation zone arranged downstream.

**[0023]** The gas flow obtained from separation, or residual gas, is a gas having a high concentration of methane, i.e. rich in methane, which gas, according to the invention, is then subjected to a liquefaction process. In order to obtain liquefaction of the methane-rich gas flow it is necessary to ensure favorable conditions for this process. Such favorable conditions mainly provide that the methane-rich gas flow reaches a very low temperature, preferably between -125°C and -150°C, more preferably a temperature of -130°C.

**[0024]** For this purpose, advantageously, the liquefaction station of the plant according to the invention is configured to carry out a series of steps of cooling the methane-rich gas flow originating in the separation zone. By virtue of the fact that the liquefaction station is configured to carry out cooling of the methane-rich gas flow for subsequent steps, until the desired temperature in the above-mentioned range is reached, a reduction in the energy consumption of the whole plant is achieved.

**[0025]** Advantageously, carrying out liquefaction of methane by means of a series of cooling steps results in that the methane does not actively take part in the liquefaction processe, unlike what happens in those liquefaction processes employing expansion processes, such as, for example, the known Linde or Claude processes; in contrast, the invention provides for indirect liquefaction of methane by using a chain of refrigeration units in order to allow methane to change from the gaseous state to the liquid state. Preferably, the change of methane from the gaseous state to the liquid state takes place by exploiting the frigories of the state change from the gaseous state to the liquid state of the refrigerant fluid used in the last cooling step of the chain of refrigeration units employed.

**[0026]** The liquefaction station comprises corresponding compression means allowing the methane-rich gas flow to reach certain pressure levels favorable to the liquefaction process. According to the invention, said pressure levels are preferably about 30 bar.

[0027] Advantageously, according to the invention, the liquefaction station comprises a set of heat exchangers arranged to facilitate reaching of the liquefaction temperature during carrying out of the series of cooling steps. The heat exchangers are suitably devised for reaching certain values of heat exchange coefficient in the various steps of cooling the methane-rich gas flow. Preferably, said heat exchangers comprise cylindrical elements, or tubes, having a hollow structure and a plurality of radial fins arranged concentrically along respective generatrices both inside and outside said hollow structure. The

fins extend radially from the center of said cylindrical elements and, in addition, they may be at least partially interrupted along said generatrices, i.e. they have interruptions in the axial direction, in order to create more turbulences during the heat exchange step, i.e. said fins may be interrupted along the corresponding generatrices.

**[0028]** According to the invention, preferably, the cylindrical elements are arranged coaxially in series and preferably rotated relative to one another, so as to cause a discontinuity in the radial fins in order to generate a corresponding greater turbulence effect over the methane-rich gas flow.

**[0029]** Still according to the invention, all the heat exchangers of the plant are made according to the above-described configuration.

**[0030]** The liquefaction station further comprises pump means arranged to distribute the liquefied gas flow, i.e. the liquid methane, and suitable storage tanks for subsequent transport and use thereof as a fuel.

[0031] In order to ensure correct operation of the plant in case of downtime, the liquefaction station comprises a safety system, equipped in turn with a plurality of expansion vessels, or reservoirs, to be associated with each cooling step. The expansion vessels or reservoirs preferably contain hydraulic oil and are provided each with a safety valve. The expansion vessels are also associated with a main tank containing hydraulic oil and are configured to convert a pressure change above a threshold value into an increase in volume inside said expansion vessel, this causing exit of the oil towards the main tank

**[0032]** The safety system further comprises respective pump means configured to bring the hydraulic oil back from the main tank towards the corresponding expansion vessels in order to allow the gas flow to reach its working pressure in a corresponding cooling step.

**[0033]** The method for obtaining liquid methane according to the invention comprises the steps of:

- providing a gas mixture containing methane;
- subjecting said gas mixture to a step of separating the various components of the gas mixture;
- subjecting at least one component obtained from the separation step to a liquefaction step, wherein said liquefaction step is carried out at low temperature, and wherein said component to be liquefied comprises a gas flow rich in methane.

**[0034]** The separation step provides for a first pretreatment step in which the gas mixture is subjected to a purification step in order to eliminate traces of polluting components present in the gas mixture.

**[0035]** Advantageously, the liquefaction step provides for subjecting the gas flow rich in methane to a compression step adapted to bring the gas flow to a certain pressure of about 30 bar and cool down said component by means of several subsequent cooling steps, until a tem-

perature between -125°C e -150°C, more preferably closer to -130°C, is reached.

**[0036]** Advantageously, subjecting the component to be liquefied to subsequent cooling steps makes it possible to reduce the energy consumption of the plant according to the invention.

**[0037]** The plant and corresponding method for obtaining liquid methane according to the invention preferably find application in the field of the production of biogases from degradation of organic residues of animal or vegetal origin. More preferably, the plant according to the invention is configured to produce certain amounts of liquid methane, by operating with even very low biogas flows, typically below 1000 m<sup>3</sup>/h, and flows of methane to be liquefied lower than 300 m<sup>3</sup>/h, preferably ranging from 125 m<sup>3</sup>/h to 250 m<sup>3</sup>/h.

#### Brief Description of Drawings

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**[0038]** A preferred embodiment of the invention will be described by way of non-limiting example with reference to the annexed figures, in which:

- Fig. 1 shows a layout of the separation zone for separating the components of a mixture containing methane;
- Fig. 2A is a layout of the liquefaction station for liquefying a first component of said mixture;
- Fig. 2B is a layout of the liquefaction station for liquefying a second component of said mixture;
  - Fig. 3 is a perspective view of a compressor used in the plant according to the invention;
  - Fig. 4 is a schematic sectional view of the compressor of Fig. 3;
  - Fig. 5 is a perspective view of a heat exchanger according to a preferred embodiment of the invention;
  - Fig. 6 is a top view of the heat exchanger of Fig. 5;
  - Fig. 7 is a layout of the safety system of the liquefaction station.

## Description of a Preferred Embodiment of the Invention

**[0039]** The plant for obtaining liquid methane according to the invention comprises a set of mutually interlinked stations in which a methane-containing mixture is subjected to a set of treatments in order to obtain liquid methane, which can then be used as a fuel.

**[0040]** The plant for obtaining methane of the present invention is associated with a source of gaseous mixture of gases including, among others, methane.

**[0041]** In the preferred embodiment of the invention, which will be described in detail below with reference to the annexed drawings, the source of gas mixture comprises a fermentation system, or digester, (not shown) and the gas mixture comprises a biogas flow F1 originating from the degradation of organic residues that has taken place in said digester. In this embodiment, the bi-

ogas flow F1 coming from the fermentation system comprises methane, carbon dioxide, sulphur dioxide, ammonia and other components such as, for example, oxygen, hydrogen and nitrogen.

**[0042]** According to this preferred embodiment of the invention, the plant comprises a separation zone 100, schematically shown in Fig. 1, in which separation of the various carbon dioxide component from the biogas flow F1 coming from the source of gas mixture takes place.

**[0043]** The separation zone 100 comprises, in turn, a first pre-treatment station 101 for pre-treating the biogas flow F1 coming from the fermentation system, said pre-treatment station being arranged to purify the gas flow F1, and a second treatment station 120 arranged to separate the components of the purified gas flow F1.

[0044] In the illustrated embodiment, the first pre-treatment station 101 comprises at least one active carbon filter 103, in the illustrated embodiment two filters 103 arranged in parallel, equipped with active carbon filtration means and arranged to purify the biogas flow F1 by removing the sulphur dioxide component and, at least partially, other polluting components such as, for example, ammonia. Ammonia in particular is removed only partially by means of the filter 103, as said filter is specific for sulphur dioxide, whereas the remaining part is dissolved in the liquid of the treatment station 120, typically water, as will become more apparent from the following description.

**[0045]** The first pre-treatment station 101 further comprises compression means 105 and heat exchangers 107, 109, which allow the biogas flow F1 to reach certain pressure and temperature condition, which will promote the separation process carried out by the second treatment station 120.

**[0046]** In the illustrated embodiment, the heat exchangers 107, 109 may be of the air-gas type. In addition, the heat exchanger 109 is preferably suitably devised, as will be described in the present description here below. The heat exchanger 107 may be of the conventional type, or it may also be devised similar to the heat exchanger 109.

**[0047]** The second treatment station 120 is configured to separate the carbon dioxide and methane components from the biogas flow F1 by means of a spraying or wet process.

[0048] In the illustrated embodiment, the second treatment station 120 comprises a plurality of separation modules 121 comprising spraying towers 123 associated with a circuit for delivering an incoming liquid L1 to said towers 123. The separation modules 121 further comprise suitable extraction membranes 125 allowing separation of carbon dioxide F2 in order to obtain a methane-rich gas flow F3. In the illustrated embodiment, the liquid L1 supplied by said circuit consists mainly of water. The membranes 125 are selective for  ${\rm CO_2}$  and the ammonia present in the flow F1 is dissolved in the water L1 and, as it accumulates, the pH value of said liquid L1 rises. A pH meter 124 connected to each tower, or reactor, 124

opens a water purge circuit for purging water from each tower 123 when the pH value exceeds a predetermined threshold, and directs the recirculated liquid to the osmotic membrane separating ammonia inside the tower 123.

**[0049]** Referring to Fig. 1, the path of the biogas flow F1 in the separation zone 100 of the plant according to the invention will be described below.

**[0050]** In the separation zone 100, the biogas flow F1 coming from the fermentation system or digester follows a specific path aimed at separating the biogas flow F1 into its various components such as, for example, the carbon dioxide component F2 and the methane component F3, as will become more apparent from the following description.

**[0051]** In a preferred embodiment of the invention, the biogas flow F1 entering the separation zone 100 has a temperature between 36°C and 46°C, and the pressure value is substantially equal to the value of the atmospheric pressure.

[0052] The biogas flow F1, in the illustrated embodiment, is subjected to a preliminary filtration step to eliminate dusts, which step is carried out by means of appropriate mechanical separators 102, and thereafter it is subjected to a compression step carried out by means of a volumetric compressor 105 in order to obtain a pressure value of about 5 bar for said biogas flow F1. During the compression step, the temperature of the biogas flow F1 rises until it reaches a value of about 170°C. Subsequently, in order to promote the process of separation of the biogas flow into its various components, the biogas flow F1 is subjected to a cooling step carried out by means of the air-gas heat exchanger 107. As a result of said cooling, the temperature of the biogas flow F1 is brought again to a temperature around the value of the ambient temperature, which in this particular case is about 35°C. [0053] After the biogas flow F1 has reached the temperature and pressure levels favorable to the separation process, it is subjected to a purification step through the filter 103 provided with active carbon filtration means and arranged downstream of the air-gas heat exchanger 107. [0054] After the step of purification of the biogas flow F1, it is necessary to cool down the biogas flow F1 until a temperature closer to 0°C, more preferably of about 1°C, is reached. Cooling of the purified biogas flow F1 is carried out by means of the heat exchanger 109 in which a refrigerating fluid such as, for example, R448A circulates, cooled by a chiller 112 associated with a further air-gas heat exchanger 110.

[0055] Under the conditions in which the biogas flow F1 has been cooled down and has reached the temperature of 1°C and the pressure value is about 5 bar, the solubility of carbon dioxide F2 in the liquid L1 is ensured. It is thus possible to subject the biogas flow F1 to the separation process carried out by the second treatment station 120 by means of the separation modules 121.

[0056] The purified gas flow F1 exiting the first pretreatment station 101 flows into the second treatment

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station 120, which is adapted to separate the purified biogas flow F1 essentially into two main components: the carbon dioxide component F2 and the methane component F3.

[0057] In the embodiment illustrated in Fig. 1, the second treatment station 120 comprises three separation modules 121 arranged in series. Each of the separation modules 121 comprises in turn a spraying tower 123 associated with a corresponding extraction membrane 125 and further associated with a shared circuit adapted to deliver an incoming liquid L1 to said spraying towers 123. Preferably, the liquid L1 entering said towers essentially consists of water.

**[0058]** In the illustrated embodiment, the purified gas flow F1 exiting the first pre-treatment station 101 flows through the separation modules 121, in succession one by one, gradually effecting extraction of the carbon dioxide component F2 by means of the corresponding membranes 125 associated with the spraying towers 123.

[0059] Therefore, the purified biogas flow F1 enters the first separation module 121, i.e. the first spraying tower 123, where separation of a part of the carbon dioxide component F2 from said biogas flow F1 takes place by means of a water L1 spraying process. Said CO2 component is further extracted by means of the corresponding extraction membrane 125 arranged downstream of said spraying tower 123. The gas flow F3 resulting from extraction of a part of the carbon dioxide component F2 is a gas flow comprising a high concentration of methane. **[0060]** The gas flow exiting the first separation module 121 enters the second separation module 121 where it is subjected to the same separation process, i.e., inside the respective spraying tower 123, a further part of the carbon dioxide component F2 is separated by means of spraying or wet separation process, which further part is further extracted by means of the corresponding membrane 125 arranged downstream of the spraying tower 123.

**[0061]** The gas flow exiting the second separation module 121 consists of a gas flow with a methane concentration even higher than that of the gas flow exiting the first separation module 121. Subsequently, the gas flow passes through the third separation module 121, where, by means of a spraying process with water L1, there is carried out a further separation of the carbon dioxide component F2, which is extracted by means of the corresponding extraction membrane 125 associated with the tower 123. Therefore, at the outlet of the third separation module 121, two flows exiting the spraying tower 123 are obtained, namely a flow of the carbon dioxide component F2 on one hand, and a gas flow F3 rich in methane, i.e. with very high methane concentrations, on the other hand.

[0062] The temperature of the water L1 used by the separation modules 121 is preferably 1°C and once the carbon dioxide CO<sub>2</sub> component has been separated by means of the membranes 125, the water L1 is advantageously re-circulated, this being of advantage for the

management of the plant.

[0063] At the end of the separation process, separation of the biogas flow F1 into its main components, i.e. the carbon dioxide component F2 and the methane component F3, is obtained. The methane component F3 also comprises a part of moisture that will be discarded by means of a dehydration filter, as described below.

**[0064]** At this stage, it is possible to subject the respective carbon dioxide F2 and methane F3 components to a liquefaction process.

**[0065]** Advantageously, the plant for obtaining liquid methane according to the invention comprises a lique-faction station 200, 300 configured to carry out liquefaction of at least one component F2, F3 of the biogas flow F1, which component has been obtained from the separation of the various components. The liquefaction station 200, 300 is advantageously configured to operate at low temperature.

**[0066]** Advantageously, according to the invention, the component that is subjected to the liquefaction step is the methane component F3.

**[0067]** According to a preferred embodiment of the invention, it is further provided to also subject the carbon dioxide component F2 to a liquefaction process.

[0068] Referring to Fig. 2A, there is schematically illustrated the process of liquefaction of the carbon dioxide component F2. The flow of carbon dioxide F2 exiting the separation zone 100 is at a temperature of about 0°C and a pressure of 5 bar. In the liquefaction step, the carbon dioxide component F2 is subjected to a first compression step carried out by means of an appropriate compressor 201 according to the invention. In the illustrated embodiment, a volumetric compressor 201 is provided, the operation of which will be described in detail below. The compressor 201 allows bringing the carbon dioxide component F2 to a pressure of about 20 bar. During the compression step, the temperature of the carbon dioxide component F2 rises until it reaches a temperature value of essentially 40°C. In order to effect liguefaction of the carbon dioxide component F2, the carbon dioxide component F2 is subsequently cooled down by means of an appropriate heat exchanger 203 arranged downstream of the compressor 201 associated with a gas-air heat exchanger 205 to obtain a temperature of about -20°C. At the outlet of said heat exchanger 203 there is obtained liquid carbon dioxide F2'.

**[0069]** Referring to Fig. 2B, there is illustrated the liquefaction station 300 used to carry out the process of liquefaction of the methane component F3 exiting the separation zone 100.

**[0070]** In order to obtain liquefaction of the methane component F3 it is necessary to ensure favorable conditions for said process. Such favorable conditions provide for the methane component F3 to reach a very low temperature of about -130°C, called liquefaction temperature.

[0071] To this aim, advantageously, the liquefaction station 300 of the plant according to the invention is con-

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figured to effect a series of cooling steps for cooling the methane component F3.

[0072] By effecting cooling of the methane component F3 by means of a series of cooling steps until the liquefaction temperature is reached, a remarkable reduction in the energy consumption of the whole plant is obtained. Therefore, the plant according to the invention will be capable of producing liquid methane with an energy consumption between 0.3 and 0.35 kW/kg of liquid methane, remarkably lower than the consumptions of current technologies, which are about 0.5 kW/kg of liquid methane. The invention is thus particularly advantageous in the making of small-sized plants, or micro-plants, i.e. plants capable of operating with extremely small incoming biogas flows, for example smaller than 1000 m<sup>3</sup>/h, and flows of methane to be liquefied smaller than 300 m<sup>3</sup>/h. [0073] The liquefaction station 300 comprises a dehydration filter 302 arranged at the inlet of the station 300, in order to separate especially moisture from the flow F3. Moisture is discarded from the dehydration filter 302, which is a molecular sieve with activated alumina, with hot air regeneration. Downstream of the filter 302, in the flow F3 there will remain traces of oxygen, hydrogen and nitrogen, which are gases that are incondensable at the temperature of liquefaction of methane and will therefore be found in the dome of an expansion vessel, or reservoir, 317 located upstream of the outlet of the station 300. Said residual gases are preferably sent to a small stripping column for recovering gaseous methane, present in small quantities in the mixture, whereas the remaining gases are evacuated through a torch.

**[0074]** Downstream of the filter 302 there are provided compression means 301 allowing the methane component F3 entering the liquefaction station 300 to reach certain pressure levels favorable for the liquefaction process.

**[0075]** Advantageously, the liquefaction station 300 further comprises, according to the invention, heat exchangers 305, 307, 313, 315 arranged in series and adapted to carry out the series of cooling steps according to the invention.

**[0076]** In the illustrated embodiment, the process of liquefaction of the methane component F3 provides for carrying out substantially five subsequent cooling steps. Still referring to Fig. 2B, the process of liquefaction of the methane component F3 will now be explained.

**[0077]** As a result of the fact that the liquefaction of methane takes place by means of a series of cooling steps, methane takes no active part in the liquefaction process, but it is liquefied indirectly by using the frigories of a set of refrigeration units employed for obtaining said series of cooling steps.

**[0078]** When entering the liquefaction station 300, the methane component F3 has a temperature of about 1°C and a pressure of 5 bar, which are substantially the temperature and pressure values reached by the methane component F3 exiting the separation zone 100. At first, the methane component F3 entering the liquefaction sta-

tion 300 will be subjected to a compression step by means of an appropriate compressor 301 in order to reach a pressure value of about 30 bar. In the illustrated embodiment, the compressor 301 is a volumetric compressor. As a result of the compression step, the methane component F3 reaches a temperature of about 130°C, which is then brought to ambient temperature, here substantially of about 35°C, by means of a gas-air heat exchanger 303 located downstream of the compressor 301. **[0079]** After bringing the methane component F3 to ambient temperature by means of the heat exchanger 303, the subsequent cooling steps are carried out, which provide for reaching gradually lower temperatures, until the methane component F3 reaches the liquefaction temperature of about -130°C.

**[0080]** The first cooling step provides for bringing the methane component F3 from ambient temperature, i.e. from 35°C, to a temperature lower than 0°C, namely to a temperature of about -30°C. In order to carry out said cooling step, the liquefaction station 300 comprises a first heat exchanger 305 using a refrigerating fluid, for example of the type R448A, said first heat exchanger being arranged downstream of the heat exchanger 303 and associated with a corresponding air-gas heat exchanger 306.

**[0081]** Thereafter, the methane component F3 will be subjected to a second cooling step providing for bringing said methane component F3 from the temperature of about -30°C to a temperature of about -60°C. In order to carry out the second cooling step, the liquefaction station 300 comprises a second heat exchanger 307, arranged downstream of the heat exchanger 305, which, by using a refrigerating component, such as, for example, ethylene at the gaseous state, at a pressure of about 0 relative bar and a temperature level of about -30°C, reduces the temperature level of the methane component F3.

[0082] The second cooling step further comprises a step of liquefaction of the refrigerating ethylene component used, which will be recirculated in order to exploit the sensible heat of said ethylene component in a subsequent cooling step for the methane component F3, i.e. in the fourth cooling step of the series of cooling steps. For this purpose, the liquefaction station 300 comprises a third heat exchanger 309 associated with an air-gas heat exchanger 311, which heat exchangers promote the cooling process in the second and fourth cooling steps by using the sensible heat of ethylene. Between said heat exchangers 309, 311 there is further provided a compressor 308 arranged to effect compression of ethylene from 0 bar to 30 bar. The compressor 308 is preferably a compressor of the scroll type. The temperature of ethylene is brought back to ambient temperature by means of the gas-air heat exchanger 311 and thereafter, by means of the heat exchanger 309 containing refrigerating fluid R448A, the ethylene will be brought to the temperature of -30°C, thus liquefying it, and subsequently it will be recirculated for being used in the fourth cooling step illustrated below. Inside the heat exchanger 309, ethyl-

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ene changes its state from gaseous to liquid at a constant temperature by condensation and the refrigerating fluid R448A also changes its state at a constant temperature from liquid to gaseous by evaporation. By virtue of the fact that the temperature remains constant during the step of change of state of the ethylene component from gaseous to liquid, the yield of the plant as well as of the liquefaction process is increased remarkably.

[0083] The third cooling step, following the second cooling step, provides that the methane component F3 reaches an even lower temperature level, changing from -60°C to -80°C. In order to carry out said third cooling step, the liquefaction station 300 comprises a corresponding heat exchanger 313 arranged downstream of the heat exchanger 307 and further promoting recovery of the sensible heat of the methane component F3 evaporated in the fifth cooling step before liquefaction and is recirculated inside said heat exchanger 313. Therefore, the temperature of the methane component F3 in the third cooling step is lowered by recovery of the sensible heat of said component, by recirculating said component from the fifth cooling step to the third cooling step.

[0084] The fourth cooling step provides for lowering the temperature of the methane component F3 from -80°C to -100°C. In order to carry out said fourth cooling step, the liquefaction station 300 comprises a fourth heat exchanger 315, arranged downstream of the heat exchanger 313 of the third cooling step, said fourth heat exchanger, by recirculating the ethylene component from the second cooling step, lowers the temperature of the methane component F3 to a value of about -100°C. In this step of the process, the methane component F3 changes from the gaseous state to the liquid state at a constant temperature by condensation, whereas the ethylene component changes from the liquid state to the gaseous state at constant temperature by evaporation. By virtue of the fact that the temperature remains constant during the step of change of state of the methane component F3 from gaseous to liquid, the yield of the plant as well as of the liquefaction process is increased remarkably.

[0085] The fifth cooling step provides for effecting further cooling in order to bring the methane component F3 from the temperature value of -100°C to the liquefaction temperature of about -130°C. For this purpose, in the illustrated embodiment, the liquefaction station 300 comprises an expansion vessel 317 which is arranged downstream of the heat exchanger 315, and in which expansion of the methane component F3 from a pressure value of 30 bar to a pressure value of 8 bar is effected. The pressure value of 8 bar is usually the pressure value at which the liquefied methane is stored in storage tanks or reservoirs for subsequent transport

**[0086]** In this fifth cooling step, about 30% of the methane component F3 is evaporated and recirculated, in order to recover the sensible heat of said methane component F3, necessary to effect cooling in the third cooling step.

**[0087]** Advantageously, operation of the heat exchangers 309 and 315 effecting liquefaction of the methane component and ethylene is based on latent heat rather than sensible heat.

**[0088]** The liquefaction station 300 further comprises pump means 319 associated with the expansion vessel 317 and arranged to distribute the liquefied gas flow F4 exiting the expansion vessel 317, i.e. the liquid methane, to appropriate storage tanks for subsequent transport and use of the same as a fuel.

**[0089]** Referring to Fig. 3, there is illustrated a compressor 201, 301 used in the liquefaction station 300. Advantageously, the compressor 201, 301 is preferably a volumetric compressor provided with a hydraulic feeding system arranged to receive an incoming gas flow with flow rates between 100 and 500 m<sup>3</sup>/h.

**[0090]** In the illustrated embodiment, the compressor 201, 301 comprises a first compression cylinder 211 and a second compression cylinder 311 which are arranged opposite to each other and have each a corresponding compression piston 221, 223, and in which compression of the biogas flow entering the liquefaction station 200, 300 is effected. Advantageously, the first compression cylinder 211 and the second compression cylinder 213 are coaxial to each other and are arranged opposite to each other.

**[0091]** The compressor 201, 301 comprises a third cylinder 215 also provided with a corresponding piston 225, said third cylinder being arranged centrally between the first compression cylinder 211 and the second compression cylinder 213 and coaxial to said first 211 and second 213 compression cylinders.

**[0092]** The hydraulic feeding system of the compressor 201, 301 comprises a reserve tank 217 containing hydraulic oil and arranged so as to define a substantially "T"-shaped configuration together with the coaxial cylinders, i.e. the first compression cylinder 211, the second compression cylinder 213 and the third central cylinder 215.

**[0093]** The hydraulic feeding system of the compressor 201, 301 further comprises adjusting means 219 and pump means 220 for the hydraulic oil flow, said pump means being arranged between the reserve tank 217 and the coaxial cylinders 211, 213, 215. In the illustrated embodiment, the adjusting means 219 are a proportional valve and the pump means 220 are a variable flow rate hydraulic pump.

**[0094]** Referring to Fig. 4, the operation of the compressor 201, 301 will now be described. The gas flow Fi entering the compressor 201, 301 has a pressure of substantially 5 bar and can be formed either by the methane component F3 or by the carbon dioxide component F2, depending on the component to be subjected to the liquefaction process in the respective liquefaction station 200, 300.

**[0095]** The gas flow Fi entering the compressor 201, 301 enters the first compression cylinder 211 and actuates, by means of its pressure, the piston 221 of said first

cylinder 211. The piston 225 of the central cylinder 215 is actuated by the oil flow coming from the reserve tank 217. At the beginning of the stroke, the thrust of the piston 225 of the central cylinder 215 is substantially reduced because the counter-pressure of the gas flow Fi is minimal. In this initial step, the flow rate of the oil coming from the reserve tank 217 will be high, in accordance with the flow rate capacity of the pump means 220. As the pressure of the incoming gas Fi rises, it is necessary to increase also the thrust of the piston 225 of the central cylinder. To this aim, it is necessary to increase the pressure of the oil coming from the reserve tank 217. The flow rate adjusting means, during the stroke of the piston 225 of the central cylinder 215, perform a function of adjusting the flow rate of the pump means 220 so as to maintain the energy consumption thereof constant. The exiting flow gas Fu subjected to compression in the first compression cylinder 213 or in the second compression cylinder 215 and intended to be subjected to the liquefaction process will have a pressure of about 30 bar.

[0096] The compressor 201, 301 further comprises detection means such as a sensor arranged to detect the end of the stroke of the piston 225 of the central cylinder 215 and generate an electric control signal for reversing the oil flow towards the reserve tank 217. In a preferred embodiment of the invention, the piston 225 comprises a small magnet which cooperates with an inductive sensor, arranged on the wall of the cylinder 215 housing said piston 225, to generate a signal indicative of the end of the stroke of the piston 225 in order to allow reversing the direction of the linear movement of said piston 225. [0097] Referring to Figs. 5 and 6, there is illustrated a

**[0098]** Advantageously, the heat exchangers 305, 307, 313, 315 used especially in the liquefaction station 300 are configured to reach high values of exchange coefficient in the various cooling steps.

heat exchanger 305, 307, 313, 315 according to the in-

[0099] The heat exchanger comprises a tubular cylindrical element 3 having a hollow structure and a plurality of radial fins arranged concentrically along respective generatrices both inside and outside said hollow structure. The fins 5 extend radially from the center of the cylindrical element 3 and at least some of them 5 can be interrupted in order to create more turbulences during the heat exchange step, i.e. said fins may be interrupted along the corresponding generatrices. In the illustrated embodiment, the diameter of the inner part of the cylindrical element 3 is between 60 cm and 70 cm, more preferably of 65 cm, whereas the outer diameter of the heat exchanger is between 80 cm and 95 cm, more preferably of 90 cm.

**[0100]** Advantageously, said cylindrical elements 3 can be associated in series along the longitudinal axis in order to attain the desired length for the heat exchanger 305, 307, 313, 315. For this purpose, the ends 7,9 of a heat exchanger 305, 307, 313, 315 are suitably configured to allow interlinking with other similar heat exchang-

ers in order to attain the desired length. In the illustrated embodiment, the cylindrical element 3 has a length of about 200 cm.

**[0101]** According to the invention, the cylindrical elements 3 are arranged coaxially in series and are preferably rotated with respect to one another, so as to cause discontinuity in the radial fins 5 in order to generate a corresponding greater turbulence effect on the methanerich gas flow touching said radial fins. More preferably, a first cylindrical element 3 is rotated relative to a following second cylindrical element 3 so that a fin 5 of the first cylindrical element 3 is substantially sandwiched between two fins of an adjacent cylindrical element 3. In order to ensure proper operation of the plant in case of downtime, the liquefaction station 300 further comprises a safety system 400 schematically shown in Fig. 7.

**[0102]** The safety system 400 is preferably applied to all the refrigerating circuits of the plant in which a liquid phase is present. The safety system is necessary to remedy the detrimental effects due to a sudden pressure increase caused by boiling of the liquid phase, in case of sudden stoppage of the plant. According to the illustrated embodiment of the invention, the safety system 400 is applied to all refrigerating circuits of the plant, because a liquid phase is present in them all.

**[0103]** The safety system 400 mainly comprises a plurality of expansion vessels, or tanks, 401 preferably containing hydraulic oil balancing the cooling pressure during the various cooling steps. The safety system further comprises a main tank 403 containing in turn hydraulic oil. The expansion vessels 401 are further provided with a safety valve 402 arranged downstream of the expansion vessels. The valve 402 connects said expansion vessels 401 with the main tank 403. The expansion vessels 401 are further configured to turn a pressure change, above a threshold level, into a corresponding increase in volume within the expansion vessel 401, this resulting in the oil being transferred from the expansion vessel 401 towards the main tank 403, through the valve 402 which is opened when said threshold is exceeded.

**[0104]** The safety system 400 further comprises corresponding pump means 405, arranged between said expansion vessels 401 and the main tank 403 and configured to bring back the hydraulic oil from the main tank 403 towards the corresponding expansion vessels 401, thus allowing the gas flow to reach the correct operating pressure.

**[0105]** The valve 402 is calibrated to intervene at a pressure slightly higher than that of the refrigerating circuit with which it is associated. For example, in the case of the refrigerating circuit in which ethylene circulates, described in Fig. 2B, the valve 402 is calibrated to intervene when the pressure in the circuit exceeds 32 bar, the maximum pressure of the gas in the circuit being 30 bar.

**[0106]** The invention as described and illustrated is susceptible to several modifications and variations all falling within the same inventive concept.

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1. A plant for obtaining liquid methane from a methane-containing mixture, said plant comprising a separation zone (100) in which the methane, contained in a gas flow (F1) coming from a source of gas mixture is separated from the other gases (F2, F3) forming said gas flow (F1), characterized in that it comprises a liquefaction station (200, 300) configured to obtain liquefaction of at least one component (F2, F3) of the gas flow (F1) obtained from said separation, wherein said liquefaction station (200, 300) operates at low temperature, and in that said component (F3) to be liquefied comprises methane.

- 2. The plant according to claim 1, wherein said separation zone (100) comprises a first pre-treatment station (101) in which purification of the gas flow (F1) coming from the source of gas mixture takes place by removing respective pollutant components, and a second treatment station (120) in which separation of the gases forming the gas flow (F1) takes place.
- 3. The plant according to claim 2, wherein the component (F2) is carbon dioxide and the carbon dioxide has gradually decreasing percent concentrations upon passage from a spraying tower (123) to a successive spraying tower (123), whereby a last spraying tower (123) of the chain will effect absorption of a percent carbon dioxide remarkably lower than the one absorbed and extracted by the first spraying tower (123) belonging to the chain.
- 4. The plant according to claim 2 or 3, wherein said liquid entering said spraying towers (123) is water, and the component (F2) of said gas flow (F1) is subjected to a first absorption step in which said component (F2) is absorbed with the water, and to a second extraction step in which said component (F2) is extracted by means of extraction membranes (125) associated with the spraying towers (123).
- 5. The plant according to claim 2, 3 or 4, wherein the first pre-treatment station comprises compression means (105) and respective heat exchangers (107, 109) capable of bringing the gas flow (F1) to favorable conditions for the process of separation of the various components (F2, F3), and at least one mechanical separator (102) and active carbon filtration means (103) for removing said pollutant components.
- 6. The plant according to any of claims 2 to 5, wherein said second treatment station (120) comprises a plurality of separation modules (121) arranged in series and comprising spraying towers (123) associated with a circuit adapted to deliver an incoming liquid (L1) to said towers (123) and a plurality of appropriate

- extraction membranes (125) in which extraction of at least one component (F2) of said gas flow (F1) takes place.
- 7. The plant according to any of the preceding claims, wherein said liquefaction station (300) is configured to carry out a set of steps of cooling at least one component to be liquefied (F3) of said gas flow (F1) and comprises compression means (301) causing said component (F3) to be liquefied to reach a certain pressure, and a set of heat exchangers (305, 307, 313, 315) which are arranged downstream of said compression means (301) and in which said cooling steps of said component to be liquefied (F3) are carried out.
  - 8. The plant according to claim 7, wherein said component to be liquefied (F3) comprises methane, and wherein the liquefaction of said component (F3) takes place indirectly by using the frigories of said set of heat exchangers (305, 307, 313, 315) arranged in a chain-like manner and arranged to cause a change of state of said component (F3) from a gaseous state to a liquid state.
  - **9.** The plant according to claim 7 or 8, wherein said heat exchangers comprise:
    - at least one first heat exchanger (305) using a cooling fluid, preferably R448A;
    - at least one third heat exchanger (309) located downstream of said first heat exchanger (305) and having an ethylene-based cooling component capable of changing from a gaseous state to a liquid state at constant temperature by condensation;
    - at least one second heat exchanger (307) interposed between said first heat exchanger (305) and said third heat exchanger (309) and arranged to receive the cooling fluid of the first heat exchanger and the ethylene-based cooling component at the gaseous state of the third heat exchanger (309);
    - at least one fourth heat exchanger (315) arranged to receive said liquefied ethylene-based component changing from a liquid state to a gaseous state by evaporation, and wherein the component to be liquefied (F3) changes from a gaseous state to a liquid state by condensation.
- 10. The plant according to claim 7 or 8, wherein a respective heat exchanger of said set of heat exchangers (305, 307, 313, 315) comprise at least one cylindrical element (3) having a hollow structure and a plurality of fins (5) arranged concentrically along respective generatrices both inside and outside said hollow structure and extending radially from the center of said cylindrical element (3), and wherein at least

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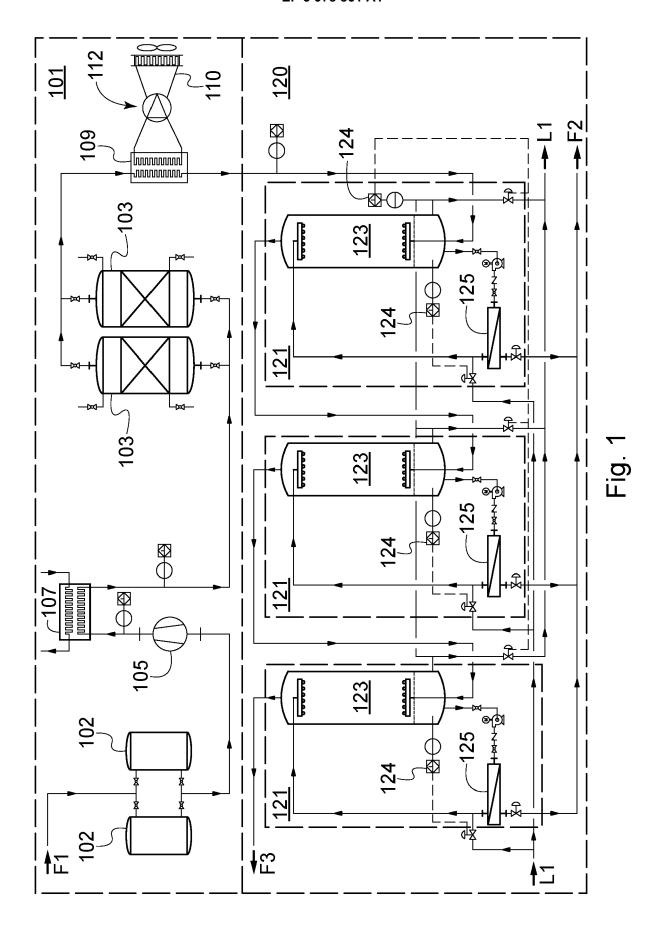
some of said fins (5) are interrupted along said generatrices.

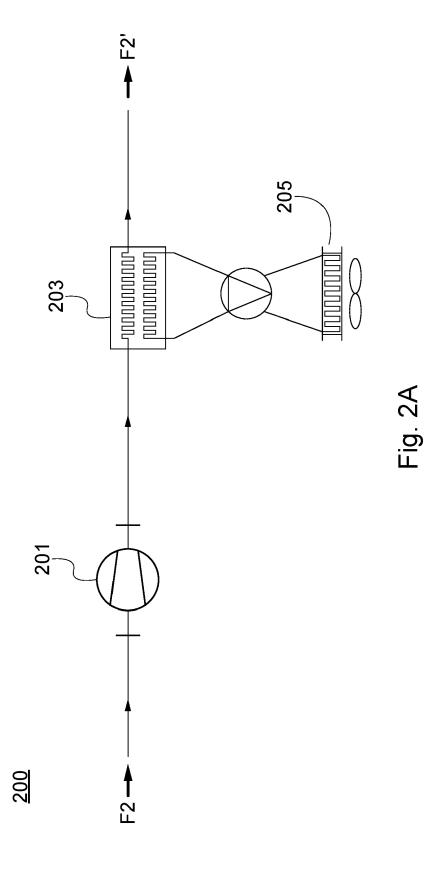
- **11.** The plant according to any of claims 5 to 10, wherein said compression means comprise at least one volumetric compressor (201,301) comprising:
  - a first compression cylinder (211) provided with a corresponding compression piston (221) and adapted to receive an incoming gas flow (Fi) and to discharge a compressed gas flow (Fu);
  - a second compression cylinder (213) provided with a corresponding compression piston (223) and adapted to receive an incoming gas flow (Fi) and to discharge a compressed gas flow (Fu), wherein said first (211) and said second (213) compression cylinders are arranged coaxially opposite to each other;
  - a third central cylinder (215) provided with a corresponding piston (225) and arranged centrally between said first (211) and said second (213) compression cylinders, wherein said central cylinder (215) is arranged coaxial to said first (211) and said second (213) compression cylinders:
  - a hydraulic feeding system comprising a reserve tank (217) containing hydraulic oil and arranged substantially to define a "T"-shaped configuration with said first (211) and second (213) compression cylinders and said central cylinder (215) coaxial to one another.
- 12. The plant according to claim 7, 8, 9 or 10, wherein said liquefaction station (300) comprises a safety system (400) including a plurality of expansion vessels (401) associated with each cooling step, wherein said expansion vessels (401) contain hydraulic oil and are provided in turn with a safety valve (402), said expansion vessels being associated with a main tank (403), this too containing hydraulic oil and being associated with pump means (405) provided between said main tank and said expansion vessels (401), wherein said expansion vessels (401) are configured to turn a pressure change, above a threshold value, into an increase in volume within said expansion vessel (401) with consequent oil transfer to the main tank (403).
- **13.** A method for obtaining liquid methane from a gas mixture, said method comprising the steps of:
  - providing a gas flow (F1) comprising a number of component gases, among which at least methane (F3);
  - subjecting said gas flow (F1) to a step of separating the various components (F2, F3);
  - subjecting at least one component (F2, F3) obtained from the separation step to a liquefaction

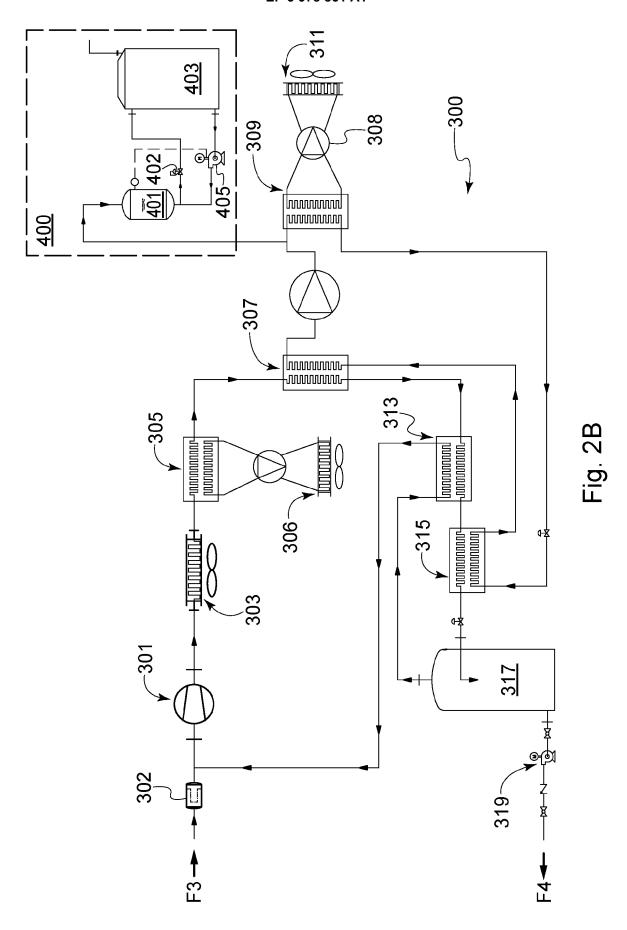
step,

**characterized in that** said liquefaction step is carried out at low temperature, and **in that** said component (F3) to be liquefied comprises methane.

- 14. The method according to claim 13, wherein said separation step comprises a first step of purifying the gas flow (F1) and a second step of separating the components (F2, F3) of said purified gas flow (F1), and wherein the liquefaction step further provides for subjecting said component (F3) to be liquefied to a compression step, adapted to bring said component (F3) to a certain pressure, and for subjecting said component (F3) to a cooling process carried out by means of series of cooling steps until a liquefaction temperature of said component (F3) to be liquefied is reached.
- **15.** The method according to claim 12 or 13, wherein the liquefaction step provides for:
  - subjecting a methane-containing component (F3) to a first cooling step providing for bringing said component (F3) from the ambient temperature to a temperature lower than 0°C, preferably of about -30°C;
  - subjecting said component (F3) to a second cooling step providing for bringing said component (F3) from the temperature of about -30°C to a temperature of about -60°C;
  - subjecting said component (F3) to a third cooling step providing for bringing said component (F3) from the temperature of about -60°C to a temperature of about -80°C;
  - subjecting said component (F3) to a fourth cooling step providing for bringing said component (F3) from the temperature of about -80°C to a temperature of about -100°C;
  - subjecting said component (F3) to a fifth cooling step providing for bringing said component (F3) from the temperature of about -100°C to a temperature of about -130°C.







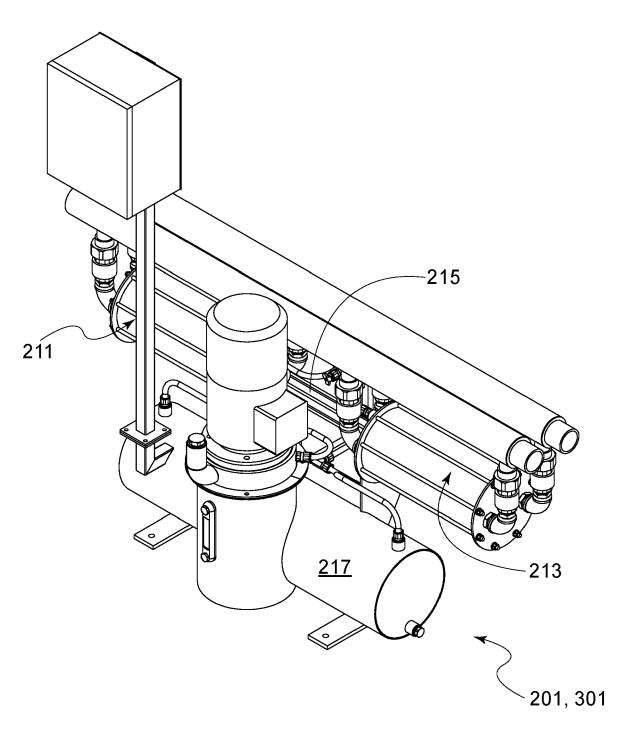


Fig. 3

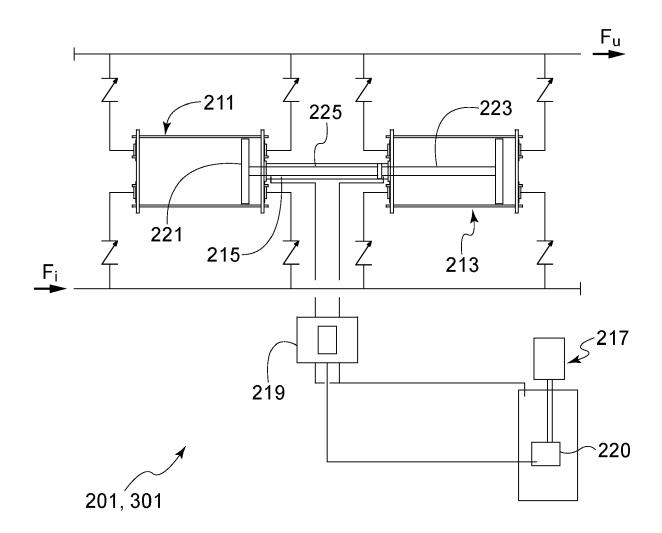
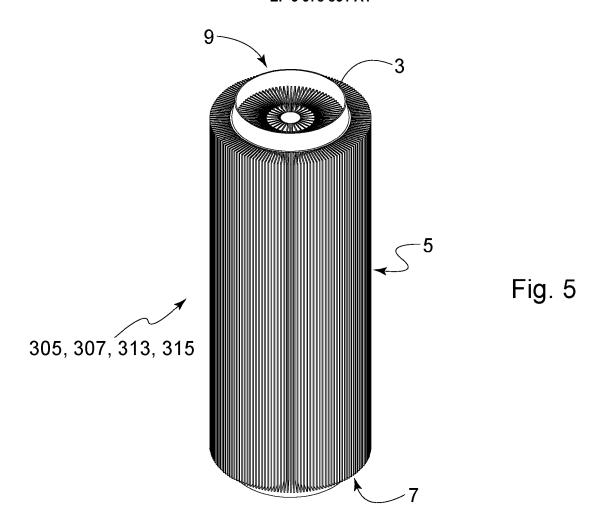
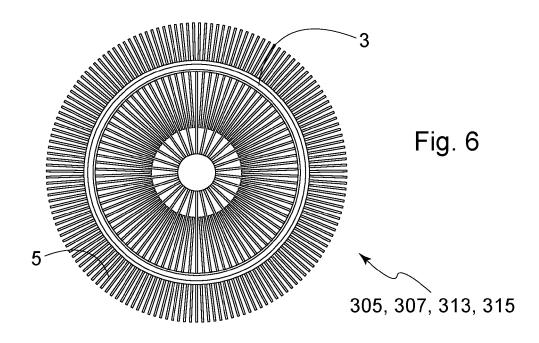


Fig. 4





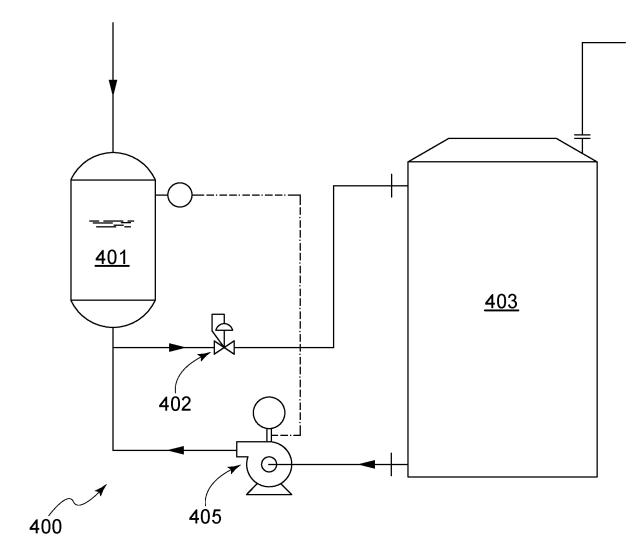


Fig. 7



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Application Number

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Place of search		te of completion of the search		Examiner
	Munich 1	8 February 2022	Sch	opfer, Georg
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D: document cited in the application
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