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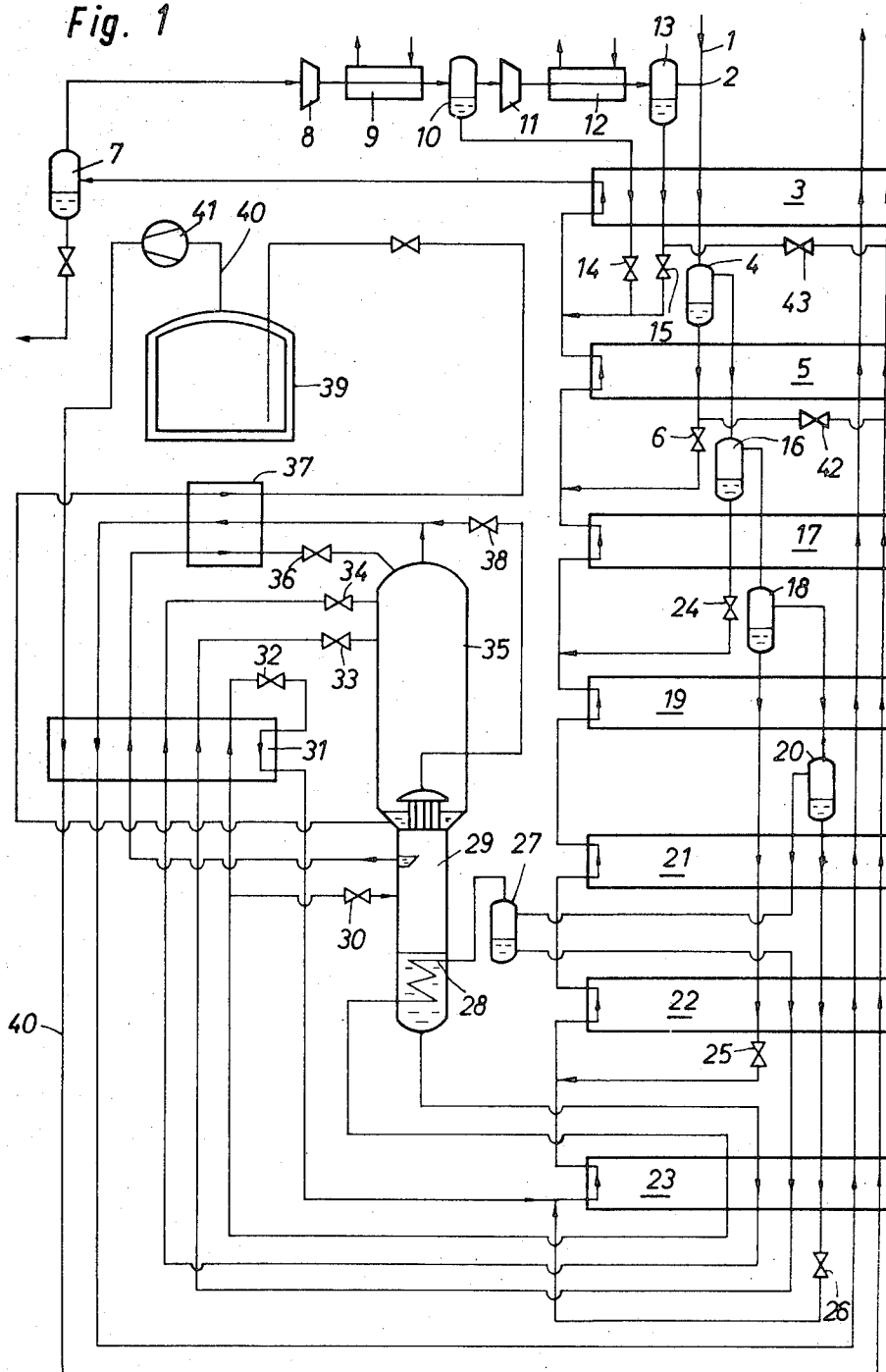
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FRACTIONAL CONDENSATION OF NATURAL GAS

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4 Sheets-Sheet 1

Fig. 1



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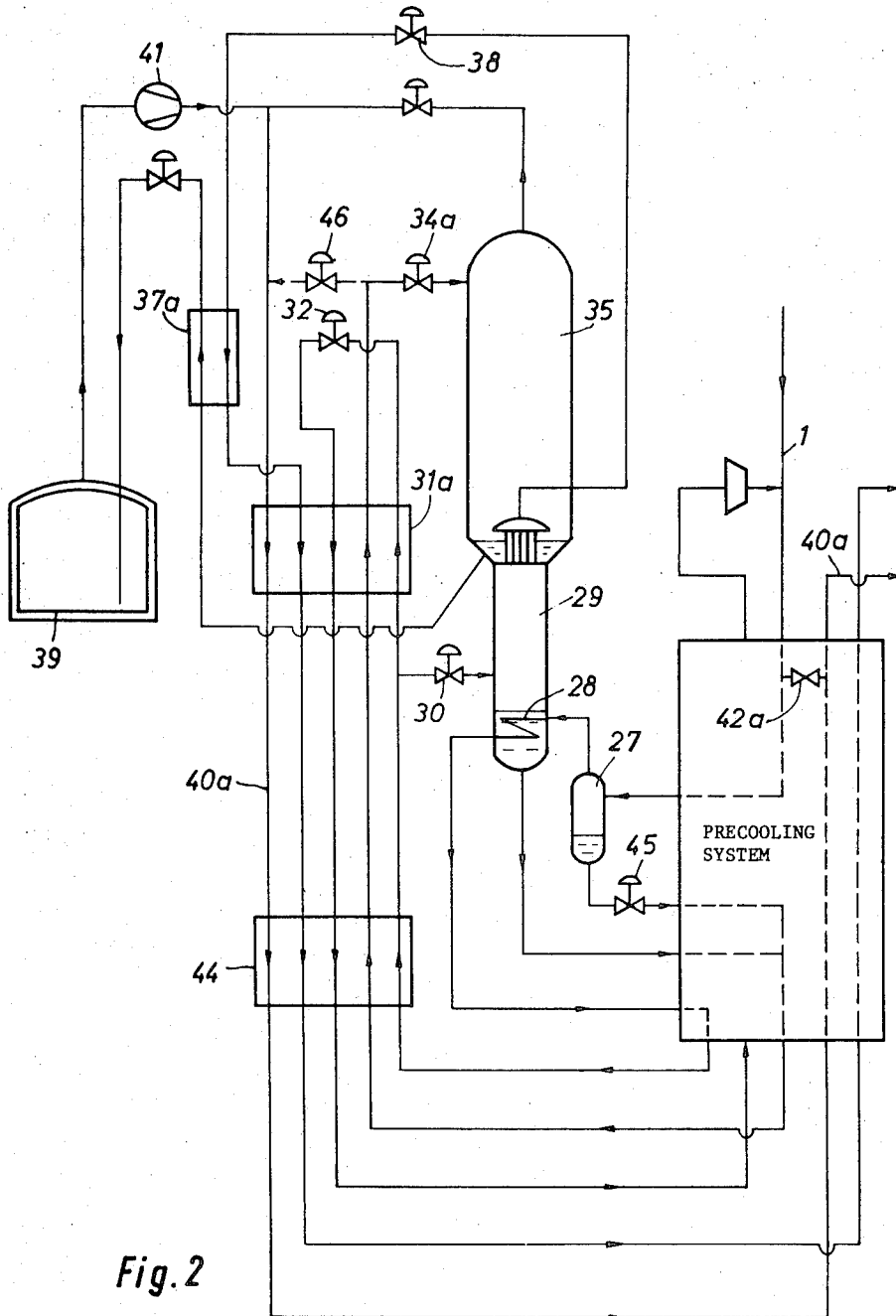


Fig. 2

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4 Sheets-Sheet 3

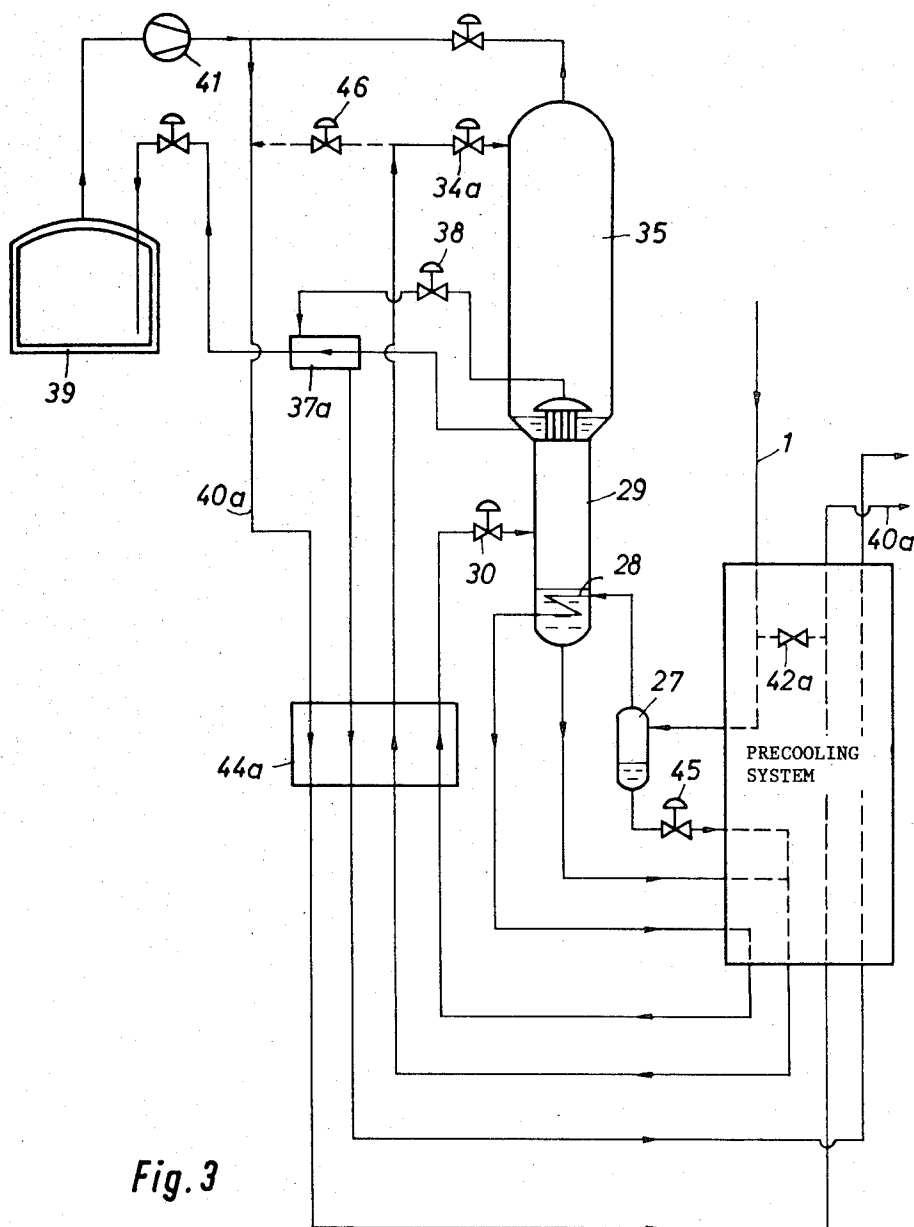


Fig. 3

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4 Sheets-Sheet 4

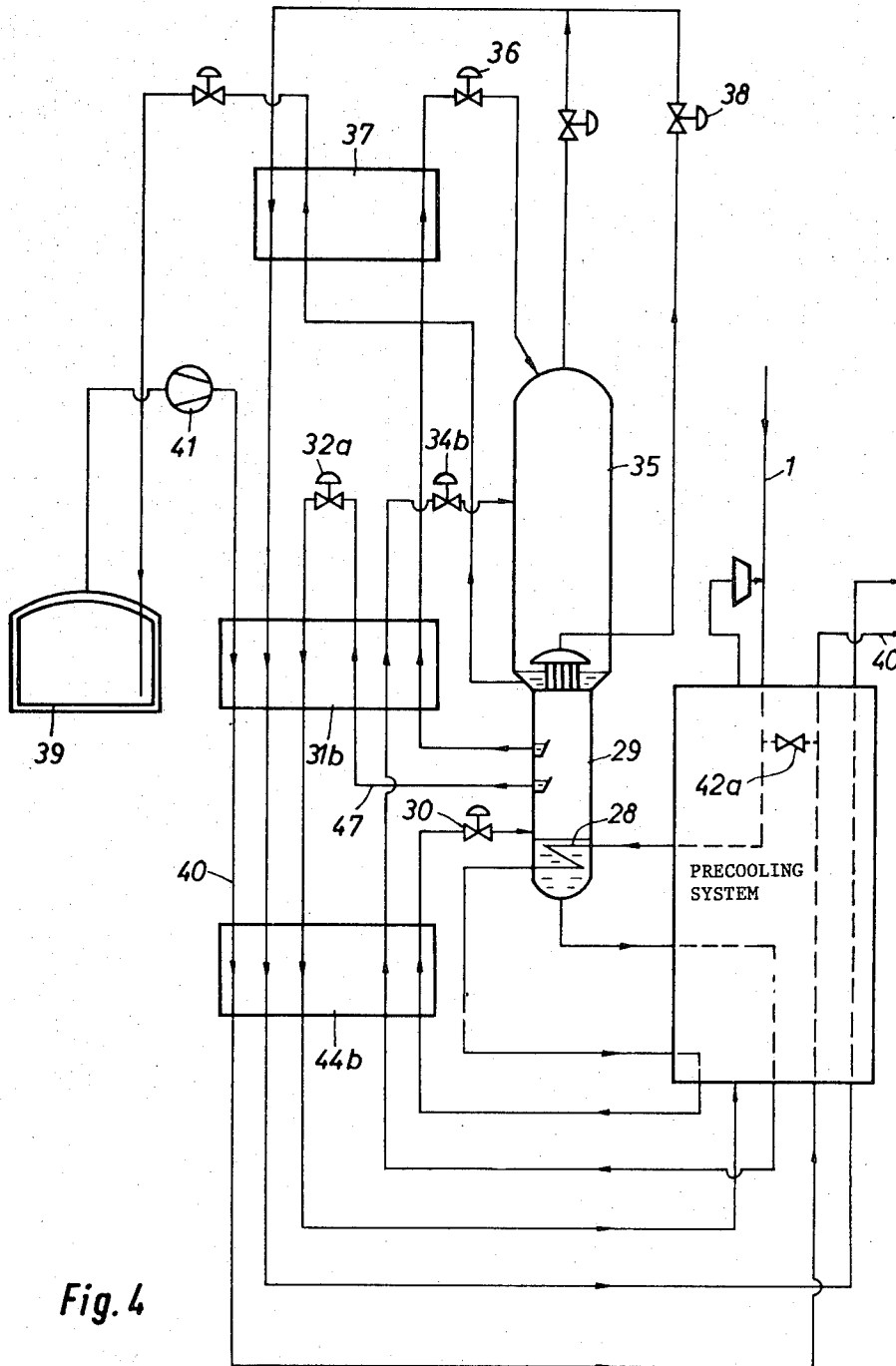


Fig. 4

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FRACTIONAL CONDENSATION OF
NATURAL GAS

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10 Claims

ABSTRACT OF THE DISCLOSURE

A system for the liquefaction of natural gas wherein nitrogen is removed from the gas by rectification, involves fractional condensation of the natural gas and passing a gaseous portion of the thus-treated gas through the sump of a rectification column, in indirect heat exchange with sump liquid. At least a portion of the resultant condensed gaseous portion is returned to the rectification column as feed through an expansion valve. Heat exchangers are employed to subcool natural gas liquid passing to the storage tank and reflux liquid passing to the rectification column, said heat exchangers being cooled by nitrogen fractions withdrawn from the rectification column. The overall system provides for savings in refrigeration energy, and also provides peak refrigeration loads for start-up procedures and the like.

BACKGROUND OF THE INVENTION

This invention relates to a system for the liquefaction of natural gas by fractional condensation under pressure, wherein the nitrogen in the natural gas is separated by rectification.

During natural gas liquefaction, it is advantageous to separate the nitrogen, otherwise it would significantly depress the boiling point and storage temperature of the resultant liquid natural gas. In other words, the presence of nitrogen in the liquefied gas would make it necessary to generate refrigeration at a temperature level below that of boiling temperature of methane. Furthermore, since the nitrogen-containing methane continuously evaporating because of imperfect insulation in the storage tank initially exhibits a high nitrogen concentration, but then decreases with increasing storage time, neither the composition of the liquid gas nor that of the evaporating gas remains constant. This means that for end uses, the composition of the supplied gas must be constantly monitored and must be adjusted to the desired, constant composition by admixing in each case the necessary quantity of one or more other gases.

SUMMARY OF THE INVENTION

It is thus an object of this invention to provide an improved process and apparatus for the removal of a low boiling component during the liquefaction of a higher boiling component, and especially wherein nitrogen is the low boiling component and natural gas is the high boiling component.

Still another object is to provide an improved system wherein nitrogen is removed from the natural gas in such a manner that minimal refrigeration energy is required.

Upon further study of the specification and appended claims, other objects and advantages of the present invention will become apparent.

These objects are attained, with reference to nitrogen removal from natural gas, by providing a nitrogen-methane-ethane mixture which remains in the gaseous phase after the fractional condensation of the hydrocar-

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bons required for the production of refrigeration in the precooling system. This mixture is liquefied by heat exchange against the sump liquid of a high-pressure column of a double rectification column (or equivalent thereof) and is expanded, at least partially, into the high-pressure column, to be separated therein into a substantially pure nitrogen fraction, as the overhead product, and into a methane fraction containing nitrogen and ethane, as the sump liquid.

Any portion of said mixture not fed into the high pressure column is expanded, evaporated and warmed in heat exchange with fractions to be subcooled and fed to the low pressure rectification stage. Said portion is then employed in the precooling system, and subsequently compressed and recycled into the natural gas to be liquefied.

The sump liquid from the high-pressure column is likewise subcooled and expanded into the upper column. From the sump of the upper column, there is withdrawn liquid methane low in nitrogen and containing ethane, and this product is then subcooled prior to being fed into the storage tank. The subcooling is conducted by heat exchange with expanded nitrogen withdrawn in the gaseous phase from the high-pressure column or from both the high-pressure column and the low-pressure column, said nitrogen being additionally warmed against fractions to be subcooled and fed to the rectification column.

An important advantage of this invention is that the pressure of the natural gas is utilized in an especially energy-conserving manner, on the one hand for the production of peak refrigeration levels and, on the other hand, for the separation of nitrogen. The latter is withdrawn from the rectification column either as a substantially pure nitrogen fraction and used for various known purposes as such, or it is employed as a component of town gas in the form of an impure nitrogen fraction to which have been added higher hydrocarbons for adjusting the heating value.

DETAILED DISCUSSION OF THE INVENTION

In a further embodiment of the invention, liquid methane containing nitrogen and ethane is separated, by partial condensation in the precooling system, from the nitrogen-methane-ethane gaseous mixture remaining in the gaseous phase after the fractional condensation of the hydrocarbons required for the production of refrigeration in the precooling system, and prior to the heat exchange of said gaseous mixture with the sump liquid of the high-pressure column. This liquid methane, optionally in admixture with the sump liquid of the high-pressure column, and after a subcooling step, is preferably 100% expanded into the upper low-pressure column, but can also be expanded, evaporated and warmed in heat exchange with fractions to be subcooled and fed to the low-pressure rectification stage. In this manner preliminary nitrogen enrichment is achieved in the gaseous phase.

Another preferred embodiment of the process according to the invention resides in providing that the liquid formed by heat exchange with the sump liquid of the high-pressure column is expanded into the high-pressure column, and that liquid is withdrawn from this column at another point, expanded, evaporated and warmed in heat exchange with fractions being fed to the rectification column; thereafter passed through the precooling system; and then compressed and recycled into the natural gas to be liquefied. The advantage of this technique is that the point of liquid withdrawal from the column, i.e. the composition of the liquid and thus the boiling point thereof, can be adjusted so that exactly the desired temperature is ambient at the cold end of the heat exchanger wherein this liquid is evaporated.

It is especially suitable first to subcool the sump liquid of the high-pressure column and/or the liquid formed in

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heat exchange therewith, after leaving the high-pressure column, in the precooling system. Also the liquid which has been formed (after separating the liquid required in the precooling stage for cold production) by additional partial condensation of the remaining gaseous mixture in the precooling system is advantageously first subcooled in the precooling system. This procedure serves to increase the amount of liquid to be introduced into the column.

In a still further preferred embodiment, the residual portion of the liquid formed by heat exchange with the sump liquid of the high-pressure column, or the liquid withdrawn from the high-pressure column at another point, is subcooled prior to expansion thereof, by counter-current heat exchange with itself and/or with fractionation products, in order to reduce the amount of vapor formed during the subsequent expansion.

Any residual portion of the liquid formed by heat exchange with the sump liquid of the high-pressure column, or the liquid withdrawn from the high-pressure column at another point, after expansion and partial warming, is further warmed, preferably in a mixture with the recycled cycle gas of the precooling system, and then compressed and recycled into the natural gas to be liquefied.

The apparatus for conducting the process of this invention is characterized in that a middle section of the high-pressure column of a double rectification column is in communication with a precooling system for the partial condensation of natural gas via a feed conduit, provided with an expansion valve, for liquid methane containing nitrogen and ethane. The sump of the high-pressure column is in communication with the upper column of the double rectification column via a liquid conduit provided with an expansion valve. A conduit for gaseous nitrogen is connected from the head of the high-pressure column, by way of an expansion valve, with the refrigerating medium side of a heat exchanger, the flow path of said heat exchanger for material to be cooled being in communication, on the one hand, with the sump of the upper column and, on the other hand, with a storage tank for liquid methane. The precooling system is connected downstream of the refrigerating medium side of said heat exchanger via at least one additional heat exchanger for subcooling liquids being fed to the rectification stage.

BRIEF DESCRIPTION OF THE DRAWINGS

All the figures are schematic flowsheets depicting various preferred embodiments of the invention.

FIG. 1 shows the details of the precooling system whereas the other figures depict same in "block" form.

DETAILED DESCRIPTION OF THE DRAWINGS

The invention will now be explained by way of example with reference to the drawings. Identical components are characterized in all figures by the same numerals.

An example of a natural gas to be processed exhibits the following composition:

	Mol percent
He -----	0.04
CO ₂ -----	0.80
N ₂ -----	14.30
O ₂ -----	0.01
CH ₄ -----	81.59
C ₂ H ₆ -----	2.60
C ₃ H ₈ -----	0.40
C ₄ H ₁₀ -----	0.16
C ₅ H ₁₂ -----	0.04
C ₆₊ -----	0.06
	100.00

In general, however, natural gas contains a predominant amount of methane and about 2 to 40% by volume of nitrogen.

Prior to entering the low-temperature plant, the CO₂ is removed by a monoethanolamine scrubbing step, and the gaseous water is removed by exchangeable molecular sieve

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adsorbers. The natural gas then enters the low-temperature plant through conduit 1 at a pressure of about 30 to 45, preferably 35 atmospheres, but can also be fed into one of the previous cycle gas compressor stages, and is mixed with cycle gas at 2, and cooled in heat exchanger 3. Thereby, a condensate is produced containing predominantly higher-boiling hydrocarbons, which condensate is separated from the gas in the phase separator 4. Both the gas and the liquid are further cooled in heat exchanger 5.

The liquid is then expanded in valve 6 into the returning cycle gas, the latter having been formed by the evaporation of lower-boiling liquids in heat exchangers operating at a lower temperature. It is then evaporated and warmed in heat exchangers 5 and 3, and finally fed, via the phase separator 7, to the first compressor stage 8 and the intercooling stage 9 connected thereafter. The thus-formed liquid collects in the phase separator 10. Another compressor stage 11 follows, with cooler 12 and separator 13. The portion remaining in the gaseous phase is fed at 2 into the natural gas to be liquefied. The liquid separately withdrawn from the separators 10 and 13 is subcooled in heat exchanger 3 and expanded (pressure reduced) into the cycle gas through valves 14 and 15, respectively.

The gas leaving the separator 4 and partially condensed in heat exchanger 5 is separated, in the separator 16, from the thus-formed liquid and again subjected to a partial condensation in the heat exchanger 17, passing via the separator 18 and the additional heat exchanger 19 into the separator 20. The amount of liquid produced up to this point is sufficient to compensate for the refrigerating requirements of the precooling system. This liquid is then subcooled, namely, the liquid from separator 16 in the heat exchanger 17, the liquid from separator 18 in heat exchangers 19, 21 and 22, and the liquid from separator 20 in heat exchangers 21, 22 and 23. Thereafter, the liquid is expanded into the cycle gas by means of the valves 24 and 25 and 26, respectively. The thus-formed fluid mixture is then recycled, as cycle gas, into the natural gas to be liquefied, as set out above.

The gas discharged from the separator 20 contains the entire amount of methane to be liquefied, a corresponding proportion of nitrogen, as well as minor amounts of ethane. This gas is cooled in heat exchanger 21 to such an extent that the gaseous phase in the separator 27 contains approximately 25% nitrogen and the liquid phase, in correspondence with the equilibrium, contains about 5% nitrogen. The resultant gas is liquefied and subcooled in the pipe coil 28 which heats the sump of the high-pressure column 29 of the double rectification column, and is passed thereafter through the heat exchanger 23 involved in the precooling system.

A portion of the liquid from heat exchanger 23 is expanded, via valve 30, into the high-pressure column 29 operating at 22 atmospheres absolute; in this high-pressure column, the head product obtained is a substantially pure nitrogen fraction containing only about 5% methane, and the sump product is a liquid, the composition of which is approximately the same as that of the liquid obtained in the separator 27, i.e. 5% nitrogen.

The remaining liquid from the pipe coil 28, instead of being passed through valve 30, is subcooled in heat exchanger 31, expanded in valve 32, and evaporated in heat exchanger 31, thereby transferring its latent "cold" of evaporation during this step, to the fractions passing to the upper column. The resultant evaporated liquid finally passes, together with the liquid expanded in valve 26 (the evaporation temperature of the latter liquid representing the lowest of the precooling system) through the cycle back to the compressor.

The liquid from the separator 27 is subcooled in heat exchanger 22 and then, like the sump liquid of the high-pressure column, further subcooled in heat exchangers 23 and 31, and introduced through valves 33 and 34, respectively, into the upper column 35 of the double rectifier, operating at about 2.5 atmospheres absolute. To this

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upper column is fed, via expansion valve 36, liquid reflux nitrogen previously subcooled in the heat exchangers 31 and 37, and derived from the top part of the high-pressure column. As the overhead product of the upper column there is obtained a gas having 95 molar percent of nitrogen, and to this gas is added, via valve 38, helium-enriched nitrogen from the head of the high-pressure column. This resultant gaseous mixture is warmed in the heat exchangers 37 and 31, as well as in the precooling system, and is then withdrawn from the plant. Owing to the low methane concentration in the nitrogen, the losses in methane are kept at a minimum.

In the sump of the low-pressure column 35 there is collected liquid methane containing about 0.5% nitrogen and approximately 2% ethane. This liquid methane is subcooled in heat exchanger 37 by gaseous nitrogen and expanded into the tank 39, the latter being operated at a superatmospheric pressure of about 350 mm. H₂O. Continuously evaporating gas formed due to the imperfect insulation of the tank 39 is withdrawn via conduit 40, compressed in the cold gas blower 41 and warmed in heat exchanger 31 and in the precooling system. During this procedure, the higher-boiling hydrocarbons to be separated in the precooling section are admixed to this gas via valves 42 and 43.

The system according to FIG. 2 differs from the above-described system in that there is discharged from the plant, in addition to liquid methane and a pure nitrogen fraction, a gaseous mixture having approximately the heating value of the natural gas. The gas coming from the phase separator 27 is liquefied and subcooled in the coil 28 and in the precooling system, and is then further subcooled in countercurrent heat exchanger 44. Thereafter it is expanded, in part, through valve 30 into the high-pressure column 29 and, in part, after being subcooled in the countercurrent heat exchanger 31, through valve 32. The thus liberated refrigeration is transferred, in heat exchangers 31 and 44, to the liquid fractions passing to the rectification stage. Finally, the resultant fluid is warmed in the precooling system and then recycled into the natural gas to be liquefied.

The liquid from the separator 27 is expanded in valve 45 to the pressure of the high-pressure column and, together with the sump liquid of the high-pressure column, subcooled first in the precooling system and thereafter in the heat exchangers 44 and 31, and then expanded via valve 34a into the upper column 35. The gaseous stream withdrawn from the head of this upper column contains, in addition to nitrogen, about 80% methane; this gaseous stream is mixed with the gas evaporating in the storage tank, the latter gas having been compressed by the cold gas blower 41 to about 1.8 atm. abs., and warmed, via conduit 40a, in heat exchangers 31 and 44, as well as in the precooling system. In order to adjust the heating value of this gaseous mixture to that of natural gas, there are added via valve 42a higher-boiling hydrocarbons previously condensed during the course of the precooling step.

From the high-pressure column 29 a gaseous substantially pure nitrogen fraction at a pressure of 22 atm. abs. is withdrawn as the overhead product, expanded in valve 38, and first warmed in the heat exchanger 37a against methane so that the latter is subcooled. The residual refrigeration capacity of the nitrogen stream is transferred, in heat exchangers 31 and 44, to the fractions passing to the rectification column and then further utilized in the precooling system.

Optionally, it is also possible to branch off reflux liquid and expand same through valve 46 into conduit 40a in order to have additional peak refrigeration available, for example, when placing the plant on stream, for the operation of the heat exchanger 31.

FIG. 3 depicts an embodiment of the invention wherein the liquid formed in the pipe coil 28, after being subcooled in the precooling system and in the heat exchanger

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44a, is fed in its entirety into the high-pressure column via valve 30. For subcooling the liquid methane in heat exchanger 37a, there is again employed the substantially pure gaseous nitrogen fraction withdrawn from the high-pressure column 29 via expansion in valve 38. This nitrogen stream, after being warmed in heat exchangers 37a and 44a and then in the precooling system, is discharged from the plant.

FIG. 4 depicts an embodiment of the invention wherein, as in the process shown in FIG. 1, no gas need be discharged as town gas, so that the undesired nitrogen can be separated in as enriched a form as possible as a purer nitrogen fraction. In this connection, the natural gas to be liquefied, after the hydrocarbons necessary for refrigeration production in the precooling system have been liquefied and separated, is liquefied without prior partial condensation in the pipe coil 28 and then subcooled in the precooling system, and in the heat exchanger 44b. It is then completely expanded via valve 30 into the high-pressure column 29. The sump liquid of the high-pressure column is passed via expansion valve 34b into the upper column. To generate peak refrigeration, liquid is withdrawn from the high-pressure column via conduit 47, subcooled in heat exchanger 31b, and expanded in valve 32a. In heat exchangers 31b and 44b, this refrigeration is transferred to the fractions fed to the low-pressure column. The thus-produced gas is further warmed in the precooling system, then compressed, and finally recycled into the natural gas to be liquefied. The advantage of this embodiment resides in that the composition of the liquid to be expanded in valve 32a, and thus the temperature at the cold end of the heat exchanger 31b, can be adapted to existing conditions in an especially advantageous manner by an appropriate selection of the point of withdrawal. Since, just as in the process according to FIG. 1, only the evaporation loss of the storage tank may be discharged as town gas, nitrogen in as pure a form as possible must, in turn, be obtained from the head of the low-pressure column. For this purpose, pure liquid nitrogen is withdrawn at the head of the high-pressure column, subcooled in heat exchangers 31b and 37, and fed via valve 36 as reflux to the upper column. From the head of this column, a gaseous pure nitrogen fraction is withdrawn which still contains approximately 5% methane. Helium-enriched gaseous nitrogen from the head of the high-pressure column is admixed via valve 38 to the aforementioned fraction. The resultant nitrogen stream is first utilized in heat exchanger 37 for subcooling both liquid methane and reflux liquid for the upper column. It then transfers additional refrigeration, in heat exchangers 31b and 44b, to the fractions passing to the rectification column, and is finally warmed in the precooling system.

From the foregoing description, one skilled in the art can easily ascertain the essential characteristics of this invention, and without departing from the spirit and scope thereof, can make various changes and modifications of the invention to adapt it to various usages and conditions.

What is claimed is:

1. In a process for the liquefaction of natural gas by fractional condensation under pressure, and with the separation of nitrogen from said gas by rectification, wherein at least one liquid hydrocarbon fraction is, for purposes of refrigeration production, expanded in a precooling cycle, evaporated and warmed in heat exchange with media to be cooled, compressed, and recycled into the natural gas to be cooled,

the improvement comprising liquefying a gaseous nitrogen-methane-ethane mixture remaining after the fractional condensation of the hydrocarbon fraction required for the production of refrigeration in the precooling system, said liquefying being conducted by passing said mixture in heat exchange with sump liquid of a high-pressure column of a double rectifi-

cation column; expanding resultant liquid at least partially, into the high-pressure column; separating said expanded liquid in said high-pressure column into a substantially pure gaseous nitrogen fraction, as the head product of the high-pressure column, and into a methane fraction containing nitrogen and ethane, as the sump liquid; withdrawing and expanding said substantially pure gaseous nitrogen fraction; subcooling and expanding the sump liquid from the high-pressure column and passing same into the low-pressure column; withdrawing from the sump of the low-pressure column liquid methane low in nitrogen and containing ethane, and subcooling said liquid methane by passing same in indirect heat exchange with said substantially pure expanded nitrogen fraction, whereby peak cooling is attained.

2. A process as defined by claim 1 wherein said resultant liquid, not expanded into the high-pressure column, is expanded, evaporated and warmed in heat exchange with streams to be subcooled and fed to the rectification column.

3. A process as defined by claim 1 wherein said liquid methane containing nitrogen and ethane is separated by partial condensation in the precooling system, from the nitrogen-methane-ethane mixture remaining in the gaseous phase after the fractional condensation of the hydrocarbons required for the production of refrigeration in the precooling system, prior to the heat exchange of said mixture with the sump liquid of the high-pressure column, said liquid methane being then expanded, after subcooling, into the upper column.

4. A process as defined by claim 1 wherein said resultant liquid mixture is expanded in its entirety into the high-pressure column.

5. A process as defined by claim 1 wherein the sump liquid from the high-pressure column is subcooled in the precooling cycle after leaving the high-pressure column.

6. A process as defined by claim 3 wherein said liquid methane is subcooled in the precooling cycle before being passed as feed to the low-pressure column.

7. A process according to claim 2 wherein said resultant liquid not expanded into the high-pressure column is warmed, after expansion and partial warming in admixture with returning cycle gas of the precooling cycle, then compressed and recycled into the natural gas to be liquefied.

8. Apparatus for liquefying natural gas and removing nitrogen therefrom, said apparatus comprising precooling means for the partial condensation of natural gas comprising an open cycle containing a plurality of serially connected heat exchangers, a plurality of serially connected phase separators, expansion means, compressor

means, and natural gas feed means, a high-pressure column (29) of a double rectification column, said high-pressure column (29) being in communication with the gas side of the most downstream phase separator, a conduit for said communication provided with cooling means (21); phase separator means (27); coil means (28) in the sump of the high-pressure column and an expansion valve (30), the sump of the high-pressure column (29) being in communication with an upper column (35) of the double rectification column via a liquid conduit provided with an expansion valve (34; 34a; 34b); a heat exchanger (37; 37a); a conduit for gaseous nitrogen being connected from the head of the high-pressure column (29), by way of an expansion valve (38), with the refrigerating medium side of said heat exchanger (37; 37a), the flow path of said heat exchanger for medium to be cooled being in communication with the sump of the upper column (35) and with a storage tank (39) for liquid methane.

9. Apparatus as defined by claim 8 further comprising conduit means for effect communication between the liquid side of said phase separator (27) and the top of the upper column (35), said conduit means being provided with an expansion valve (34a); and further heat exchange means (44a) for cooling feed streams to the double rectification column against nitrogen fractions withdrawn from said column.

10. A process as defined by claim 1 wherein said gaseous nitrogen-methane-ethane mixture is derived from a gaseous stream withdrawn from a single phase separator in said precooling cycle, said stream containing the entire amount of methane to be liquefied, said gaseous stream being subjected to further cooling and further phase separation to obtain said gaseous nitrogen-methane-ethane mixture.

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