DEVICE AND METHOD FOR PRE-COOLING A STREAM OF A TARGET FLUID TO A TEMPERATURE LESS THAN OR EQUAL TO 90 K

Technical field of the invention

The present invention is directed to a device for cooling a flow of a target fluid to a temperature of less than or equal to 90 K and a method for cooling a flow of a target fluid to a temperature of less than or equal to 90 K. It applies, for example, to the field of hydrogen liquefaction.

State of the art

A fluid liquefaction method is schematically divided into three major temperature technology blocks: compression, pre-cooling and refrigeration. The purpose of pre-cooling, for example, is to lower inlet temperatures between 273 K and 320 K of a hydrogen flow of interest and the fluid used for refrigeration in the next block to a so-called pre-cooling temperature between 78 K and 120 K.

In known systems, the cooling step is historically performed using liquid nitrogen flowing in the opposite direction in a heat exchanger. This nitrogen enters at a temperature of about 78 K, exits at ambient temperature and is discharged into the atmosphere. Such systems are hereinafter referred to as “P1”.

In such systems, the implementation of an open loop of liquid nitrogen has the drawbacks of involving logistical management of its supply, storage of large amounts of nitrogen and low energy performance (approximately from 3.5 to 4.5 kWh/kg LH_2). Economic and practical advantages of these systems are justified in the context of small productions of less than 5 tonnes per day, but are not viable or operationally complex beyond this. Finally, these systems are not suitable for productions in remote and hard-to-reach areas due to the need to create a liquid nitrogen supply chain.

Other known systems focus on recycling nitrogen in a closed cycle (or circuit). Such systems are hereinafter referred to as “P2”.

This is achieved by a series of compression and cooling operations with a final expansion allowing temperature of the nitrogen to be lowered to about 80 K. Using heat exchangers, fluids to be cooled are also brought to about 80 K. Such systems are hereinafter referred to as “P2.1”.

An improvement of this loop performs several expansion operations during cooling to optimise cold supply within the exchangers. Such systems are hereinafter referred to as “P2.2”.

An improvement of P2.2 can be achieved by implementing a so-called dual closed nitrogen cycle as there are two simultaneous inlet pressures in the compressors. Such systems are hereinafter referred to as “P2.3”.

All such systems are alternatives to solution P1 in that they operate in a closed nitrogen cycle, avoiding all the problems recited above. The P2.3 solution is an improvement of the P2.2 and P2.1 solutions making it possible to optimise cold supply within the exchangers. Nonetheless, these solutions require high investment in equipment, especially compressors, due to their high nitrogen throughput (approximately 8 tonnes per day to produce one tonne of liquid hydrogen per day).

Other systems known as “MRC” (Mixed-Refrigerant Cycle) use a mixture of hydrocarbons and nitrogen as a refrigerant, the composition of which varies depending on the solutions. By the same

operating principle of compression, cooling and expansion, the refrigerant is cooled to about 90 to 130 K. Using heat exchangers, the fluids to be cooled are also brought to about 90 to 130 K. Such systems are hereinafter referred to as “P3”.

A variant to this solution produces cold in the 90–110K range using a blended refrigerant consisting of nitrogen, methane, ethane and propane. Such systems are hereinafter referred to as “P3.1”.

A variant to this solution applies the concept to hydrogen liquefaction with a three-stage expansion pre-cooling carried out through a Joule-Thomson valve (referred to as “J-T”). Such systems are hereinafter referred to as “P3.2”.

A variant provides a similar cycle, but turbines were used instead of J-T valves. On the other hand, the composition then comprised nitrogen, C_1-C_5 hydrocarbons, ethylene, tetrafluoromethane R14 and neon. Such systems are hereinafter referred to as “P3.3”.

One variant simplifies the single-stage expansion and five-component refrigerant (N2 and C1-C4) mixing cycle. Such systems are hereinafter referred to as “P3.4”.

Another variant also results in a simplified composition (H_2, N_2, C_1, C_2 and C_4) in a three-stage expansion system and adds a compression stage partly performed by a pump. Such systems are hereinafter referred to as “P3.5”.

Another variant uses a two-stage expansion cooling mixing system, where compression is partly made with a pump and the composition is limited to four components (N_2, C_1, C_2, iC_4). Such systems are hereinafter referred to as “P3.6”.

These P3 systems resort to a combined cooling cycle and optimise the cycle energy efficiency compared to previous systems by enabling heat exchanges between cold and hot fluids to be adapted by virtue of successive partial evaporation operations of the different compounds. Variations between these solutions are due to changes in compositions, number of expansion stages and addition of a compression stage, partly provided by a pump.

However, pre-cooling using a mixed refrigerant described above allows the target fluid to be cooled down to a temperature limited to approximately 90K without the risk of crystallisation. This limit requires the cooling cycle to perform cooling above 90 K or take the risk of crystallisation, increasing the need for the compressive power thereof compared to a cycle using pure nitrogen.

US patent application 2018,347,897 A is known, which discloses a device for pre-cooling a target gas by implementing two separate cooling fluid circuits. However, such a device does not allow pre-cooling of a gas to a temperature below 90 K.

US patent application 2019,063,824 is also known, which discloses a device for pre-cooling a target gas by implementing four separate circuits of natural gas joint cooling and liquefaction fluid. However, such a device implements a large number of target fluid and cooling fluid circuits. Furthermore, this device is complex and costly due to the joint use of some elements, such as expansion turbines, in cooling circuits.

Scientific publication *"A novel Hydro liquefaction process configuration with combined mixed refrigerant systems"* by Asadnia *et al* is also known, which discloses a device for pre-cooling a target

gas by implementing two separate cooling fluid circuits. However, such a device is complex due to some elements, such as expansion turbines, used in cooling circuits.

DISCLOSURE OF THE INVENTION

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

For this purpose, according to a first aspect, the present invention is directed to a device for pre-cooling a flow of a target gas at a temperature of less than or equal to 90 K, which comprises:

- a group of at least two heat exchangers between the flow of target gas, a flow of a first cooling fluid and at least one flow among a flow of a second cooling fluid and a flow of a third cooling fluid, the target gas downstream of the group of exchangers remaining in gas form,
- a closed circulation circuit for a flow of a second cooling fluid, said fluid comprising at least methane, said circuit comprising:
- at least one compression stage of the flow of the second fluid,
- at least one liquid-gas separation stage of the flow of the second fluid to form a liquid part and a gas part, at least one of the two parts being provided to at least one said heat exchanger; and
- at least one expansion stage of the flow of the second fluid and
- a circulation circuit for a flow of the third cooling fluid through at least one of said heat exchangers, said circuit not comprising an expansion turbine, said third fluid having a circulation temperature in said exchanger of less than 90K,

said circulation circuits of each cooling fluid being separate.

By virtue of these provisions, it is possible to achieve pre-cooling temperatures below 90K at low energy and economic cost while maintaining optimised exchanges enhanced between the refrigerant mixture and the fluids to be cooled without the risk of crystallisation.

The present invention is distinguished by a pre-cooling to overcome the compromise between energy efficiency and pre-cooling temperature found in existing solutions. Indeed, the method provided is both very energy efficient by virtue of the use of a mixture of refrigerants and both capable of reaching the coldest temperatures usually reached without risk of crystallisation by a pre-cooling circuit in the liquefaction of a target fluid, especially hydrogen, by virtue of the nitrogen loop.

The present invention thus makes it possible to significantly reduce energy consumption of the cooling circuit of the entire method.

Furthermore, the present invention makes it possible to avoid the use of an expansion turbine in the circulation circuit for a flow of the third cooling fluid. This simplifies the circuit, therefore reducing the cost of the device.

In embodiments, the circulation circuit for a flow of the third cooling fluid comprises at least one expansion stage for the flow of third fluid, the expansion stage comprising a Joule-Thompson valve, upstream of said at least one heat exchanger.

In embodiments, the circulation circuit for the flow of third fluid is configured so that the third cooling fluid has a mixture of liquid and gas downstream of the expansion stage.

In embodiments, the circulation circuit for the flow of third fluid is configured to pass through at least two of the heat exchangers of the heat exchanger group.

In embodiments, the circulation circuit for a flow of the third cooling fluid comprises the expansion stage for the flow of third fluid between a third fluid outlet of a heat exchanger among the two of said heat exchangers and a third fluid inlet of a heat exchanger among the two of said heat exchangers.

In embodiments, the circulation circuit for a flow of the third cooling fluid comprises at least one compression stage for the flow of the third fluid between a third fluid outlet of a heat exchanger among the two of said heat exchangers and a third fluid inlet of a heat exchanger among the two of said heat exchangers.

In embodiments, the circulation circuit for a flow is a closed circulation circuit for a flow of a third cooling fluid.

In embodiments, the circulation circuit for a flow is a closed circulation circuit for a flow of a third cooling fluid through at least two of said heat exchangers, comprising:

- at least one compression stage for the flow of third fluid between a third fluid outlet of a heat exchanger among the two of said heat exchangers and a third fluid inlet of a heat exchanger among the two of said heat exchangers and

- at least one expansion stage for the flow of third fluid between a third fluid outlet of a heat exchanger among the two of said heat exchangers and a third fluid inlet of a heat exchanger among the two of said heat exchangers.

These embodiments make it possible to form pre-cooling with two integrated loops, further optimising the operation of the device.

In embodiments:

- the target fluid successively passes through at least a first heat exchanger and a second heat exchanger,

- the closed circulation circuit for the flow of the third cooling fluid also passes through at least the first heat exchanger and the second heat exchanger and

- at least one compression stage of the closed circulation circuit for the flow of the third cooling fluid is positioned between a third fluid outlet of the first heat exchanger and an inlet for third compressed fluid of said first heat exchanger.

These embodiments make it possible to condition, in pressure, the third refrigerant fluid upstream of the injection of this fluid into the group of exchangers performing pre-cooling of the target fluid.

In embodiments:

- the target fluid successively passes through at least a first heat exchanger and a second heat exchanger,

- the closed circulation circuit for the flow of the third cooling fluid also passes through the first heat exchanger and the second heat exchanger and

- at least one expansion stage of the closed circulation circuit for the flow of third cooling fluid is positioned downstream of the second heat exchanger.

These embodiments make it possible to regenerate frigories in the third fluid, in particular upstream of an inverse passage through the heat exchanger group.

In embodiments, the circulation circuit for a flow of a third fluid is an open circulation circuit for a flow of a third cooling fluid through at least one heat exchanger.

These embodiments make it possible to reduce the number of exchanger channels and decrease complexity of the refrigeration cycle in the event of a third fluid open circuit.

In embodiments, the open circulation circuit for the flow of third fluid is configured to pass through at least two of the heat exchangers of the heat exchanger group.

These embodiments make it possible to reduce the number of exchanger channels and decrease complexity of the refrigeration cycle in the event of a third fluid open circuit.

In embodiments, the flow of third cooling fluid is a nitrogen flow.

In embodiments, the flow of third cooling fluid has a liquefaction temperature at atmospheric pressure less than or equal to the liquefaction temperature at atmospheric pressure of the second cooling fluid.

In embodiments, the circulation circuit of the nitrogen flow is configured so that the nitrogen flow is constrained by at least one of the following operating conditions:

- a high pressure of between 22 and 100 bara,
- a low pressure of between 1 and 2.2 bara,
- a mass ratio of nitrogen to target fluid of between 1 and 8 and/or
- an inlet temperature in at least one heat exchanger between 78 K and 88 K.

These operating conditions have the best pre-cooling efficiencies.

In embodiments:

- a target fluid circuit successively passes through a first heat exchanger and a second heat exchanger of the group,
- the closed circulation circuit for the flow of the second cooling fluid also passes through the first heat exchanger and the second heat exchanger and
- at least one expansion stage of the closed circulation circuit for the flow of the second cooling fluid is positioned downstream of a second fluid outlet of a heat exchanger.

These embodiments make it possible to restore part of the frigories of the second cooling fluid upstream of the complete or partial reverse passage through the heat exchanger group.

In embodiments:

- a target fluid circuit successively passes through a first heat exchanger and a second heat exchanger of the group,
- the closed circulation circuit for the flow of the second cooling fluid also passes through the first heat exchanger and the second heat exchanger of the group and
- at least one compression stage of the closed circulation circuit for the flow of the second cooling fluid is positioned between a second fluid outlet of the first heat exchanger and a second compressed fluid inlet of said first heat exchanger.

These embodiments make it possible to condition the second cooling fluid upstream of the pre-cooling step.

In embodiments:

- the target fluid successively passes through a first heat exchanger and a second heat exchanger,

- the closed circulation circuit for the flow of the second cooling fluid also passes through the first heat exchanger and the second heat exchanger and

- at least one liquid-gas separation stage of the flow of the second fluid is positioned upstream of the first heat exchanger, at least one of the liquid and gas parts being provided to said first heat exchanger.

Separations make it possible to constitute flows mainly comprised of light species vaporising at low temperature and flows mainly comprised of heavy species vaporising at medium or high temperature (cryogenic baseline).

The benefits are many:

- refine the boiling temperature steps to have an optimised partial vaporisation and

- reduce the risk of crystallisation of heavy compounds at a low temperature.

In embodiments:

- the heat exchanger group has an intermediate heat exchanger,

- a target fluid circuit successively passes through a first heat exchanger in the group, the intermediate heat exchanger and a second heat exchanger in the group,

- the closed circulation circuit for the flow of the second cooling fluid also passes through the first heat exchanger, the intermediate heat exchanger and the second heat exchanger and

- at least one liquid-gas separation stage for the flow of the second fluid is positioned downstream of the first heat exchanger and upstream of the intermediate heat exchanger, at least one of the liquid and gas parts being provided to said intermediate heat exchanger.

In embodiments, the device object of the present invention comprises, downstream of a compression stage for the second cooling fluid:

- a liquid-gas separation stage for the second cooling fluid to form a gas part and a liquid part,

- a compression stage for the gas part,

- a means for compressing the liquid part and

- a means for mixing the compressed gas part and the compressed liquid part.

In embodiments, the circulation circuit of the second refrigerant fluid is configured so that the flow of the second refrigerant fluid is constrained by at least one of the following operating conditions:

- a high pressure of between 20 and 36 bara,

- a low pressure of between 1 and 2 bara,

- a mass ratio of nitrogen to target fluid of between 17.5 and 28,

- an inlet temperature at the heat exchanger group of between 86 K and 100 K,

- an inlet temperature at an intermediate exchanger of the group of heat exchangers of between 166 K and 210 K and/or

- an inlet temperature at a second heat exchanger of the heat exchanger group of between 95 K and 132 K.

These operating conditions have the best pre-cooling efficiencies.

In embodiments, the flow of target fluid is a hydrogen and/or helium flow.

In embodiments, the flow of first refrigerant fluid is a flow comprising or at least or consisting of:

- dihydrogen,
- neon, helium or a mixture of neon and helium or
- a mixture of neon, helium and dihydrogen.

In embodiments, the flow of second refrigerant fluid is a flow comprising at least or consisting of a mixture from:

- a mixture of nitrogen, methane, ethylene or ethane, propane or propene and n-butane or i-butane or but-1-ene,
- a mixture of methane, ethylene or ethane, propane or propene and n-butane or i-butane or but-1-ene and
- a mixture of nitrogen, methane, ethylene or ethane, propane or propene, n-butane or i-butane or but-1-ene and n-pentane or i-pentane.

In embodiments, the flow of the second refrigerant fluid consists, in mole percent, of:

- 4% to 14% nitrogen,
- 26.4% to 40% methane,
- 14.9% to 36.4% ethylene,
- 21.5% to 35% propane and
- 14.8% to 25% butane.

According to a second aspect, the present invention is directed to a method for pre-cooling a flow of a target gas to a temperature of less than or equal to 90 K, which comprises:

- a step of passing, by the target gas flow, through a group of at least two heat exchangers for exchanging heat between the target fluid flow, a flow of a first cooling fluid and at least one flow among a flow of a second cooling fluid and a flow of a third cooling fluid,
- a step of circulating a flow of a third cooling fluid through at least one of said heat exchangers, said circulation step not comprising an expansion step carried out by an expansion turbine, said third fluid having a circulation temperature in said exchanger of less than 90K and
- a step of circulating a flow of a second cooling fluid in a closed circuit, said fluid comprising at least methane, said circulation step comprising:
- at least one step of compressing the flow of the second fluid,
- at least one liquid-gas separation step for the flow of the second fluid to form a liquid part and a gas part, at least one of the two parts being provided to at least one said heat exchanger and
- at least one step of expanding the flow of the second fluid.

The method object of the present invention has the same advantages as the device object of the present invention.

Brief description of the figures

Further particular advantages, purposes and characteristics of the invention will become clearer from the non-limiting description that follows of at least one particular embodiment of the device and method object of the present invention, with regard to the attached drawings, wherein:

Fig. 1 schematically represents a first particular embodiment of the device object of the present invention,

Fig. 2 schematically represents a second particular embodiment of the device object of the present invention,

Fig. 3 schematically represents a third particular embodiment of the device object of the present invention,

Fig. 4 schematically represents a fourth particular embodiment of the device object of the present invention,

Fig. 5 schematically represents in the form of a flowchart, a first succession of particular steps of the method object of the present invention,

Fig. 6 schematically represents in the form of a flowchart, a second succession of particular steps of the method object of the present invention,

Fig. 7 schematically represents in the form of a flowchart, a third particular sequence of steps of the method object of the present invention, and

Fig. 8 schematically represents in the form of a flowchart, a fourth sequence of particular steps of the method object of the present invention.

Description of the embodiments

This description is provided for non-limiting purposes, wherein each characteristic of an embodiment can be advantageously combined with any other characteristic of any other embodiment.

From now on, it is noted that the figures are not drawn to scale.

As used herein, “fluid” of a given compound denotes a fluid comprising at least said compound in major proportion. “Major proportion” refers to at least one relative major proportion.

In variants, the term “major proportion” means a proportion corresponding to at least 30%, 40%, 50%, 60%, 70%, 80%, 90%, 95% or 99% in volume of the flow.

A “target fluid” denotes a gas to be liquefied by the action of one of the variants of the device or method object of the present invention. Such a gas may, for example, correspond to dihydrogen. Such a target fluid is, for example, configured to initially have a temperature of 298 K. Such a target fluid is, for example, configured to initially have a pressure of 21 bara.

“First cooling fluid” denotes any gas or liquid likely to allow the device or method to cool the target flow to a temperature of 90 K or less. Such a first fluid comprises at least or consists of, for example:

- dihydrogen,
- neon, helium or a mixture of neon and helium,
- neon, dihydrogen or a mixture of neon and dihydrogen or
- a mixture of neon, helium and dihydrogen.

Such a flow of first fluid is, for example, configured to operate in a closed circuit between 298 K and 22 K as the circulation in the closed circuit proceeds.

“Second cooling fluid” denotes any gas or liquid likely to allow the device or method to cool the target flow to a temperature of 90 K or less and preferably 80 K or 83 K or less.

Preferably, the second refrigerant fluid circulating, for example, in a closed circulation circuit is in liquid or two-phase form, that is a liquid-gas mixture, in most of the circuit.

This second refrigerant fluid has several variants:

In a first variant, the second fluid consists of or comprises five compounds:

- nitrogen,
- methane,
- ethylene or ethane,
- propane or propene and
- an n-butane or a i-butane or a but-1-ene.

In a second variant, the second fluid consists of four compounds:

- methane,
- ethylene or ethane,
- propane or propene and
- an n-butane or a i-butane or a but-1-ene.

In a third variant, the second fluid consists of six compounds:

- nitrogen,
- methane,
- ethylene or ethane,
- propane or propene,
- an n-butane or a i-butane or a but-1-ene and
- an n-pentane or an i-pentane.

“Third cooling fluid” denotes any gas or liquid likely to allow the device or method to cool the target flow to a temperature of 90 K or less. In other words, the third cooling fluid has a circulation temperature in a heat exchanger of less than 90 K. Such a third fluid is, for example, nitrogen or argon.

In embodiments, the flow of third cooling fluid has a liquefaction temperature at a predetermined pressure, for example, at atmospheric pressure, less than or equal to the same predetermined pressure liquefaction temperature of the second cooling fluid.

In embodiments, the flow of third cooling fluid has a dew point less than or equal to the dew point of the second cooling fluid at a predetermined pressure, for example, at atmospheric pressure. Preferably, the flow of third cooling fluid also has a bubble point less than or equal to the bubble point of the second cooling fluid at a predetermined pressure, for example, at atmospheric pressure.

In the description below, “heat exchanger” denotes any heat exchanger likely to be suitable for the operating conditions allowing cooling of less than 90 K of the target fluid. For example, such a heat exchanger is a multiple-flow plate and fin heat exchanger.

It is noted that devices of the same type, for example compressors or exchangers, may not be separate devices, but stages of a single device for all or part of the devices of a given type. For example, the exchangers, 106, 107, 108 and 136, can correspond to three separate stages of a single exchanger.

A schematic view of one embodiment of the device 100 object of the present invention is observed in Fig. 1, which is not to scale. This device 100 for cooling a flow 101 of a target fluid to a temperature less than or equal to 90 K, comprises:

- a group 105 of at least two heat exchangers, 106, 107, 108 and/or 136, for exchanging heat between the target fluid flow, a flow 102 of a first cooling fluid, a flow of a second cooling fluid and/or a flow of a third cooling fluid,

- a closed circulation circuit 110 of a flow of a second cooling fluid, said fluid comprising at least methane, said circuit comprising:

- at least one stage, 111 and/or 112, of compressing the flow of the second fluid,

- at least one liquid-gas separation stage, 115 and/or 116, for the flow of the second fluid to form a liquid part and a gas part, at least one of the two parts being provided to at least one said heat exchanger and

- at least one stage, 120, 121 and/or 122, for expanding the flow of the second fluid and

- a circuit 125 of circulating a flow of the third cooling fluid through at least one, or even two, of said heat exchangers.

In embodiments, the third fluid circulating in the circulation circuit 125 has a circulation temperature in an exchanger of less than 90 K. In other words, in these embodiments, the third cooling fluid cools the target fluid to a temperature of less than or equal to 90 K in such a heat exchanger.

In embodiments, the circuit 125 of circulating a flow of the third cooling fluid does not comprise an expansion turbine ("turboexpander"). In particular, the amount of frigories produced in the closed circuit 110 of the second circulation refrigerant fluid is sufficient to avoid the use of a circuit 125 expansion turbine for circulating a flow of the third cooling fluid. In other words, the closed circulation circuit 110 of the second refrigerant fluid, comprising different stages, is therefore sufficiently energy efficient and thus makes it possible to simplify the circulation circuit 125 for a flow of the third cooling fluid. In addition, the presence of an expansion turbine limits the use of the third fluid to specific pressure and temperature ranges, as such a fluid should remain in gas form during the expansion in order not to damage the turbine. Preferably, such a limitation should be avoided in circuit 125 of the third fluid since the liquid part of the third fluid participates in intensifying heat exchanges in one or more exchangers. In addition, such a liquid part of the third fluid allows effective cooling of the target fluid to a temperature of 90 K or less.

In embodiments, the circulation circuit 125 for the flow of the third cooling fluid comprises an expansion stage. Such a circulation circuit 125 for the flow of the third cooling fluid may be closed or open. Preferably, such an expansion stage does not have an expansion turbine (turboexpander).

In embodiments, the expansion stage of the circuit 125 of circulating a flow of the third cooling fluid is a Joule-Thompson valve. Even more preferentially, the flow of the third cooling fluid is a two-phase fluid, that is the flow of the third cooling fluid comprises a mixture of liquid and gas, downstream of the expansion stage.

The term group 105 denotes at least two exchangers, 106, 107, 108 and/or 136, preferably heat exchangers belonging to a group for pre-cooling the target fluid 101.

The group 105 of exchangers is characterised by the fact that, in at least one exchanger, 106, 107, 108 and/or 136, the target fluid flow 101, the first cooling fluid 102 and the second cooling fluid interact therein. This group 105 of exchangers may also, in variants, be a place of exchange between the aforementioned fluids and a third cooling fluid.

In at least one exchanger, 106, 107, 108 and/or 136, of the group 105 of exchangers, the first cooling fluid 102 has a temperature lower than the temperature of the target fluid 101 passing through each said exchanger, 106, 107, 108 and/or 136.

Preferably, each exchanger, 106, 107, 108 and 136, of the group 105 of exchangers has both the target fluid 101 and the first cooling fluid 102 passing therethrough. The target fluid 101 and the first cooling fluid 102 may circulate in a co-current and/or counter-current manner with each other.

The cooling device 100 may further comprise a plurality of additional heat exchangers downstream of the heat exchanger group 105. These heat exchangers correspond to the ordinary implementation of the cooling stage of a liquefaction device for the target fluid 101.

Thus, as is understood, in embodiments such as those shown in Figs. 1 to 4, the target fluid 101 initially passes through the group 105 of heat exchangers in a pre-cooling stage and then a succession of at least one heat exchanger in a cooling stage.

At the outlet of these two stages, the liquefied target fluid 101 may be discharged or inserted into a gas-liquid separation stage, the liquid fraction of the target fluid 101 being discharged and the gaseous fraction of the target fluid 101 being recirculated in at least one of the exchangers of the pre-cooling or cooling stage.

Such a mechanism is represented in Figs. 1 to 4 without being referenced.

The first refrigerant fluid 102 may pass through all or part of the heat exchangers through which the target fluid 101 passes, whether in group 105 of exchangers or in at least one exchanger positioned upstream or downstream of said group 105 of exchangers.

Figs. 1 to 4 show a variant in which the first fluid 102 flows through all the exchangers through which the target fluid 101 pass.

Preferably, the device 100 comprises a closed circulation circuit for the first refrigerant fluid 102 in all or part of the heat exchangers of the device, in the group 105 of exchangers, upstream and/or downstream of said group 105 of exchangers. In these embodiments, represented in Figs. 1 to 4, the refrigerant fluid 102 passes through the exchangers in a first direction, co-currently from the target fluid 101, and then counter-currently from this target fluid 101 in a second direction.

This closed circuit may comprise intermediate stages of compression, expansion, division or mixing of the flow 102 of the first refrigerant fluid, as represented in Figs. 1 to 4.

The closed circulation circuit 110 of a flow of a second cooling fluid is intended to contribute to cooling, in at least one heat exchanger, 106, 107, 108 and/or 136, of the group 105 of exchangers, the first refrigerant fluid 102 and/or the target fluid 101.

The exact architecture of the circuit 110 depends on a compromise between theoretical performance and cycle complexity.

The number of exchangers is additionally related to the number of separations, as is the number of expansion operations related to the number of separation operations.

A circuit with a single separation (and therefore two exchangers and two expansions) is adapted if the composition of the refrigerant is adapted and the risks of crystallisation of the heaviest species at low temperatures are limited. Also, some variants do not implement separation. Such variants further restrict the composition and lowest achievable temperature.

Both variants degrade method performance by increasing energy consumption, but simplify the device variants set out in Figs. 1-4.

Conversely, with additional separation (i.e. four exchangers and four expansion operations) reduces energy consumption, but makes the cycle more complex.

This closed circuit 110 comprises:

- at least one stage, 111 and/or 112, of compressing the flow of the second fluid,

- at least one liquid-gas separation stage, 115 and/or 116, for the flow of the second fluid to form a liquid part and a gas part, at least one of the two parts being provided to at least one said heat exchanger and

- at least one stage, 120, 121 and/or 122, of expanding the flow of the second fluid.

At least one compression stage, 111 and/or 112, or a compression means 150 is, for example, a turbocompressor, a mechanical or reciprocating compressor.

At least one liquid-gas separation stage, 115 and/or 116 is, for example, a separation column.

At least one expansion stage, 120, 121 and/or 122 is, for example, a Joule-Tompson valve.

As is understood, there are many embodiments for this closed circuit 110.

In some embodiments, such as the one represented in Fig. 1, the closed circuit 110 for the second refrigerant fluid comprises a stage 111 of compressing the second refrigerant fluid at the outlet of a heat exchanger 106. This heat exchanger 106 is preferably the first exchanger in which the target fluid 101 in the group 105 of heat exchangers travels.

In some embodiments, such as that represented in Fig. 1, the closed circuit 110 for second refrigerant fluid comprises a stage 140 of separating the second refrigerant fluid at the outlet of the compression stage 111.

In some embodiments, such as the one represented in Fig. 1, the closed circuit 110 for the second refrigerant fluid comprises a gas-liquid separation stage 140 of the second refrigerant at the outlet of the compression stage 111.

In some embodiments, such as the one represented in Fig. 1, the closed circuit 110 for the second refrigerant comprises a compression stage 112 of a gas part derived from a gas-liquid separation stage 140.

In some embodiments, such as that represented in Fig. 1, the closed circuit 110 for the second refrigerant fluid comprises a means 150 of compressing a liquid part from a gas-liquid separation stage 140.

In some embodiments, such as that represented in Fig. 1, the closed circuit 110 for a second refrigerant fluid comprises a means 155 of mixing a compressed liquid part derived from a compression stage 112 and a compressed gas part derived from a compression means 150.

In some embodiments, such as the one represented in Fig. 1, the closed circuit 110 for a second refrigerant fluid comprises a gas-liquid separation stage 115 for the flow of a second refrigerant fluid from a mixing means 155 to form a gas part and a liquid part.

In some embodiments, such as the one represented in Fig. 1, the liquid part of the second refrigerant flow from the gas-liquid separation stage 115 is provided to a heat exchanger 106. This heat

exchanger 106 is preferably the first exchanger in which the target fluid 101 in the group 105 of heat exchangers travels.

In some embodiments, such as the one represented in Fig. 1, the liquid part of the second refrigerant flow from a heat exchanger 106 is provided to an expansion stage 120 and then injected back into the exchanger 106 before being provided to a compression stage 111.

In some embodiments, such as that represented in Fig. 1, the gas part of the second refrigerant flow from the gas-liquid separation stage 115 is provided to a heat exchanger 106. This heat exchanger 106 is preferably the first exchanger in which the target fluid 101 in the group 105 of heat exchangers travels.

In some embodiments, such as that represented in Fig. 1, the second refrigerant flow from a heat exchanger 106 is provided to a liquid-gas separation stage 116 to form a gas part and a liquid part. In some embodiments, such as that represented in Fig. 1, the liquid part of the second refrigerant flow from the gas-liquid separation stage 116 is provided to a heat exchanger 107. This heat exchanger 107 is preferably the second exchanger in which the target fluid 101 in the group 105 of heat exchangers travels.

In some embodiments, such as that represented in Fig. 1, the liquid part of the second refrigerant flow from a heat exchanger 107 is provided to an expansion stage 121 and then injected back into the heat exchanger 107, and then optionally into a heat exchanger 106, before being provided to a compression stage 111.

In some embodiments, such as that represented in Fig. 1, the gas part of the second refrigerant flow from the gas-liquid separation stage 116 is provided to a heat exchanger 107. This heat exchanger 107 is preferably the second exchanger in which the target fluid 101 in the group 105 of heat exchangers travels.

In some embodiments, such as that represented in Fig. 1, the second refrigerant flow from the heat exchanger 107 is provided to a third heat exchanger 108. This heat exchanger 108 is preferably the third exchanger in which the target fluid 101 in the group 105 of heat exchangers travels.

In some embodiments, such as that represented in Fig. 1, the flow of the second refrigerant from a heat exchanger 108 is provided to an expansion stage 122 and then injected back into the heat exchanger 108, then optionally into a heat exchanger 107 and/or then into a heat exchanger 106, before being provided to a compression stage 111.

As is understood, the following diagram can be implemented iteratively:

- liquid gas separation of the flow of a second refrigerant fluid from a previous heat exchanger or previous iteration,

- injection of the liquid part into a heat exchanger through the target fluid 101 passes, expansion of the liquid part at the outlet of the exchanger, reinjection into the exchanger and then transport, optionally to all or part of the previous heat exchangers,

- injection of the gas part into a heat exchanger through which the target fluid 101 passes and injection into a separation stage of the next iteration.

The last step, at the end of the iteration of the diagram, is characterised by the absence of separation upstream of injection in a heat exchanger.

In embodiments, such as those represented in Figs. 1 and 2, the device, 100 or 200, comprises a closed circuit 125 of circulating a flow of a third cooling fluid through at least two of the heat exchangers, 106, 107, 108 and/or 136, comprising:

- at least one stage 130 of compressing the flow of third fluid between a third fluid outlet of a heat exchanger and a third fluid inlet of a heat exchanger and

- at least one stage 135 of expansing the flow of third fluid between a third fluid outlet of a heat exchanger and a third fluid inlet of a heat exchanger.

As is understood, there are many ways to implement the closed circuit 125. Below, several particular embodiments are set forth.

In a first embodiment, represented in Fig. 1, a compression stage 130 is positioned at one end of the closed circuit 125, that is between a third fluid outlet and a third fluid inlet of a same heat exchanger 106.

In embodiments, such as those represented in Figs. 1 and 2:

- the target fluid 101 successively passes through at least one first heat exchanger 106 and a second heat exchanger 108, or preferably the target fluid 101 successively passes through at least one first heat exchanger 106, an intermediate heat exchanger 107 and a second heat exchanger 108,

- the closed circulation circuit 125 of the flow of the third cooling fluid also passes through at least the first heat exchanger 106 and the second heat exchanger 108, or preferably at least one first heat exchanger 106, an intermediate heat exchanger 107 and a second heat exchanger 108, and

- at least one stage 130 of compressing the closed circulation circuit for the flow of the third cooling fluid being positioned between a third fluid outlet of the first heat exchanger 106 and a third compressed fluid inlet of said first heat exchanger 106.

Here, “successively” denotes a direct or indirect sequence of the staps of passing through the heat exchangers, 106, 107, 108 and/or 136, by the third cooling fluid.

In these embodiments, at least one compression stage 130 is positioned at the junction between a co-current through-passage and a counter-current through-passage of the third refrigerant fluid.

In embodiments, such as those represented in Figs. 1 and 2:

- the target fluid 101 successively passes through at least one first heat exchanger 106 and a second heat exchanger 108, or preferably the target fluid 101 successively passes through at least one first heat exchanger 106, an intermediate heat exchanger 107 and a second heat exchanger 108,

- the closed circulation circuit 125 of the flow of the third cooling fluid also passes through the first heat exchanger and the second heat exchanger, or preferably at least one first heat exchanger 106, an intermediate heat exchanger 107 and a second heat exchanger 108, and

- at least one stage 135 of expanding the closed circulation circuit for the flow of the third cooling fluid being positioned downstream of the second heat exchanger 108.

In these embodiments, at least one expansion stage 135 is positioned at the junction between a co-current through-passage and a counter-current through-passage of the third refrigerant fluid.

In embodiments, such as that represented in Fig. 2, the device 200 comprises a dedicated heat exchanger 205 for exchanging heat between the flow of the third compressed fluid and at least part of the flow of the third fluid from the additional heat exchanger 136.

In these embodiments, the closed circuit 125 comprises a separator 202 positioned downstream of the compression stage 130 and upstream of the group 105 of heat exchangers, along the path in which the third refrigerant fluid travels. This separator 202 is configured to separate a predetermined or variable part, according to a command issued by an automaton for example. The part thus separated is provided to the dedicated heat exchanger 205 so as to cool the flow of the third decompressed refrigerant fluid from the expansion stage 135.

The third cooling fluid from the dedicated heat exchanger 205 is provided to a mixing means 201, in which said fluid and the part not provided to the dedicated heat exchanger 205 are mixed.

At the outlet of the mixing means 201, the third refrigerant fluid is provided to the expansion stage 135.

In embodiments, such as those represented in Figs. 3 and 4, the device, 300 or 400, comprises an open circuit 305 of circulating a flow of a third cooling fluid through at least the heat exchanger 136. In embodiments, such as that represented in Fig. 3, the open circulation circuit 305 for the flow of third fluid is configured to pass through at least one of the heat exchangers, 106, 107, 108, and/or 136, of the heat exchanger group 105.

In embodiments, preferably adapted to the embodiment represented in Fig. 1, the third cooling fluid is a nitrogen flow constrained by at least one of the following operating conditions:

- a high pressure of between 22 and 100 bara,
- a low pressure of between 1 and 2.2 bara,
- a mass ratio of nitrogen to target fluid of between 1 and 8 and/or
- an inlet temperature in at least one heat exchanger between 78 K and 88 K.

In embodiments, preferably adapted to the embodiment represented in Fig. 1, the second refrigerant fluid is constrained by at least one of the following operating conditions:

- a high pressure of between 20 and 36 bara,
- a low pressure of between 1 and 2 bara,
- a mass ratio of nitrogen to target fluid of between 17.5 and 28,
- an inlet temperature at the heat exchanger group of between 86 K and 100 K,
- an inlet temperature at an intermediate exchanger of the group of heat exchangers of between 166 K and 210 K and/or
- an inlet temperature at a second heat exchanger of the heat exchanger group of between 95 K and 132 K.

For example, in variants, the target fluid flow 101 is comprised of normal hydrogen (25% para-hydrogen and 75% orthohydrogen) at 21 bara, 298K (25°C) with a mass flow rate of 0,116 kg/s. The flow is first cooled to 90K (-183°C) by three heat exchangers, 106, 107, 108 and 136. The target fluid 101 then enters a first heat exchanger 136, for example catalytic exchanger, performing the first step of the ortho-para conversion. The target fluid flow 101 exits the pre-cooling part at 80K (-193°C) and 48% of para-hydrogen. In the downstream cooling section, the supply hydrogen reaches 26K (-247°C) and 98% para-hydrogen through five serial catalytic heat exchangers (not referenced). The target fluid flow 101 is mixed with evaporative gas (boil-off gas) from the last stage of liquefaction and enters the last catalytic heat exchanger to reach 22K (-251°C). At this stage, the supply hydrogen is at 22K (-251°C),

20 bara and 99% para-hydrogen. The final liquefaction step is performed with a Joule-Thompson valve which lowers pressure to 2 bara. The liquid part of the flow (98%) exits the liquefier and the remaining gas part is liquefied.

Performing the conversion of orthohydrogen to parahydrogen during liquefaction can be performed in several ways and therefore leads to variants.

The advantage of using a catalytic exchanger is to carry out a first stage of conversion in the pre-cooling circuit to avoid doing so in the cooling circuit.

However, there may not be a catalytic exchanger and a dedicated reactor can be used, or even a conversion may not be performed at this location.

The general idea is to perform a conversion step and dissipate conversion heat with the third refrigerant, especially with a catalytic exchanger.

In variants, the circuit 102 of first refrigerant fluid is a double pressure Claude loop and the refrigerant used is normal hydrogen. The refrigerant fluid is first compressed to 29 bara by a multi-stage compressor (not referenced). The fluid is cooled to 90 K (-183°C) in three heat exchangers, 106, 107, 108 and 136 by exchange against the flow of the third refrigerant fluid, then cooled to 80 K (-193°C) in the exchanger 136 by exchange against the flow of the third refrigerant fluid. The first refrigerant fluid then enters the cooling section and is cooled to 69 K (-204°C) in the first cooling heat exchanger (not referenced). The refrigerant is separated, 89% of the total flow rate is expanded to 18.5 bara and reaches 60K (-213°C). The first refrigerant fluid is then cooled to 51K (-222°C) in a heat exchanger (not referenced) and is again expanded with a two-stage expander to 4.5 bara to reach 31.5K (-241.5°C). From this point on, the first refrigerant fluid is used as a refrigerant in the first four cooling heat exchangers (not referenced). The remaining part (11%) is cooled to 26K through four heat exchangers (not referenced). This part is then expanded with a Joule-Thompson valve to 1.5 bara to reach 22K. The liquid refrigerant cools the target fluid 101 to 22K in two (unreferenced) two-phase heat exchangers and seven multi-flow heat exchangers, comprising the heat exchanger group 105 and exchanger 136. The two refrigerant flows at 4.5 and 1.5 bara exit the pre-cooling part at ambient temperature. The one at low pressure is compressed to 4.5 bara in a first compressor (not referenced). It is then mixed with the medium pressure flow before entering the second compression stage (not referenced).

The third refrigerant fluid, for example nitrogen, cools the target fluid 101 from 90 K (-183°C) to 80 K (-193°C). Nitrogen is first compressed from 1 bara to 40 bara by a multi-stage compressor 130. Nitrogen is then cooled to 90 K (-183°C) in three heat exchangers, 106, 107, 108 and 136. Nitrogen is then partially liquefied using a Joule-Thompson valve to reach 78 K (-195°C) and nitrogen operates in the heat exchanger 106 as the main refrigerant. The remaining cooling capacity of nitrogen is used in the pre-cooling heat exchangers 106, 107, 108 and 136.

In variants, the second refrigerant fluid comprises a mixture of five components, the mole percents of which, relative to the total amount of the mixture of the five components, are as follows: R728 (nitrogen), 4–14%, R50 (methane) 26.4-40%, R1150 (ethylene) 14.9-36.4%, R290 (propane) 21.5-35% and R600 (butane) 14.8-25%. These refrigerants have different boiling points ranging from 78K (-195°C) to 261K (-12°C) at atmospheric pressure, making the second refrigerant partially liquid during most of the process.

The second refrigerant fluid is first compressed from 1 bara to 11 bara by a compression stage 111. At 11 bara a liquid fraction appears (about 10%) after intermediate cooling to ambient temperature, the phases are separated and the gas part finishes its compression in a compressor 112 while the liquid part finishes it in a pump 150. The use of a pump reduces the compression power and thus the energy consumption of the facility. The compressed flows are then mixed and the phases are separated again. The liquid part (30%) is cooled to 182K (-91°C) in the first heat exchanger 106 and expanded with a Joule-Thompson valve at 1 bara. The gas part (80%) is cooled to 182K (-91°C) in the first heat exchanger 106 and the phases are separated once again. The liquid part (73%) is cooled to 115K (-158°C) in the intermediate heat exchanger 107 and expanded to 1 bara with a Joule-Thompson valve. The gas part (27%) is cooled to 90K (-183°C) in two heat exchangers, 107 and 108, before being expanded in a Joule-Thompson valve to 1 bara and providing its cold power in the heat exchanger 108. The two previous flows are mixed and provide cooling power to the heat exchanger 107. Finally, the remaining two flows are mixed, provide cooling power in the first heat exchanger 106 and are provided to the compression stage 111.

In Fig. 5, schematically, a succession of particular steps of the method 500 object of the present invention is observed. This method 500 of cooling a flow of a target fluid to a temperature less than or equal to 90 K, comprises:

- a step 505 of passing through, by the target fluid flow, a group of at least two heat exchangers between the target fluid flow, a flow of a first cooling fluid and a flow of a third cooling fluid,
- a step 506 of circulating a flow of a third cooling fluid through at least two of said heat exchangers and
- a step 510 of circulating a flow of a second cooling fluid in a closed circuit, said fluid comprising at least methane, said circulation step comprising:
- at least one step 515 of compressing the flow of the second fluid,
- at least one liquid-gas separation step 520 for the flow of the second fluid to form a liquid part and a gas part, at least one of the two parts being provided to at least one said heat exchanger and
- at least one step 525 of expanding the flow of the second fluid.

This method is described, mutatis mutandis, in different embodiments and variants, with respect to the device, 100, 200, 300, and/or 400, illustrated in Figs. 1 to 4.

In Fig. 6, schematically, a succession of particular steps of the sub-method 600 object of the present invention is observed. This sub-method 600 describes a particular embodiment of the interactions of the first refrigerant fluid with other components of a device, 100, 200, 300 or 400, object of the present invention.

This sub-method 600 comprises:

- a step 605 of compressing the first low-pressure refrigerant fluid,
- a step 610 of mixing the first compressed refrigerant fluid and the first medium pressure refrigerant fluid,
- a step 615 of compressing the mixture of the first refrigerant fluid,
- a step 620 of cooling the first compressed refrigerant fluid by heat exchange, in the group 105 of exchangers, with the second refrigerant fluid, to reach a temperature between 90 K and 120 K,

- a step 625 of cooling the first compressed refrigerant fluid by heat exchange, in a heat exchanger 136, with the third refrigerant fluid, to reach a temperature between 78 K and 90 K,

- a step 630 of cooling the first refrigerant fluid in a third heat exchanger,

- a step 635 of separating the flow of the first refrigerant fluid into a low pressure flow and a low pressure flow,

- and then, on the one hand:

- a step 640 of cooling the flow of the first low pressure refrigerant in a set of cooling exchangers, a step 645 of expanding the flow of the first low pressure refrigerant fluid,

- a step 650 of cooling hot fluids in at least one exchanger of the device by the first refrigerant fluid,

- a step 655 of circulating the first low-pressure refrigerant to the compression step 605,

- and on the other hand:

- a step 660 of expanding the flow of the first medium pressure refrigerant fluid from the separation step 635,

- a step 665 of cooling the first medium pressure refrigerant fluid expanded,

- a step 670 of expanding the flow of the first medium pressure refrigerant fluid from the cooling step 665,

- a step 675 of cooling hot fluids in at least one exchanger of the device by the first refrigerant and

- a step 680 of circulating the first refrigerant fluid at medium pressure to the mixing step 610.

A succession of particular steps of the sub-method 700 object of the present invention is schematically observed in Fig. 7. This sub-method 700 describes a particular embodiment of interactions of the third refrigerant fluid with other components of a device, 100, 200, 300, or 400, object of the present invention.

This sub-method 700 comprises:

- a step 705 of compressing the third refrigerant fluid,

- a step 710 of cooling the third refrigerant fluid in the group 105 of heat exchangers,

- a step 715 of expanding the third refrigerant fluid cooled,

- a step 720 of cooling the hot fluids, by the third refrigerant, in the group 105 of heat exchangers and in an exchanger 136 and

- a step 725 of circulating the third cooling fluid up to the compression step 705.

In a variant, step 720 is performed only in the exchanger 136.

A sequence of particular steps of the sub-method 800 object of the present invention is schematically observed in Fig. 8. This sub-method 800 describes a particular embodiment of the interactions of a second refrigerant fluid with other components of a device, 100, 200, 300 or 400, object of the present invention.

This sub-method 800 comprises:

- a step 805 of compressing the second refrigerant fluid,

- a step 810 of separating the second refrigerant fluid into a liquid phase and a gas phase,

- a step 815 of compressing the liquid phase of the second refrigerant fluid,

- a step 820 of compressing the gaseous phase of the second refrigerant fluid,
- a step 825 of mixing the gaseous and liquid phases compressed of the second refrigerant fluid,
- a step 830 of separating the second refrigerant fluid into a liquid part and a gas part,
- and then, on the one hand:
- a step 835 of cooling the liquid part, of the second cooling fluid, derived from the separation step 830,
- a step 840 of expanding the liquid part of the second refrigerant fluid cooled,
- and on the other hand:
- a step 860 of cooling the gas part, of the second refrigerant fluid, derived from the separation step 830,
- a step 865 of separating the second cooling fluid from the cooling step 860 into a liquid part and a solid part,
- and then, on the one hand:
- a step 870 of cooling the liquid part of the second refrigerant fluid and
- a step 871 of expanding the liquid part cooled,
- and on the other hand:
- a step 875 of cooling the gas part of the second refrigerant fluid,
- a step 876 of expanding the gas part cooled and
- a step 877 of cooling the hot fluids, by the second refrigerant fluid from the expansion step 876, in the exchangers 107 and 108,
- a step 880 of mixing the liquid and solid parts from the expansion 871 and cooling 877 steps and,
- a step 885 of cooling the hot fluids, by the second refrigerant fluid, in the intermediate exchanger 107 and
- a step 890 of circulating the second refrigerant fluid from the cooling step 885 to a mixing step 845,
- the step 845 of mixing the flows originating on the one hand from the expansion step 840 and circulation step 890,
- a step 850 of cooling the hot fluids, by the second refrigerant fluid, in the first exchanger 106 and
- a step 855 of circulating the second refrigerant fluid from the cooling step 850 to the compression step 805.

CLAIMS

1. A device (100, 200, 300, 400) for pre-cooling a flow (101) of a target gas to a temperature less than or equal to 90 K, characterised in that it comprises:

- a group (105) of at least two heat exchangers (106, 107, 108, 136) for exchanging heat between the target gas flow, a flow (102) of a first cooling fluid and at least one flow among a flow of a second cooling fluid and a flow of a third cooling fluid,

the target gas downstream of the group of exchangers remaining in gas form,

- a closed circulation circuit (110) for a flow of a second cooling fluid, said fluid comprising at least methane, said circuit comprising:

- at least one stage (111, 112) of compressing the flow of the second fluid,

- at least one stage (115, 116) of liquid-gas separation of the flow of the second fluid to form a liquid part and a gas part, at least one of the two parts being provided to at least one said heat exchanger and

- at least one stage (120, 121, 122) of expanding the flow of the second fluid and

- a circulation circuit (125, 305) for a flow of the third cooling fluid through at least one of said heat exchangers, said circuit not comprising an expansion turbine, said third fluid having a circulation temperature in said exchanger of less than 90K,

said circulation circuits of each cooling fluid being separate.

2. The device (100, 200) according to claim 1, wherein the circulation circuit (125) for a flow of the third cooling fluid comprises at least one stage (135) of expanding the flow of the third fluid, the expansion stage comprising a Joule-Thompson valve, upstream of the at least one heat exchanger.

3. The device (100, 200) according to claim 2, wherein the circulation circuit (125, 305) for the flow of the third fluid is configured so that the third cooling fluid comprises a mixture of liquid and gas downstream of the expansion stage (135).

4. The device (100, 200, 400) according to any of claims 1 to 3, wherein the circulation circuit (125) for the flow of the third fluid is configured to pass through at least two of the heat exchangers (106, 107, 108, 136) of the heat exchanger group (105).

5. The device (100, 200) according to any of claims 2 or 3 and according to claim 4, wherein the circulation circuit (125) for a flow of the third cooling fluid comprises the stage (135) of expanding the flow of the third fluid between a third fluid outlet of a heat exchanger (108) among the two of said heat exchangers and a third fluid inlet of a heat exchanger (136) among the two of said heat exchangers.

6. The device (100, 200) according to any of claims 4 or 5, wherein the circulation circuit (125) for a flow of the third cooling fluid comprises at least one stage (130) of compressing the flow of the third fluid

between a third fluid outlet of a heat exchanger (136) among the two of said heat exchangers and a third fluid inlet of a heat exchanger (106) among the two of said heat exchangers.

7. The device (200) according to claim 6, which comprises a dedicated heat exchanger (205) for exchanging heat between the flow of the third compressed fluid and at least one part of the flow of the third fluid from a heat exchanger (136).

8. The device (100, 200) according to any of claims 1 to 7, wherein the circulation circuit (125) for a flow is a closed circulation circuit (125) for a flow of a third cooling fluid.

9. The device (300, 400) according to any of claims 1 to 7, wherein the circulation circuit (305) for a flow of a third fluid is an open circulation circuit for a flow of a third cooling fluid through at least one heat exchanger (136).

10. The device (100, 200, 300, 400) according to any of claims 1 to 9, wherein the flow of third cooling fluid is a nitrogen flow.

11. The device (100, 200, 300, 400) according to claim 10, wherein the circulation circuit of the nitrogen flow is configured so that the nitrogen flow is constrained by at least one of the following operating conditions:

- a high pressure of between 22 and 100 bara,
- a low pressure of between 1 and 2.2 bara,
- a mass ratio of nitrogen to target fluid of between 1 and 8 and/or
- an inlet temperature at at least one heat exchanger of between 78 K and 88 K.

12. The device (100, 200, 300, 400) according to any of claims 1 to 11, wherein the flow of third cooling fluid has a liquefaction temperature at atmospheric pressure less than or equal to the liquefaction temperature at atmospheric pressure of the second cooling fluid.

13. The device (100, 200, 300, 400) according to any of claims 1 to 12, wherein:

- a target fluid circuit (101) successively passes through a first heat exchanger (106) and a second heat exchanger (108) of the group (105),
- the closed circulation circuit (110) for the flow of the second cooling fluid also passes through the first heat exchanger and the second heat exchanger and
- at least one liquid-gas separation stage (115) for the flow of the second fluid is positioned upstream of the first heat exchanger, at least one of the liquid and gas parts being provided to said first heat exchanger.

14. The device (100, 200, 300, 400) according to any of claims 1 to 13, wherein:

- the heat exchanger group (105) has an intermediate heat exchanger (107),

- a target fluid circuit successively passes through a first heat exchanger (106) of the group (105), the intermediate heat exchanger and a second heat exchanger (108) of the group (105),

- the closed circulation circuit (110) for the flow of the second cooling fluid also passes through the first heat exchanger, the intermediate heat exchanger and the second heat exchanger and

- at least one liquid-gas separation stage (116) for the flow of the second fluid is positioned downstream of the first heat exchanger and upstream of the intermediate heat exchanger, at least one of the liquid and gas parts being provided to said intermediate heat exchanger.

15. The device (100, 200, 300, 400) according to one of claims 1 to 14, which comprises, downstream of a stage (111) of compressing the second cooling fluid:

- a liquid-gas separation stage (140) for the second cooling fluid to form a gas part and a liquid part,

- a means (112) of compressing the gas part,

- a means (150) of compressing the liquid part and

- a means (155) of mixing the compressed gas part and the compressed liquid part.

16. The device (100, 200, 300, 400) according to any of claims 1 to 15, wherein the circulation circuit for the second refrigerant fluid is configured so that the flow of the second refrigerant fluid is constrained by at least one of the following operating conditions:

- a high pressure of between 20 and 36 bara,

- a low pressure of between 1 and 2 bara,

- a mass ratio of nitrogen to target fluid of between 17.5 and 28,

- an inlet temperature at the heat exchanger group of between 86 K and 100 K,

- an inlet temperature at an intermediate exchanger of the group of heat exchangers of between 166 K and 210 K and/or

- an inlet temperature at a second heat exchanger of the heat exchanger group of between 95 K and 132 K.

17. The device (100, 200, 300, 400) according to any of claims 1 to 16, wherein the flow (101) of target fluid is a hydrogen and/or helium flow.

18. The device (100, 200, 300, 400) according to any of claims 1 to 17, wherein the flow of first refrigerant fluid is a flow comprising or at least or consisting of:

- dihydrogen,

- neon, helium or a mixture of neon and helium or

- a mixture of neon, helium and dihydrogen.

19. The device (100, 200, 300, 400) according to any of claims 1 to 18, wherein the flow of the second refrigerant fluid is a flow comprising or consisting of a mixture from:

- a mixture of nitrogen, methane, ethylene or ethane, propane or propene and n-butane or i-butane or but-1-ene,

- a mixture of methane, ethylene or ethane, propane or propene and n-butane or i-butane or but-1-ene and

- a mixture of nitrogen, methane, ethylene or ethane, propane or propene, n-butane or i-butane or but-1-ene and n-pentane or i-pentane.

20. The device (100, 200, 300, 400) according to any of claims 1 to 19, wherein the flow of the second refrigerant fluid consists, in mole percent, of:

- 4% to 14% nitrogen,
- 26.4% to 40% methane,
- 14.9% to 36.4% ethylene,
- 21.5% to 35% propane and
- 14.8% to 25% butane.

21. A method (500) for pre-cooling a flow of a target gas to a temperature of less than or equal to 90 K, characterised in that it comprises:

- a step (505) of passing through, by the target gas flow, a group of at least two heat exchangers for exchanging heat between the target gas flow, a flow of a first cooling fluid and at least one flow among a flow of a second cooling fluid and a flow of a third cooling fluid,

- a step (506) of circulating a flow of a third cooling fluid through at least one of said heat exchangers, said circulation step not comprising an expansion step performed by an expansion turbine, said third fluid having a circulation temperature in said exchanger less than 90K and

- a step (510) of circulating a flow of a second cooling fluid in a closed circuit, said fluid comprising at least methane, said circulation step comprising:

- at least one step (515) of compressing the flow of the second fluid,

- at least one liquid-gas separation step (520) for the flow of the second fluid to form a liquid part and a gas part, at least one of the two parts being provided to at least one said heat exchanger and

- at least one step (525) of expanding the flow of the second fluid.