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(54) **Title:** PROCESS AND APPARATUS FOR SEPARATION OF HYDROCARBONS AND NITROGEN

(57) **Abstract:** The invention provides a process and apparatus for the separation of nitrogen from a gaseous feed comprising a mixture of hydrocarbons and nitrogen gas, the process comprising the steps of: (i) cooling and at least partially condensing the gaseous feed; (ii) feeding the cooled and at least partially condensed feed from step (i) to a first fractionation to produce an overhead vapour stream having an enriched nitrogen content and a condensed product having a reduced nitrogen content which is subjected to a second fractionation, which comprises reboil, at a lower pressure than the first fractionation; (iii) partially condensing the overhead vapour stream, and separating to provide a liquid stream, which is used to provide reflux to the first fractionation, and a separated vapour stream, which is condensed to provide reflux to the second fractionation; and (iv) sub-cooling the condensed product of the first fractionation and dividing the resulting sub-cooled product into at least two streams: a first stream being expanded and fed to the second fractionation, and a second stream being expanded and reheated in heat exchange with the separated vapour stream from step (ii) before being fed to the second fractionation; (v) removing a hydrocarbon product stream low in nitrogen from the second fractionation; and (vi) removing a nitrogen rich stream from the second fractionation.

Process and Apparatus for Separation of Hydrocarbons and Nitrogen

This invention relates to processes and apparatus for the low temperature separation of nitrogen from a gaseous mixture comprising nitrogen gas and hydrocarbons. Such mixtures occur naturally in geological formations and can also result from nitrogen injection as a method of improving oil or gas production. Nitrogen separation may be required as part of an overall processing of gaseous hydrocarbons to meet sales specifications, such as maximum inert content or minimum calorific value.

10 Low temperature fractionation presents an energy efficient method for the separation of nitrogen from gaseous hydrocarbon streams, in particular gaseous hydrocarbon streams wherein the hydrocarbons comprise predominantly methane, such as natural gas. Separated nitrogen streams of high purity can be produced, thereby maximising hydrocarbon recovery and, where the nitrogen stream is vented to atmosphere, 15 minimising environmental impact.

Where the nitrogen content is higher than approximately 35 to 40 mol%, a double column arrangement (such as disclosed in US 7,127,915) similar to that used in air separation is conventional and is often the most economical choice considering both capital cost and 20 energy consumption. The columns are typically configured in a stacked arrangement, with the upper fractionation column operating at low pressure, just above atmospheric, and the lower fractionation column operating at high pressure, typically at approximately 27 bar (2700 kPa).

25 An example of a conventional double column arrangement is shown in Figure 1.

A feed gas (01) is cooled and at least partially condensed in heat exchanger (02). The partially condensed feed (03) is expanded across valve (06) to form a two-phase feed (07) which is fed to a high pressure column (08). The high pressure column (08) separates the two-phase feed (07) into a nitrogen rich overhead vapour fraction (19) and a hydrocarbon rich liquid fraction (09). 30

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The hydrocarbon rich liquid fraction (09) from the high pressure column (08) is sub-cooled in heat exchanger (10) to form stream (12) which is expanded across valve (13) to form a further two-phase feed stream (14) which is fed to an intermediate stage of a low pressure column (15).

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The overhead vapour (19) from the high pressure column (08) is fully condensed in heat exchange with boiling liquid at the bottom of the low pressure column (15) in reboil heat exchanger (04). The liquid may either be piped from the bottom tray or packed section, or the reboil heat exchanger (04) may be submerged in the liquid in the sump of the low
10 pressure column (15).

The fully condensed overhead stream (20) is split. A portion is passed as reflux (22) to the high pressure column (08) and a portion (23) is sub-cooled in heat exchangers (10) and (24) to form stream (25) which is expanded across valve (26) and passes as reflux
15 (27) to the low pressure column (15).

A hydrocarbon product (31) with low nitrogen content, from the low pressure column (15), is pumped to an elevated pressure, dependent on the composition and pressure of feed gas (01), and is rewarmed and evaporated in heat exchangers (10) and (02) to form a
20 gaseous product (35), providing the majority of the refrigeration for cooling and condensation of the feed gas (01).

A nitrogen vapour stream (28) with low methane content from the low pressure column (15) is rewarmed in heat exchanger (24) to provide refrigeration for sub-cooling of the
25 nitrogen rich reflux stream and is further rewarmed in heat exchangers (10) and (02).

The purity of the hydrocarbon product (31) from the bottom of the low pressure fractionation column (15) is ensured by provision of sufficient reboil in heat exchange with the overhead vapour from the high pressure column (08) in the reboil heat
30 exchanger (04). Purity of the nitrogen product from the low pressure fractionation column overhead (28) is ensured by conditioning the feed streams to the column and, in particular, providing sufficient flow of nitrogen rich reflux (27).

The pressure drop between high pressure fractionation column (08) and low pressure fractionation column (15) provides the required refrigeration by Joule-Thomson expansion. High purity nitrogen and hydrocarbon streams are withdrawn from the top 5 and bottom of the low pressure fractionation column (15) respectively.

When the nitrogen content of the feed gas to a conventional double column system is lower than approximately 35 mol%, insufficient reflux is normally generated to the low pressure fractionation column to maintain low losses of hydrocarbons with the nitrogen 10 product.

In such cases, options to increase the available reflux include:

- a) compressing the reject nitrogen stream and recycling it to the feed gas or other part 15 of the process to meet reflux requirements; and
- b) introducing an upstream 'pre-separation' system to condition the feed gas to produce a stream suitably enriched in nitrogen to feed to the double column system.

20 Process selection will be dependent on considerations such as plant capacity, feed gas nitrogen content and variability and feed and product gas pressure. Use of an upstream 'pre-separation' system is efficient in producing product gas at elevated pressure, hence reducing product gas compression power requirements and may also be preferred when the gaseous feed comprises contaminants such as carbon dioxide and heavy 25 hydrocarbons, which are tolerated at higher levels in the pre-separation system. Inclusion of either a pre-separation system or a nitrogen recycle system does however introduce additional equipment items.

The present invention provides an alternative double column system for the separation of 30 nitrogen from a gaseous mixture comprising nitrogen gas and hydrocarbons which uses improved heat integration to maximise the nitrogen reflux stream and to optimise the feed conditions to the low pressure fractionation column. This provides additional

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flexibility to process gases having low nitrogen content, for example less than 35 mol%, and potentially as low as 20 mol% without recourse to the use of additional equipment for pre-separation or nitrogen recycling as described above. However, the invention is also useful for the separation of gases having nitrogen content above 35 mol%, with improved 5 separation efficiency being observed in comparison with conventional separation techniques.

The present invention provides a process for the separation of nitrogen from a gaseous feed comprising a mixture of hydrocarbons and nitrogen gas, the process comprising the 10 steps of:

- (i) cooling and at least partially condensing the gaseous feed;
- (ii) feeding the cooled and at least partially condensed feed from step (i) to a first fractionation to produce an overhead vapour stream having an enriched nitrogen content and a condensed product having a reduced nitrogen content which is subjected to a second fractionation, which 15 comprises reboil, at a lower pressure than the first fractionation;
- (iii) partially condensing the overhead vapour stream, and separating to provide a liquid stream, which is used to provide reflux to the first fractionation, and a separated vapour stream, which is condensed to provide reflux to the second fractionation; and 20
- (iv) sub-cooling the condensed product of the first fractionation and dividing the resulting sub-cooled product into at least two streams: a first stream being expanded and fed to the second fractionation, and a second stream being expanded and reheated in heat exchange with the separated vapour stream from step (iii) before being fed to the 25 second fractionation;
- (v) removing a hydrocarbon product stream low in nitrogen from the second fractionation; and
- (vi) removing a nitrogen rich stream from the second fractionation. 30

The combination of steps (iii) and (iv) enables separation of feed streams with lower

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nitrogen content than is feasible with prior art processes, such as the conventional double column arrangement described above.

In particular, partial condensation of the overhead vapour stream from the first 5 fractionation allows the separated vapour stream from step (iii) to be efficiently condensed in heat exchange with reheat of the second stream of the condensed product in step (iv) to provide reflux to the second fractionation. The reheated second stream has an increased vapour fraction and its introduction into the second fractionation provides an increased quantity of stripping vapour to the second fractionation. For a 10 given feed composition, the effect of these steps is to provide improved separation, even for feed compositions comprising less than 35 mol% nitrogen, for example 20 to 35 mol%, while maintaining the overall energy efficiency of the separation process.

As a further advantage, partial condensation of the overhead stream from the first 15 fractionation allows the first fractionation and the second fractionation steps to be 'decoupled' as there is no physical requirement for the second fractionator to be elevated above the first fractionator.

In a preferred embodiment, the hydrocarbons in the gaseous feed comprise or consist of 20 methane. For instance, the gaseous feed may comprise or consist of natural gas. In further preferred embodiments, the gaseous feed comprises less than 40 mol% nitrogen, less than 35 mol% nitrogen, or less than 30 mol% nitrogen. Preferably, the gaseous feed comprises at least 20 mol% nitrogen. The gaseous feed may further comprise other inert gases, such as helium. If required, the gaseous feed may be subjected to 25 one or more pre-treatment procedures to remove impurities and/or unwanted components which could solidify in either of the fractionations.

The gaseous feed is cooled and partially condensed prior to the first fractionation. In order to minimise power consumption, heat exchange during cooling of the gaseous feed 30 may be used to provide reboil to the second fractionation. In addition, cooling of the gaseous feed may be obtained by heat exchange with the hydrocarbon product low in nitrogen from the second fractionation and/or the nitrogen rich stream from the second

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fractionation. In one preferred embodiment, the hydrocarbon product low in nitrogen from the second fractionation is pumped to elevated pressure and evaporated to provide cooling to the gaseous feed.

- 5 The gaseous feed may also be split into at least two streams, each of which may be independently processed according to any of the steps described above before being directed to the first fractionation. Where at least one of the at least two streams is used to provide reboil to the second fractionation energy efficiently via heat exchange, that stream is preferably fed to an intermediate stage of the first fractionation. Where at least
- 10 one of the at least two streams is not used to provide reboil to the second fractionation, that stream is preferably fed below the bottom stage of the first fractionation to provide stripping vapour. Preferably the first fractionation is at a pressure of 5 to 30 bar (0.5 to 3.0 MPa).
- 15 In addition, reboil in the second fractionation may be provided by heat exchange with the overhead vapour stream from the first fractionation during the partial condensation thereof, thereby reducing energy consumption.

In step (iv) of the process described above, the first stream may comprise between 10%
20 and 50% of the sub cooled product.

In a preferred embodiment, the first stream is expanded to form a two-phase feed, prior to being fed to the second fractionation. The second stream is preferably expanded to form a two-phase feed before being reheated via heat exchange with the separated
25 vapour stream from step (iii). If required, further reheating of the second stream, after expansion thereof, may be effected by heat exchange with the condensed product from the first fractionation and/or the overhead vapour stream from the first fractionation. Preferably the first stream is fed to the second fractionation at a higher stage than the second stream.

30

The hydrocarbon product stream low in nitrogen from the second fractionation is preferably removed from the second fractionation as a liquid stream.

In a further embodiment, the nitrogen rich stream from the second fractionation may be reheated by heat exchange with the overhead vapour stream from the first fractionation during partial condensation thereof and/or by heat exchange with the separated vapour stream in step (iii) during condensation thereof.

It will be appreciated by the skilled person that the residual nitrogen content of the hydrocarbon product and the residual hydrocarbon content of the nitrogen rich stream obtained from the second fractionation are dependent on the composition of the feed gas. However, the process of the present invention typically provides a hydrocarbon product comprising 2 mol% or less residual nitrogen content, and possibly a hydrocarbon product comprising less than 1 mol% residual nitrogen can be obtained. However, in other embodiments the process may be operated with a more relaxed specification so as to obtain a hydrocarbon product having, for example, up to 10 mol% residual nitrogen content.

The present invention also provides an apparatus for the separation of nitrogen from a gaseous feed comprising a mixture of hydrocarbons, the apparatus comprising of:

- 20 (i) means for cooling and at least partially condensing the gaseous feed;
- (ii) a first fractionator for producing an overhead vapour stream and a condensed product and a second fractionator operable at a lower pressure than the first fractionator;
- (iii) means for conveying the cooled and at least partially condensed feed from
25 step (i) to the first fractionator;
- (iv) means for conveying the condensed product from the first fractionator to the second fractionator;
- (v) means for partially condensing the overhead vapour stream, and means for separating the partially condensed vapour stream to provide a liquid
30 stream, and a vapour stream;

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- (vi) means for conveying the liquid stream to the first fractionator which is used to provide reflux to the first fractionation, and means for conveying and condensing the vapour stream to provide reflux to the second fractionator;
- 5 (vii) means for dividing the condensed product of the first fractionator, prior to entry into the second fractionator, into at least two streams;
- (viii) means for expanding a first stream prior to entry into the second fractionator, and means for expanding and heating prior to entry into the second fractionator;
- 10 (ix) means for conveying a hydrocarbon product low in nitrogen from the second fractionator; and
- (x) means for conveying a nitrogen rich stream from the second fractionator.

In one embodiment, the first and second fractionators may be in a stacked configuration, with the second fractionator positioned above the first fractionator.

15

In an alternate embodiment, the overall height of the apparatus may be reduced by arranging the first and second fractionators in a non-stacked configuration.

Preferably, the second fractionator comprises a reboil heat exchanger.

20

Suitable means for expanding the streams include liquid and two-phase expansion turbines.

The invention will now be described in greater detail with reference to preferred 25 embodiments and with the aid of the accompanying figures, in which:

Figure 1 shows a conventional stacked double column apparatus for the separation of nitrogen from a gaseous mixture comprising nitrogen gas and hydrocarbons, as described above.

30

Figure 2 shows a stacked double column apparatus in accordance with the present invention.

Figure 3 shows an uncoupled double column apparatus also in accordance with the present invention

5 Figure 4 also shows an uncoupled double column apparatus in accordance with the present invention.

In the embodiment of the invention shown in Figure 2, a high pressure column (08) and a low pressure column (15) are provided in a stacked arrangement, with the high pressure
10 column (08) positioned below the low pressure column (15).

A feed gas (01) is cooled and at least partially condensed in a heat exchanger (02) and is expanded across valve (06) to form two-phase feed (07) to the bottom of the high pressure column (08). The high pressure column (08) separates the two-phase feed
15 (07) into a nitrogen rich overhead vapour fraction (19) and a hydrocarbon rich liquid fraction (09)

The hydrocarbon rich liquid fraction (09) from the high pressure column (08) is sub-cooled in heat exchanger (10) and the resulting stream (12) is split into two portions. One
20 portion is expanded across valve (13) to form a two-phase feed stream (14) which is fed to an intermediate stage of the low pressure column (15). The other portion is expanded across valve (16) and is reheated at low pressure to form a two-phase feed stream (18), which has a higher vapour fraction and is fed to a lower stage of the low pressure column (15) than feed stream (14).

25

The overhead vapour (19) from the high pressure column (08) is partially condensed in heat exchange with boiling liquid at the bottom of the low pressure column (15) in a reboil heat exchanger (04). The boiling liquid may either be piped to the reboil heat exchanger (04) from a bottom tray or packed section of the low pressure column (15), or the reboil
30 heat exchanger (04) may be submerged in the liquid in the sump of the low pressure column (15).

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The partially condensed overhead stream (20) is separated into a liquid stream (22) and a separated vapour stream (23) in a phase separator (21). The liquid stream (22) is passed as reflux to the high pressure column (08). The separated vapour stream (23) is fully condensed and sub-cooled in heat exchangers (10) and (24) to form stream (25) 5 which is expanded across valve (26) and is passed as reflux (27) to the low pressure column (15).

A hydrocarbon product (31) with low nitrogen content from the low pressure column (15) is pumped to an elevated pressure by a pump (32), dependent on the composition and 10 pressure of the feed gas (01), and the resulting stream (33) is evaporated and reheated in heat exchangers (10) and (02) to form a gaseous product (35). Evaporation and reheating of the hydrocarbon stream (33) in heat exchanger (02) preferably provides at least a portion of, and more preferably the majority of, the refrigeration required for cooling and condensation of the feed gas (01).

15

A nitrogen vapour stream (28) with low hydrocarbon content from the low pressure column (15) is preferably reheated in heat exchanger (24) to provide further refrigeration for sub-cooling of the separated vapour stream (23) and is preferably further reheated in heat exchangers (10) and (02).

20

In the embodiment of the invention shown in Figure 3, a high pressure column (08) and a low pressure column (15) are provided in an uncoupled arrangement.

A feed gas (01) is cooled and at least partially condensed in a heat exchanger (02) and is 25 then sub-cooled in heat exchange with boiling liquid at the bottom of the low pressure column (15) in a reboil heat exchanger (04). The boiling liquid may either be piped to the reboil heat exchanger (04) from a bottom tray or packed section of the low pressure column (15), or the reboil heat exchanger (04) may be submerged in the liquid in the sump of the low pressure column (15). The cooled and at least partially condensed feed 30 gas (05) is expanded across valve (06) to form two-phase feed (07) to the bottom of the high pressure column (08).

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The hydrocarbon rich liquid fraction (09) from the high pressure column (08) is sub-cooled in heat exchangers (10) and (11) and the resulting stream (12) is split into two portions. One portion is expanded across a valve (13) to form a two-phase feed stream (14) to an intermediate stage of the low pressure column (15). The other portion is
5 expanded across a valve (16) and is reheated at low pressure to form a two-phase feed stream (18), which has a higher vapour fraction and is fed to a lower stage of the low pressure column (15) than feed stream (14).

The overhead vapour (19) from the high pressure column (08) is partially condensed in
10 heat exchanger (10). The partially condensed overhead stream (20) is separated into a liquid stream (22) and a separated vapour stream (23) in a phase separator (21). The liquid stream (22) is passed as reflux to the high pressure column (08). The separated vapour stream (23) is fully condensed and sub-cooled in heat exchangers (11) and (24) to form stream (25) which is expanded across a valve (26) and is passed as reflux (27) to
15 the low pressure column (15).

A hydrocarbon product (31) with low nitrogen content, from the low pressure column (15), is pumped to an elevated pressure by a pump (32), dependent on the composition and pressure of the feed gas (01), and the resulting stream (33) is evaporated and reheated
20 in heat exchanger (02) to form a gaseous product (34). Evaporation and reheating of the hydrocarbon stream (33) in the heat exchanger (02) preferably provides at least a portion of, and more preferably the majority of, the refrigeration required for cooling and condensation of the feed gas (01).

25 A nitrogen vapour stream (28) with low hydrocarbon content from the low pressure column (15) is preferably reheated in heat exchanger (24) to provide further refrigeration for sub-cooling of the separated vapour stream (23) and is preferably further reheated in heat exchangers (10) and (02).

30 In the embodiment of the invention shown in Figure 4, a high pressure column (08) and a low pressure column (15) are provided in an uncoupled arrangement, and multiple feeds are provided to the high pressure column.

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A feed gas (01) is cooled and at least partially condensed in a heat exchanger (02) and is split with a portion being expanded across valve (36) to form a two-phase feed (37) which is fed to the bottom of high pressure column (08). The remaining portion of the feed gas
5 (01) is further cooled in a heat exchanger (35) to form a stream (03) which is then sub-cooled in heat exchange with boiling liquid at the bottom of the low pressure column (15) in a reboil heat exchanger (04). The boiling liquid may either be piped to reboil heat exchanger (04) from a bottom tray or packed section of the low pressure column (15), or the reboil heat exchanger (04) may be submerged in the liquid in the sump of the low
10 pressure column (15). The cooled and at least partially condensed feed gas (05) is expanded across valve (06) to form feed stream (07) which is fed to an intermediate stage of high pressure column (08).

The hydrocarbon rich liquid fraction (09) from the high pressure column (08) is sub-
15 cooled in heat exchangers (10) and (11) and the resulting stream (12) is split into two portions. One portion is expanded across valve (13) to form a two-phase feed stream (14) which is fed to an intermediate stage of a low pressure column (15). The other portion is expanded across valve (16) and is reheated at low pressure to form a two-phase feed stream (18) which has a higher vapour fraction and is fed to a lower stage of
20 the low pressure column (15) than feed stream (14).

The overhead vapour (19) from the high pressure column is partially condensed in a heat exchanger (10). The partially condensed overhead stream (20) is separated into a liquid stream (22) and a separated vapour portion (23) in a phase separator (21). The liquid
25 stream (22) passes as reflux to the high pressure column (08). The separated vapour stream (23) is fully condensed and subcooled in heat exchangers (11) and (24) to form a stream (25) which is expanded across valve (26) and is passed as reflux (27) to the low pressure column (15).

30 A hydrocarbon product (31) with low nitrogen content, from the low pressure column (15), is pumped to an elevated pressure by pump (32), dependent on the composition and pressure of feed gas (01), and the resulting stream (33) is evaporated and reheated in

heat exchangers (35) and (02) to form a gaseous product (34). Evaporation and reheating of the hydrocarbon stream (33) in the heat exchangers (35) and (02) preferably provides at least a portion of, and more preferably the majority of, the refrigeration required for cooling and condensation of the feed gas (01).

5

A nitrogen vapour stream (28) with low hydrocarbon content from the low pressure column (15) is preferably reheated in heat exchanger (24) to provide further refrigeration for sub-cooling of the separated vapour stream (23) and is preferably further reheated in heat exchangers (11), (10), (35) and (02).

10

Examples

Comparative Example 1

15 Table 1 shows typical operating parameters for the conventional double column apparatus shown in Figure 1 when used to separate a gaseous mixture consisting of 40 mol% nitrogen gas and 60 mol% methane. It will be observed that, based on 6 theoretical separation stages in the high pressure column (08) and 6 theoretical separation stages in the low pressure column (15), the conventional double column
20 apparatus is able to separate such a mixture to obtain a nitrogen product stream (Stream 30) having a residual methane content of 0.8 mol% when producing a methane product stream (Stream 35) having a residual nitrogen content of 2.0 mol%.

Table 1

Stream¹		1	7	9	14	19	22	23
Pressure ²	MPa	3.00	2.70	2.70	0.22	2.68	2.68	2.68
Temperature	°C	35.0	-127.6	-127.7	-171.2	-146.4	-149.6	-149.6
Mass Flow	kg/h	104155	104155	84014	84014	53792	33651	20140
Molar Flow	mol/h	5000.0	5000.0	4255.5	4255.5	1988.2	1243.8	744.4
Nitrogen	mol %	40.0	40.0	30.9	30.9	92.0	92.0	92.0
Methane	mol%	60.0	60.0	69.1	69.1	8.0	8.0	8.0

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Stream ¹		27	28	30	31	35
Pressure ²	MPa	0.22	0.20	0.15	0.22	0.98
Temperature	°C	-187.7	-188.3	28.5	-155.9	28.5
Mass Flow	kg/h	20140	54562	54562	49592	49592
Molar Flow	mol/h	744.4	1954.4	1954.4	3045.6	3045.6
Nitrogen	mol%	92.0	99.2	99.2	2.0	2.0
Methane	mol%	8.0	0.8	0.8	98.0	98.0

¹As identified in Figure 1²Pressures are given as absolute values*Comparative Example 2*

5

Table 2 shows typical operating parameters for the conventional double column apparatus shown in Figure 1 when used to separate a gaseous mixture consisting of 30 mol% nitrogen gas and 70 mol% methane. It will be observed that, based on the same number of theoretical separation stages in columns (08) and (15) as per Comparative Example 1, and producing a methane product stream (Stream 35) having a residual nitrogen content of 2.0 mol%, hydrocarbon recovery in the conventional double column apparatus is reduced, with the residual methane content of the nitrogen product stream (Stream 30) increasing to 11.1mol%.

15 Table 2

Stream ¹		1	7	9	14	19	22	23
Pressure ²	MPa	3.00	2.70	2.70	0.22	2.68	2.68	2.68
Temperature	°C	35.0	-122.5	-122.6	-167.7	-147.1	-149.9	-149.9
Mass Flow	kg/h	98170	98170	85562	85562	45074	32466	12608
Molar Flow	mol/h	5000.0	5000.0	4536.0	1536.0	1658.6	1194.6	464.0
Nitrogen	mol%	30.0	30.0	23.6	69.6	93.0	93.0	93.0
Methane	mol%	70.0	70.0	76.4	30.4	7.0	7.0	7.0

Stream ¹		27	28	30	31	35
Pressure ²	MPa	0.22	0.20	0.15	0.22	0.12
Temperature	°C	-187.7	-176.4	29.3	-155.9	29.3
Mass Flow	kg/h	12608	42981	42981	55188	55188
Molar Flow	mol/h	464.0	1610.5	1610.5	3389.5	3389.5
Nitrogen	mol%	93.0	88.9	88.9	2.0	2.0
Methane	mol%	7.0	11.1	11.1	98.0	98.0

¹As identified in Figure 1

²Pressures are given as absolute values

5 Example 3

Table 3 shows typical operating parameters for the process of the invention using the apparatus shown in Figure 2, when used to separate a gaseous mixture consisting of 40 mol% nitrogen gas and 60 mol% methane. With this gaseous mixture, the first stream 10 accounts for 33% by molar flow of the sub cooled product. It will be observed that, based on the same number of theoretical separation stages in columns (08) and (15) as per Comparative Example 1, the process of the invention is able to separate such a mixture to obtain a nitrogen product stream (Stream 30) having an improved residual methane content of 0.4 mol% when producing a methane product stream (Stream 35) having a 15 residual nitrogen content of 2.0 mol%.

Table 3

Stream ¹		1	7	9	14	18	19	22
Pressure ²	MPa	3.00	2.50	2.50	0.22	0.22	2.48	2.48
Temperature	°C	35.0	-129.8	-129.9	-178.9	-158.2	-149.1	-151.3
Mass Flow	kg/h	104155	104155	84366	56508	27858	53384	33596
Molar Flow	mol/h	5000.0	5000.0	4284.0	2869.4	1414.6	1957.7	1241.7
Nitrogen	mol%	40.0	40.0	30.5	30.5	30.5	93.8	92.0
Methane	mol%	60.0	60.0	69.5	69.5	69.5	6.2	8.0

Stream ¹		23	27	28	30	31	35
Pressure ²	MPa	2.48	0.22	0.20	0.15	0.22	0.98
Temperature	°C	-151.3	-188.2	-188.9	28.5	-155.9	28.5
Mass Flow	kg/h	19789	19789	54443	54443	49712	49712
Molar Flow	mol/h	716.0	716.0	1946.7	1946.7	3053.3	3053
Nitrogen	mol%	96.9	96.9	99.6	99.6	2.0	2.0
Methane	mol%	3.1	3.1	0.4	0.4	98.0	98.0

¹As identified in Figure 2

²Pressures are given as absolute values

5 Example 4

Table 4 shows typical operating parameters for the process of the invention using the apparatus shown in Figure 2, when used to separate a gaseous mixture consisting of 30 mol% nitrogen gas and 70 mol% methane. In contrast with the conventional double column apparatus shown in Figure 1, the process and apparatus of the invention is able to maintain good separation efficiency even when the nitrogen content of the gaseous feed is below 35 mol%. With this gaseous mixture, the first stream accounts for 25% by molar flow of the sub cooled product. Thus, using the process and apparatus of the invention, based on the same number of theoretical separation stages in columns (08) and (15) as per Comparative Example 2, it is possible to obtain a nitrogen product stream (Stream 30) having a residual methane content of 1.0 mol% (compared with 11.1 mol% in Comparative Example 2), when producing a methane product stream (Stream 35) having a residual nitrogen content of 2.0 mol%.

Table 4

Stream¹		1	7	9	14	18	19	22
Pressure ²	MPa	3.00	2.00	2.00	0.22	0.22	1.98	1.98
Temperature	°C	35.0	-131.7	-131.7	-175.6	-156.3	-146.9	-153.2
Mass Flow	kg/h	98170	98170	84871	63762	21109	28263	14964
Molar Flow	mol/h	5000.0	5000.0	4513.1	3390.7	1122.5	1071.0	584.1
Nitrogen	mol%	30.0	30.0	23.1	23.1	23.1	86.4	80.0
Methane	mol%	70.0	70.0	76.9	76.9	76.9	13.6	20.0

Stream¹		23	27	28	30	31	35
Pressure ²	MPa	1.98	0.22	0.20	0.15	0.22	1.18
Temperature	°C	-153.2	-187.9	-188.0	29.3	-155.9	29.3
Mass Flow	kg/h	13299	13299	40267	40267	57903	57903
Molar Flow	mol/h	486.9	486.9	1443.7	1443.7	3556.3	3556.3
Nitrogen	mol%	94.2	94.2	99.0	99.0	2.0	2.0
Methane	mol%	5.8	5.8	1.0	1.0	98.0	98.0

¹As identified in Figure 2

5 ²Pressures are given as absolute values

CLAIMS

1. A process for the separation of nitrogen from a gaseous feed comprising a mixture of hydrocarbons and nitrogen gas, the process comprising the steps of:

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- (i) cooling and at least partially condensing the gaseous feed;
- (ii) feeding the cooled and at least partially condensed feed from step (i) to a first fractionation to produce an overhead vapour stream having an enriched nitrogen content and a condensed product having a reduced nitrogen content which is subjected to a second fractionation, which comprises reboil, at a lower pressure than the first fractionation;
- (iii) partially condensing the overhead vapour stream, and separating to provide a liquid stream, which is used to provide reflux to the first fractionation, and a separated vapour stream, which is condensed to provide reflux to the second fractionation; and
- (iv) sub-cooling the condensed product of the first fractionation and dividing the resulting sub-cooled product into at least two streams: a first stream being expanded and fed to the second fractionation, and a second stream being expanded and reheated in heat exchange with the separated vapour stream from step (ii) before being fed to the second fractionation;
- (v) removing a hydrocarbon product stream low in nitrogen from the second fractionation; and
- (vi) removing a nitrogen rich stream from the second fractionation.

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2. A process according to Claim 1, wherein the hydrocarbons in the gaseous feed comprise or consist of methane.

3. A process according to Claim 1 of Claim 2, wherein the gaseous feed comprises or consists of natural gas.

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4. A process according to any one of the preceding claims, wherein the gaseous feed comprises less than 40 mol% nitrogen.
- 5 5. A process according to Claim 4, wherein the gaseous feed comprises less than 35 mol% nitrogen.
6. A process according to Claim 5, wherein the gaseous feed comprises less than 30 mol% nitrogen.
- 10 7. A process according to any one of the preceding claims, wherein the gaseous feed comprises at least 20 mol% nitrogen.
8. A process according to any one of the preceding claims, wherein reboil in the second fractionation is provided at least in part by heat exchange during cooling of the gaseous feed.
15
9. A process according to any one of the preceding claims, wherein the gaseous feed is cooled and at least partially condensed via heat exchange with the hydrocarbon product stream low in nitrogen from the second fractionation and/or the nitrogen rich stream from the second fractionation.
20
10. A process according to Claim 9, wherein the hydrocarbon product stream low in nitrogen is pumped to elevated pressure and evaporated to provide cooling for the gaseous feed.
25
11. A process according to any one of the preceding claims, wherein the gaseous feed is expanded prior to the first fractionation.
12. A process according to any one of the preceding claims, wherein the gaseous feed is split into at least two streams, and each stream is independently processed in accordance with any one of the processes of Claims 8 to 11.
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13. A process according to Claim 12, wherein the at least two streams are directed to the first fractionation.
14. A process according to Claim 12 or 13, wherein reboil in the second fractionation is provided at least in part by heat exchange with at least one of the at least two streams.
15. A process according to Claim 14, wherein the at least one stream used for heat exchange is fed to an intermediate stage of the first fractionation.
16. A process according to Claim 15, wherein at least one of the at least two streams is not used for heat exchange in the second fractionation and is fed to a bottom stage of the first fractionation.
17. A process according to any one of the preceding claims, wherein reboil in the second fractionation is provided at least in part by heat exchange during the partial condensing of the overhead vapour stream.
18. A process according to Claim 17, wherein the heat exchange is between the overhead vapour stream and a liquid product of the second fractionation.
19. A process according to any one of the preceding claims, wherein the first fractionation is at a pressure in the range of 5 to 30 bar (0.5 to 3.0 MPa).
20. A process according to any one of the preceding claims, wherein in step (iv) the first stream is expanded to form a two-phase feed for feeding to the second fractionation.
21. A process according to any one of the preceding claims, wherein in step (iv) the second stream is expanded to form a two-phase feed and reheated via heat exchange for feeding to the second fractionation.

22. A process according to Claim 21, wherein the second stream is reheated via heat exchange with the condensed product from the first fractionation and/or the overhead vapour stream from the first fractionation.
- 5 23. A process according to any one of the preceding claims, wherein the first stream in step (iv) is fed to the second fractionation at a higher stage than the second stream.
- 10 24. A process according to any one of the preceding claims, wherein the nitrogen rich stream from the second fractionation is reheated by heat exchange with the overhead vapour stream from step (ii) and/or the separated vapour stream from step (iii).
- 15 25. A process according to any one of the preceding claims, wherein the hydrocarbon product stream is removed as an at least partly liquefied product.
26. A process according to any one of the preceding claims, wherein the gaseous feed additionally comprises further inert gases.
- 20 27. A process according to Claim 26, wherein the gaseous feed comprises helium.
- 25 28. A process according to any one of the preceding claims, wherein the gaseous hydrocarbon feed is pre-treated to remove impurities and/or other unwanted components which solidify in the first and second fractionations.
29. An apparatus for the separation of nitrogen from a gaseous feed comprising a mixture of hydrocarbons and nitrogen gas, the apparatus comprising of:
- 30 (i) means for cooling and at least partially condensing the gaseous feed;
- (ii) a first fractionator for producing an overhead vapour stream and a condensed product and a second fractionator operable at a lower pressure than the first fractionator;

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- (iii) means for conveying the cooled and at least partially condensed feed from step (i) to the first fractionator;
 - (iv) means for conveying the condensed product from the first fractionator to the second fractionator;
 - 5 (v) means for partially condensing the overhead vapour stream, and means for separating the partially condensed vapour stream to provide a liquid stream, and a separated vapour stream;
 - (vi) means for conveying the liquid stream to the first fractionator which is used to provide reflux to the first fractionation, and means for conveying and
10 condensing the separated vapour stream to provide reflux to the second fractionator; and
 - (vii) means for dividing the condensed product of the first fractionator, prior to entry into the second fractionator, into at least two streams;
 - (viii) means for expanding a first stream prior to entry thereof into the second
15 fractionator, and means for expanding and reheating a second stream prior to entry thereof into the second fractionator;
 - (ix) means for conveying a hydrocarbon product low in nitrogen from the second fractionator; and
 - (x) means for conveying a nitrogen rich stream from the second fractionator.
20
30. An apparatus according to Claim 29, wherein the first and second fractionators are in a stacked configuration.
31. An apparatus according to Claim 29, wherein the first and second fractionators
25 are in a non-stacked configuration, allowing reduction on the overall height of the apparatus.
32. An apparatus according to any one of Claims 29 to 31, wherein the second fractionator comprises a reboil heat exchanger.
30
33. An apparatus according to any one of Claims 29 to 31, wherein the means for expanding the streams comprises liquid or two-phase expansion turbines.

34. An apparatus according to any one of Claims 29 to 33 wherein the second stream in step (viii) is reheated in heat exchange with the separated vapour stream from step (v).

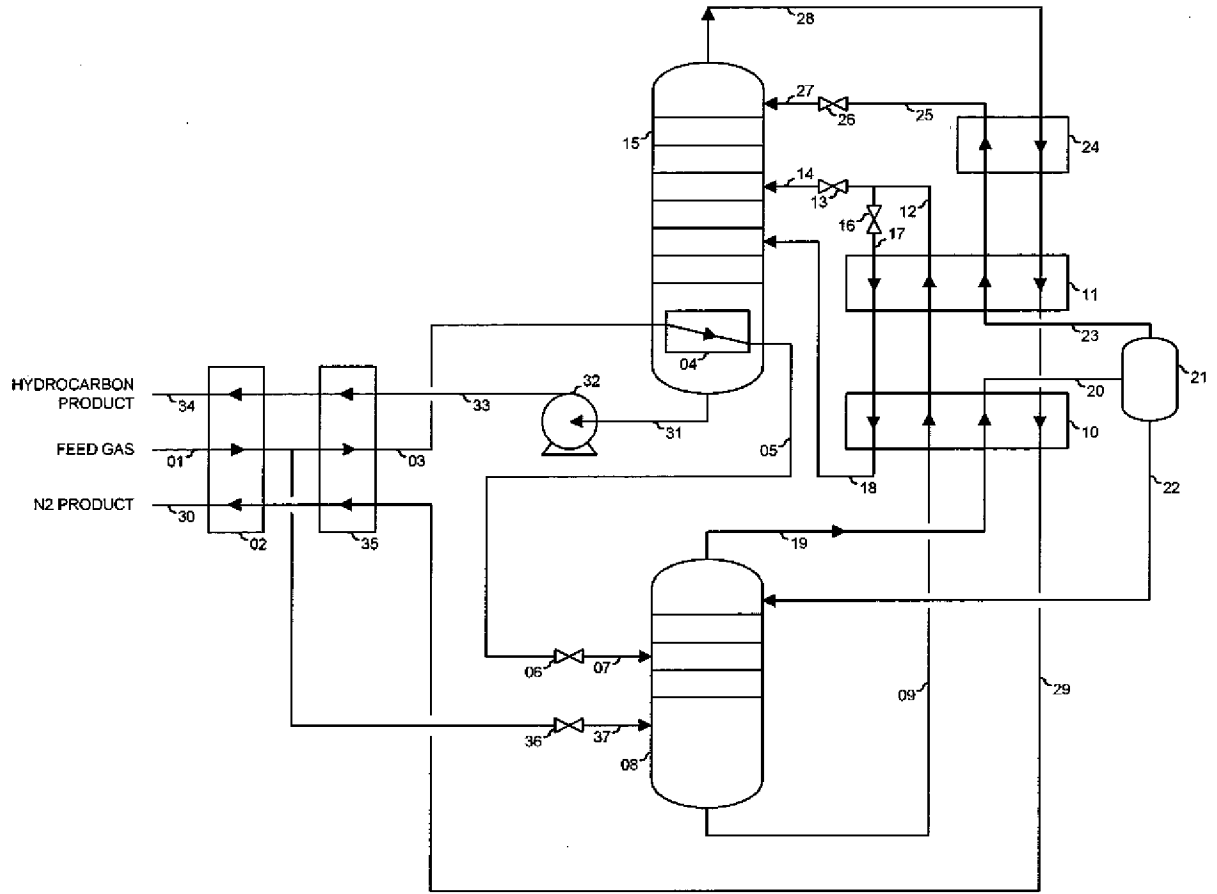


FIGURE 4