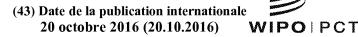
# (12) DEMANDE INTERNATIONALE PUBLIÉE EN VERTU DU TRAITÉ DE COOPÉRATION EN MATIÈRE DE BREVETS (PCT)

(19) Organisation Mondiale de la Propriété Intellectuelle

Bureau international





(10) Numéro de publication internationale WO 2016/166468 A1

(51) Classification internationale des brevets : F25J 3/06 (2006.01) F25J 1/02 (2006.01) F25J 1/00 (2006.01)

(21) Numéro de la demande internationale :

PCT/FR2016/050846

(22) Date de dépôt international :

13 avril 2016 (13.04.2016)

(25) Langue de dépôt :

français

(26) Langue de publication :

français

(30) Données relatives à la priorité : 1553430 17 avril 2015 (17.04.2015) FR

- (71) Déposant: L'AIR LIQUIDE, SOCIETE ANONYME POUR L'ETUDE ET L'EXPLOITATION DES PROCEDES GEORGES CLAUDE [FR/FR]; 75, Quai d'Orsay, 75007 Paris (FR).
- (72) Inventeurs: DEMOLLIENS, Bertrand; 79 Avenue Philippe Auguste, 75011 Paris (FR). BERNHARDT, Jean-Marc; 103 rue de l'Isère, 38500 La Buisse (FR). ALI SAID, Rawia; 8 bis rue Servan, 38000 Grenoble (FR). PICHOT, Delphine; 198 boulevard Voltaire, 75011 Paris

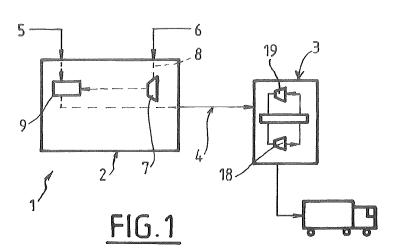
(FR). **HERNANDEZ, Antoine**; 40, avenue du Buisson Fleuri, 94510 La Queue En Brie (FR). **TSEVERY, Jean-Marc**; 134, rue des Grands Champs, 77127 Lieusaint (FR). **PEYRON, Jean-Marc**; 5, avenue de la Reine Blanche, 94000 Creteil (FR).

- (74) Mandataire : DE CUENCA, Emmanuel; L'air Liquide S.A., Direction de la Propriété Intellectuelle, 75, Quai d'Orsay, 75321 Paris Cedex 07 (FR).
  - Etats désignés (sauf indication contraire, pour tout titre de protection nationale disponible): AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BN, BR, BW, BY, BZ, CA, CH, CL, CN, CO, CR, CU, CZ, DE, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IR, IS, JP, KE, KG, KN, KP, KR, KZ, LA, LC, LK, LR, LS, LU, LY, MA, MD, ME, MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PA, PE, PG, PH, PL, PT, QA, RO, RS, RU, RW, SA, SC, SD, SE, SG, SK, SL, SM, ST, SV, SY, TH, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, ZA, ZM, ZW.
- (84) États désignés (sauf indication contraire, pour tout titre de protection régionale disponible): ARIPO (BW, GH, GM, KE, LR, LS, MW, MZ, NA, RW, SD, SL, ST, SZ,

[Suite sur la page suivante]

(54) Title: FACILITY AND METHOD FOR PRODUCING LIQUID HELIUM

(54) Titre : Installation et procédé de production d'hélium liquide



(57) Abstract: The invention relates to a facility for producing liquid helium from a source gas mixture (5) substantially comprising nitrogen and helium. The facility includes a cryogenic purifier (2) including a system (9) for separating the nitrogen from the source gas mixture with a view to producing helium at a temperature lower than the temperature of the source gas. The facility (1) also includes a helium liquefier (3) that subjects the helium to a work cycle including, in series: compressing the helium, cooling and decompressing the compressed helium, and reheating the cooled, decompressed helium. The facility includes a helium transfer pipe (4) connecting an outlet of the purifier (2) to an inlet of the liquefier in order to transfer helium produced by the purifier (2) into the work cycle of the liquefier (3). The facility is characterized in that the cryogenic purifier (2) includes a decompression system (8) that includes an inlet to be connected to a source (6) of pressurized nitrogen gas. Said system (8) for decompressing (7) the nitrogen gas exchanges heat with the separation system (9) in order to transfer cold from the decompressed nitrogen gas to said separation system (9).

(57) Abrégé:



# 

TZ, UG, ZM, ZW), eurasien (AM, AZ, BY, KG, KZ, RU, Déclarations en vertu de la règle 4.17 : TJ, TM), européen (AL, AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV, MC, MK, MT, NL, NO, PL, PT, RO, RS, SE, SI, SK, SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, KM, ML, MR, NE, SN, TD, TG).

relative à la qualité d'inventeur (règle 4.17.iv))

### Publiée :

- avec rapport de recherche internationale (Art. 21(3))
- avec revendications modifiées (art. 19.1))

Installation de production d'hélium liquide à partir d'un mélange de gaz source (5) comprenant essentiellement de l'azote et de l'hélium, l'installation comprenant un épurateur (2) cryogénique comprenant un circuit (9) de séparation de l'azote du mélange de gaz source en vue de produire de hélium à une température inférieure à la température du gaz source, l'installation (1) comprenant en outre un liquéfacteur (3) d'hélium soumettant l'hélium à un cycle de travail comprenant en série : une compression de l'hélium, un refroidissement et une détente de l'hélium comprimé et un réchauffement de l'hélium refroidi et détendu, l'installation comprenant une conduite (4) de transfert d'hélium reliant une sortie de l'épurateur (2) à une entrée du liquéfacteur pour transférer de hélium produit par l'épurateur (2) dans le cycle de travail du liquéfacteur (3), l'installation étant caractérisée en ce que l'épurateur (2) cryogénique comprend un circuit (8) de détente comprenant une entrée destinée à être raccordée à une source (6) d'azote gazeux sous pression, ledit circuit (8) de détente (7) de l'azote gazeux étant en échange thermique avec le circuit (9) de séparation pour transférer des frigories de l'azote gazeux détendu vers ledit circuit (9) de séparation.

### FACILITY AND METHOD FOR PRODUCING LIQUID HELIUM

The present invention relates to a facility and a process for producing helium.

The invention relates to helium purification and liquefaction.

5

10

15

20

25

30

The invention relates more particularly to a facility for producing liquid helium from a source gas mixture essentially comprising nitrogen and helium, the facility comprising a cryogenic purifier comprising a circuit for separating nitrogen from the source gas mixture with a view to producing helium at a temperature below the temperature of the source gas, the facility additionally comprising a helium liquefier that subjects the helium to a work cycle comprising in series: a compression of the helium, a cooling and an expansion of the compressed helium and a reheating of the cooled and expanded helium, the facility comprising a helium transfer line that connects an outlet of the purifier to an inlet of the liquefier in order to transfer helium produced by the purifier to the work cycle of the liquefier.

The invention relates in particular to the production of liquid helium in facilities that generate a mixture of helium and nitrogen and optionally other residues.

This gas source, substantially formed of nitrogen and helium in equal parts, may in particular be available in a natural gas production plant.

In this type of facility, nitrogen, which has been separated from the natural gas upstream, is generally available.

Liquid nitrogen may be used in helium liquefaction units. This makes it possible to reduce the size of the helium work cycle since, in this case, the helium from the liquefaction cycle may be cooled only between 80 K and 4 K approximately (rather than from ambient temperature to 4 K). Nevertheless, this solution requires adding an additional exchanger to the facility and a vessel for vaporizing the liquid nitrogen in a vacuum box in order to recover the cold from the liquid nitrogen.

The vacuum cold box of the liquefier also typically comprises adsorbers in order to strip the helium of the traces of atmospheric gas in order to prevent these traces from freezing in the downstream part of the process. These traces of atmospheric gas may decide the dimensions of the vacuum box.

One objective of the present invention is to overcome all or some of the drawbacks of the prior art raised above.

To this end, the facility according to the invention, furthermore in accordance with the generic definition given in the preamble above, is essentially characterized in that the cryogenic purifier comprises an expansion circuit comprising an inlet intended to be connected to a source of pressurized gaseous nitrogen, said circuit for expanding the gaseous nitrogen being in heat exchange with the separation circuit in order to transfer frigories from the expanded gaseous nitrogen to said separation circuit.

5

10

15

20

25

30

Furthermore, embodiments of the invention may comprise one or more of the following features:

- the separation circuit of the purifier comprises at least one heat exchanger in heat exchange with the source gas mixture with a view to the cooling thereof and at least one separator vessel, the circuit (8) for expanding the pressurized gaseous nitrogen is in heat exchange with the at least one heat exchanger of the separation circuit,
- the circuit (8) for expanding the pressurized gaseous nitrogen comprises at least two turbines for expanding the gaseous nitrogen and two separate portions in heat exchange with the at least one heat exchanger of the separation circuit, the two separate portions being located respectively downstream of the two expansion turbines,
- the separation circuit comprises at least one adsorption-type purification device for separating the nitrogen from the mixture,
- the helium liquefier comprises a compression station intended to carry out the compression of the helium in the work cycle and a cold box intended to carry out a cooling and an expansion of the helium compressed in the work cycle, the device for cooling the cycle helium originating from the compression station being incorporated in the cryogenic purifier in a thermally insulated common housing, the cold box of the liquefier is located in a thermally insulated separate housing that comprises vacuum insulation,
- at least one part of the compression station is incorporated in the cryogenic purifier (3) in a thermally insulated common housing that is separate from the housing incorporating the cold box of the liquefier,

- the cold box of the helium liquefier contains four turbines for expanding helium gas in the work cycle and the compression station contains a compressor stage of the work gas in the work cycle.

The invention also relates to a process for producing liquid helium from a source gas mixture essentially comprising nitrogen and helium using a facility in accordance with any one of the features above or below, wherein the source gas mixture comprising nitrogen and helium in molar concentrations respectively between 50% and 65% (for example between 55% and 60%, in particular 57%) and 35% and 50% (for example between 40% and 45%, in particular 42%), the source gas mixture optionally residually comprising at least one of the elements below: argon, oxygen, neon in proportions for example between 0.15% and 0.5%, in particular 0.22%, this source gas mixture having a pressure between 15 and 35 bar and a temperature between 273 and 323 K, and for example 300 K.

According to other possible distinctive features:

5

10

15

20

25

30

- the gaseous nitrogen inlet of the purifier is supplied with pressurized gaseous nitrogen at a pressure between 15 and 50 bar, for example 40 bar and a temperature between 273 and 323 K,
  - the helium produced by the purifier at its outlet has a pressure between 15 and 35 bar and a temperature for example between 77 and 90 K and for example 80 to 85 K, in particular 82 K,
  - the helium liquefier is configured to only cool the helium in the work cycle from the value of the temperature at the outlet of the purifier to the temperature of 4 K.

The invention may also relate to any alternative device or process comprising any combination of the features above or below.

Other distinctive features and advantages will become apparent on reading the description below, given with reference to the figures in which:

- figure 1 represents a schematic and partial figure illustrating the structure and the operation of the facility according to the invention,
- figures 2 and 3 illustrate, schematically and partially, the structure and operation of two examples of possible implementation of the invention.

The facility 1 for producing liquid helium represented schematically in figure 1 comprises a cryogenic purifier 2 (cryogenic upgrader). This purifier 2 is supplied with a source gas mixture 5 (helium and nitrogen) in order to produce, after

purification (cryogenic separation), pure or virtually pure helium, that is to say helium capable of supplying a helium liquefier 3.

For example, the nitrogen and helium are present in this source gas mixture in molar concentrations respectively between 50% and 65% (for example between 55% and 60%, in particular 57%) and 35% and 50% (for example between 40% and 45%, in particular 42%). The source gas mixture optionally residually comprises at least one of the elements below (argon, oxygen, neon) in proportions for example between 0.15% and 0.5% (in particular 0.22%). This source gas mixture may have a pressure between 15 and 35 bar and a temperature between 273 and 323 K and for example 300 K.

5

10

15

20

25

30

The purifier 2 conventionally comprises a circuit 9 for separating nitrogen from the source gas mixture with a view to producing helium at a temperature below the temperature of the source gas. The separation circuit 9 conventionally comprises steps of cooling (in particular by heat exchange with a cooling exchanger 10) and one or more passes through a separator vessel 11, 12, and an expansion (valve 20). Furthermore, the mixture may undergo one or more steps of purification via adsorption (via one or more devices 14, 15 of "PSA" pressure swing adsorption type in particular) in order to strip the mixture of its nitrogen.

As seen in figure 2, the separation circuit 9 of the purifier 2 may comprise at least one heat exchanger 10 in heat exchange with the source gas mixture with a view to the cooling thereof and two separator vessels 11, 12. The nitrogen recovered, in particular the liquefied nitrogen 21 obtained may be recovered in a recovery tank (not represented in the figures).

The circuit 8 for expanding the pressurized gaseous nitrogen may be in heat exchange with the at least one heat exchanger 10 of the separation circuit 9.

The purifier may in particular have no distillation column.

The facility 1 additionally comprises a helium liquefier 3 that conventionally subjects helium to a work cycle comprising in series: a compression of the helium (in a compression station), a cooling and an expansion of the compressed helium (in a cold box) and a reheating of the cooled and expanded helium with a view to returning it to the compression station to restart a cycle.

The facility 1 comprises a helium transfer line 4 that connects an outlet of the purifier 2 to an inlet of the liquefier 3. This transfer line 4 is provided to transfer helium produced by the purifier 2 to the work cycle of the liquefier 3. According to one advantageous distinctive feature, the cryogenic purifier 2 comprises a gaseous nitrogen inlet intended to be connected to a source 6 of pressurized gaseous nitrogen available in the facility.

As illustrated in figure 2, the purifier 2 comprises to this effect a circuit 8 for expanding 7 the pressurized gaseous nitrogen. This expansion circuit 8 is in heat exchange with the separation circuit 9 to enable the transfer of frigories from the expanded gaseous nitrogen to said separation circuit 9. That is to say that energy from the gaseous nitrogen is transferred in the process for purifying and cooling the source mixture.

5

10

15

20

25

30

More specifically, the circuit 8 for expanding 7 the pressurized gaseous nitrogen may be in heat exchange with the heat exchanger 10 of the separation circuit 9, in order to supply frigories used in the cryogenic separation of the nitrogen from the source mixture.

The circuit 8 for expanding 7 the pressurized gaseous nitrogen may comprise one or preferably at least two turbines 13 for expanding the gaseous nitrogen and two separate portions in heat exchange with the heat exchanger 10 of the separation circuit 9. The two separate portions in heat exchange with the exchanger 10 are located for example respectively downstream of the two turbines 13 for expanding the nitrogen.

This pressurized gaseous nitrogen is for example available at a pressure between 15 and 50 bar (for example 40 bar) and a temperature between 273 and 323 K.

The helium produced by the purifier 3 at its outlet has a pressure for example between 15 and 35 bar and a temperature for example between 77 and 90 K and for example 80 to 85 K. (82 K typically).

According to this configuration, the helium produced by the purifier 2 is returned cold directly to the work cycle of the liquefier 3. This makes it possible to reduce the cooling capacity of the liquefier 3 since it only needs to cool the helium between 80 K (temperature of the helium provided by the purifier 2) and 4 K (the target low liquefaction temperature).

According to the known processes, this helium had to be cooled from ambient temperature (300 K approximately) down to 4 K.

The invention makes it possible to reduce the size and capacity of the liquefier 3 of the facility 1.

Thus, the liquefier 3 may operate in "refrigerator" mode in the part of the cycle between 300 K and 80 K (that is to say that in this part of the work cycle there is as much helium that is cooled/expanded at the outlet of the compression station as there is helium that is reheated and returns to the compression station). On the other hand, between 80 K and 4 K the liquefier may operate in "liquefier" mode (that is to say that there is more helium which is in the expansion/cooling phase than in the phase of reheating and returning to the compression station).

5

10

15

20

25

30

This "refrigerator" operating mode in the part of the cycle between 300 K and 80 K is much more energy-efficient than the "liquefier" operating mode since the fluid flow rates are balanced in the work cycle (in both directions).

Specifically, this solution makes it possible to "transfer" refrigerating capacity from 300 K to 80 K from the compression station of the liquefier 3 to the nitrogen compressor of the purifier 2.

The compression of nitrogen (in particular by centrifugal compressor(s)) is much more energy-efficient than the compression of helium (in particular by oil-injected screw compressor(s)). Furthermore, the efficiency of the motor of a nitrogen compressor (which is much more powerful) will be better than that of a screw compressor. Specifically, the efficiency of a compressor motor increases with its size.

The energy efficiency of the facility 1 will therefore be improved by this change.

Obtaining cold (80 K) helium at the outlet of the purifier 2 also makes it possible to eliminate the two hot expansion turbines in the liquefier 3. These two turbines may be replaced by two nitrogen turbines on the purifier 2 side.

These two turbines 13 for nitrogen (typically oil-bearing turbines) are more efficient and less complex to produce than gas-bearing turbines for helium in the liquefier 3.

By eliminating two first turbines 18 in the liquefier 3, it is possible to considerably reduce the rate of return to medium pressure in the helium work cycle of the liquefier 3.

Another optimization of the liquefier 3 may make it possible to eliminate the return of helium to intermediate pressure in the work cycle of the liquefier 3. This may enable the liquefier 3 to operate with a single cycle compressor that will work

for example between 1 bar and 15 bar. This cycle compressor 19 may also consist of only a single oil-injected screw.

These improvements therefore make it possible to considerably reduce the cycle compressor requirements in the liquefier 3. The pressure of the cycle is thus also divorced from the feed pressure. This makes it possible to have an additional freedom parameter in order to optimize the overall plant incorporating this facility.

5

10

15

20

25

30

The adsorbers 15 of the purifier 2 (for example at a temperature of 80 K) may be incorporated into a thermally insulated cold box (conventionally insulated with perlite, the casing will preferably be insulated with rock wool in practice in order to retain the option of intervening for maintenance). This makes it possible to reduce the size of the vacuum cold box.

The regeneration of these adsorbers may be carried out with gas at the outlet of the PSA(s) 14 at ambient temperature. The (re)cooling of the purification cylinder containing the adsorber after the regeneration could be carried out by helium at the outlet (or at the inlet) of said in-line cylinder.

A portion of liquid nitrogen 21 produced may be drawn off from the facility 1. This liquid nitrogen may be consumed for other requirements in the plant (trucks, etc.).

Figure 3 represents a variant embodiment which differs from that of figure 2 only in that the initial cooling of the cycle helium originating from the compression station 16 of the liquefier 3 is incorporated into the cryogenic purifier 2 in a thermally insulated common housing whilst the cold box 17 of the liquefier 3 is located in a thermally insulated separate housing that comprises vacuum insulation.

That is to say that all the fluids having a temperature above 80 K are incorporated into one or more perlite-filled (insulated) cold boxes whereas the fluids having a temperature below 80 K are incorporated into a vacuum insulated cold box. This also makes it possible to reduce the size of the vacuum insulated cold box of the facility.

The cold box containing all the pieces of equipment may be insulated with perlite whereas the cold box containing the cryogenic adsorbers may be insulated with rock wool.

According to one distinctive feature, it is possible to share the pieces of equipment that enable the regeneration of the cold adsorbers between the liquefier

and the purifier. It is possible to recool the adsorber after regeneration with the gas at the inlet and not only with the gas at the outlet.

## **CLAIMS**

- 1. A facility for producing liquid helium from a source gas mixture (5) essentially comprising nitrogen and helium, the facility comprising a cryogenic purifier (2) comprising a circuit (9) for separating nitrogen from the source gas mixture with a view to producing helium at a temperature below the temperature of the source gas, the facility (1) additionally comprising a helium liquefier (3) that subjects the helium to a work cycle comprising in series: a compression of the helium, a cooling and an expansion of the compressed helium and a reheating of the cooled and expanded helium, the facility comprising a helium transfer line (4) that connects an outlet of the purifier (2) to an inlet of the liquefier in order to transfer helium produced by the purifier (2) to the work cycle of the liquefier (3), the cryogenic purifier (2) comprising an expansion circuit (8) comprising an inlet intended to be connected to a source (6) of pressurized gaseous nitrogen, said circuit (8) for expanding (7) the gaseous nitrogen being in heat exchange with the separation circuit (9) in order to transfer frigories from the expanded gaseous nitrogen to said separation circuit (9), wherein the helium produced by the purifier (3) at its outlet has a pressure between 15 and 35 bar and a temperature for example between 77 and 90 K and for example 80 to 85 K, in particular 82 K.
- 2. The facility as claimed in claim 1, characterized in that the separation circuit (9) of the purifier (2) comprises at least one heat exchanger (10) in heat exchange with the source gas mixture with a view to the cooling thereof and at least one separator vessel (11, 12) and in that the circuit (8) for expanding (7) the pressurized gaseous nitrogen is in heat exchange with the at least one heat exchanger (10) of the separation circuit (9).
- 3. The facility as claimed in claim 1 or 2, characterized in that the circuit (8) for expanding (7) the pressurized gaseous nitrogen comprises at least two turbines (13) for expanding the gaseous nitrogen and two separate portions in heat exchange with the at least one heat exchanger (10) of the separation circuit (9), the two separate portions being located respectively downstream of the two expansion turbines (13).

20

5

10

15

25

30

- 4. The facility as claimed in any one of claims 1 to 3, characterized in that the separation circuit (9) comprises at least one adsorption-type purification device (14, 15) for separating the nitrogen from the mixture.
- 5. The facility as claimed in any one of claims 1 to 4, wherein the helium liquefier (3) comprises a compression station (16) intended to carry out the compression of the helium in the work cycle, a device for cooling the cycle helium originating from the compression station and a cold box (17) intended to carry out a cooling and an expansion of the helium compressed in the work cycle, characterized in that the device for cooling the cycle helium originating from the compression station (16) is incorporated in the cryogenic purifier (3) in a thermally insulated common housing and in that the cold box (17) of the liquefier (3) is located in a thermally insulated separate housing that comprises vacuum insulation.
- 6. The facility as claimed in claim 5, wherein the cold box (17) of the helium liquefier (3) contains four turbines (18) for expanding helium gas in the work cycle and the compression station contains a compressor stage (19) of the work gas in the work cycle.
- 7. A process for producing liquid helium from a source gas mixture (5) essentially comprising nitrogen and helium using a facility (1) in accordance with any one of claims 1 to 6, characterized in that the source gas mixture (5) comprising nitrogen and helium in molar concentrations respectively between 50% and 65% and 35% and 50%, the source gas mixture optionally residually comprising at least one of the elements below: argon, oxygen, neon in proportions for example between 0.15% and 0.5%, in particular 0.22%, this source gas mixture having a pressure between 15 and 35 bar and a temperature between 273 and 323 K, and for example 300 K, and in that the helium produced by the purifier (3) at its outlet has a pressure between 15 and 35 bar and a temperature for example between 77 and 90 K and for example 80 to 85 K, in particular 82 K.
- 8. The process as claimed in claim 7, characterized in that the source gas mixture (5) comprises nitrogen and helium in molar concentrations respectively between 55% and 60%, in particular 57%, and 40% and 45%, in particular 42%.

15

5

10

20

25

30

9. The process as claimed in claim 6 or 7, characterized in that the gaseous nitrogen inlet of the purifier (2) is supplied with pressurized gaseous nitrogen at a pressure between 15 and 50 bar, for example 40 bar and a temperature between 273 and 323 K.

5

10. The process as claimed in any one of claims 7 to 9, characterized in that the helium liquefier (3) is configured to only cool the helium in the work cycle from the value of the temperature at the outlet of the purifier (2) to the temperature of 4 K.

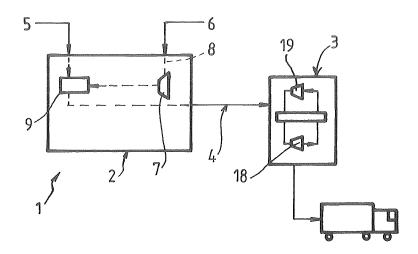


FIG.1

