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SEPARATION OF LOW BOILING HYDROCARBONS AND NITROGEN BY FRACTIONATION WITH PRODUCT STREAM HEAT EXCHANGE

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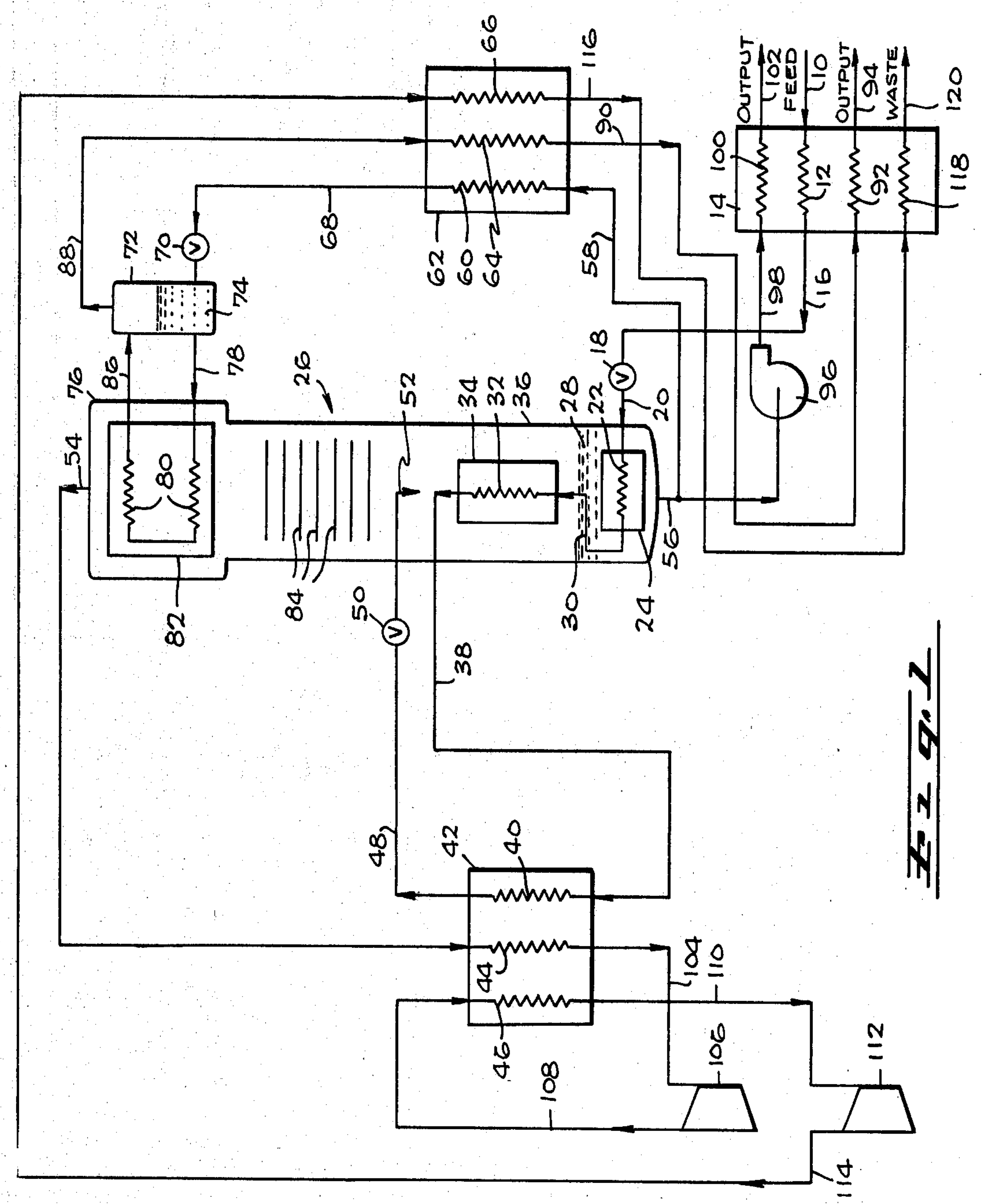

Fig. 1

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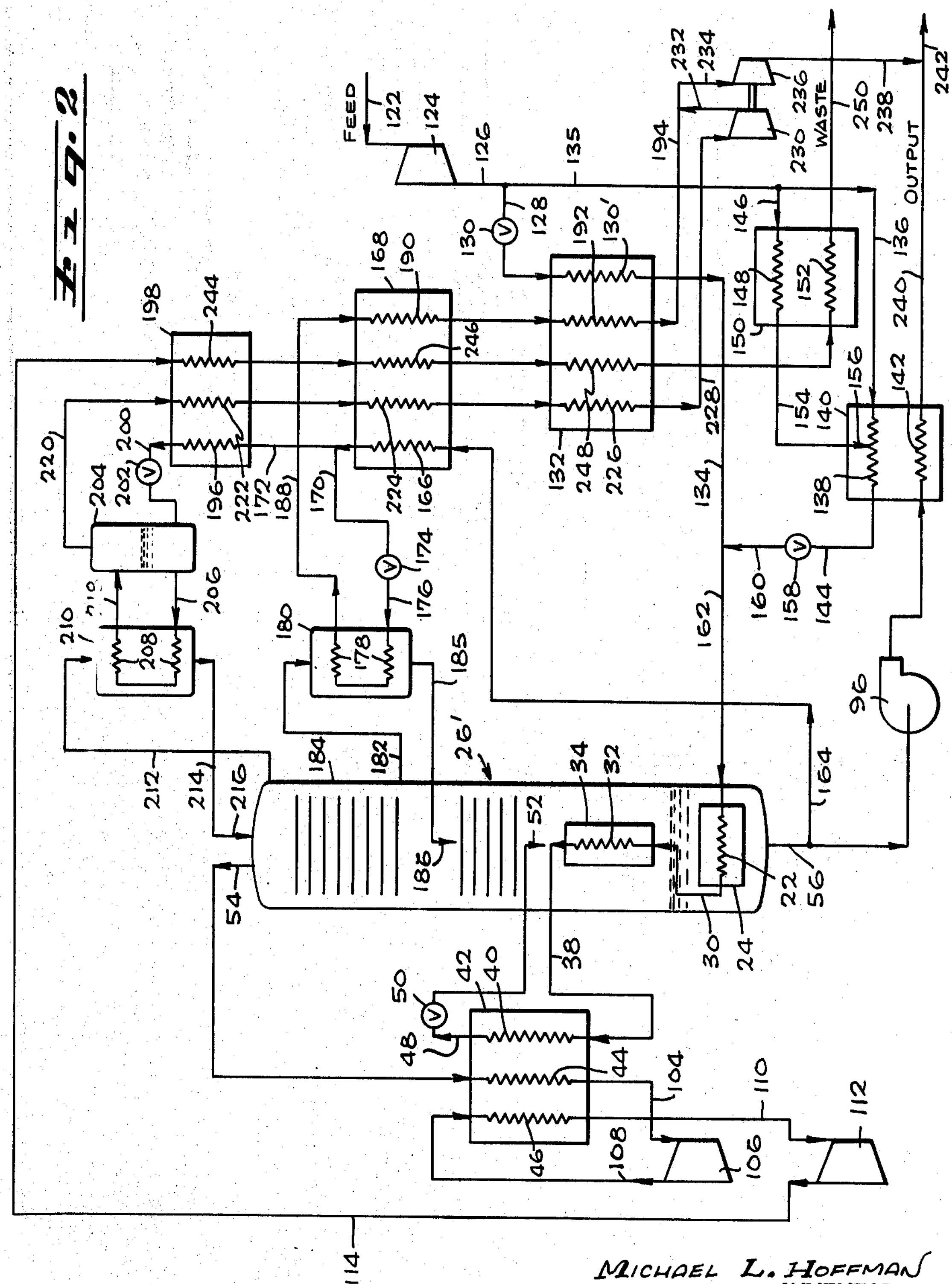
Fig. 2



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SEPARATION OF LOW BOILING HYDROCARBONS AND NITROGEN BY FRACTIONATION WITH PRODUCT STREAM HEAT EXCHANGE

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ABSTRACT OF THE DISCLOSURE

System for the separation of gases, particularly of mixtures of a low boiling hydrocarbon and nitrogen, e.g., a mixture of methane and nitrogen, which involved providing a mixture of such low boiling hydrocarbon and nitrogen at relatively high pressure, e.g., about 1,000 p.s.i., cooling the compressed gas mixture to a temperature close to its saturation temperature, passing the cooled compressed mixture in heat exchange relation with the lower portion of a fractionating column operating at a pressure substantially lower than the pressure of the compressed gas mixture, and providing reboil heat to the column, subcooling the existing compressed feed gas mixture, reducing the pressure of the subcooled compressed mixture approximately to the pressure in the column, introducing the resulting mixture as feed into the fractionating column, and effecting a separation of the mixture in the column into a nitrogen fraction and a hydrocarbon fraction, passing overhead nitrogen from the column in heat exchange relation with the compressed feed mixture for subcooling same, preferably work expanding the exiting nitrogen and recycling the expanded nitrogen again in heat exchange relation with the compressed feed mixture, work expanding the exiting nitrogen and further cooling same, withdrawing low boiling hydrocarbon in substantially pure liquid form from the lower portion of the column, passing a portion of such liquid hydrocarbon in heat exchange relation with the work expanded nitrogen for subcooling such hydrocarbon, throttling the subcooled portion of liquid hydrocarbon and further reducing the temperature and pressure thereof, employing the throttled hydrocarbon to provide condensing duty in the upper portion of the column, passing the resulting exiting vaporized hydrocarbon and the main portion of the liquid hydrocarbon withdrawn from the column in heat exchange relation with the compressed feed mixture for cooling same, compressing the hydrocarbon product to desired pressure, and passing nitrogen withdrawn from heat exchange relation with liquid hydrocarbon, into heat exchange relation with the compressed gas mixture for cooling same.

This invention relates to the separation of the components of gas mixtures, particularly mixtures of nitrogen and a low boiling hydrocarbon such as methane, ethane, ethylene or propane, by rectification, and is particularly concerned with procedure for the separation of methane in substantially pure form from mixtures thereof with nitrogen, and with a system for carrying out such procedure.

The processing or separation of the components of a gas mixture containing low boiling hydrocarbon and nitrogen, such as for example mixtures of a major portion of methane and a minor portion of nitrogen, to recover substantially pure hydrocarbon, such as methane, for use as a low cost fuel which can be readily transported by pipeline and stored, has assumed considerable importance. By the term hydrocarbons or methane "in substantially pure form" as employed herein, is intended to denote such hydrocarbons or methane of at least 95% purity, and preferably of the order of 97% or greater. In such separation operations, it is often desirable to provide a feed gas mixture to be separated which is at a pressure higher than the critical pressure of either component of the mixture, e.g., nitrogen or methane, or any mixtures thereof, and to recover the less volatile component, that is the hydrocarbon or methane component, at a pressure at or near the feed mixture pressure.

Accordingly, the process and system of the invention are particularly designed for the separation of low boiling hydrocarbon such as methane, and nitrogen, from a mixture thereof, wherein the separation of the gases is carried out by an efficient distillation procedure, to recover the hydrocarbon in substantially pure form, and particularly wherein the feed mixture is at the above noted high pressure and the hydrocarbon component is recovered close to or approximately at the feed gas pressure.

According to the present invention, there is thus provided a process for the separation of a mixture of gases consisting essentially of a low boiling hydrocarbon, e.g., methane, and nitrogen, which comprises the steps of cooling a compressed feed mixture of the gases to a temperature close to its saturation temperature, passing the cooled compressed feed mixture in heat exchange relation with the lower portion of a fractionating column to provide reboil heat thereto, said column operating at a pressure substantially lower than the pressure of the compressed gas mixture, subcooling the exiting feed gas mixture, reducing the pressure of said compressed mixture approximately to the pressure in the column, introducing the last-mentioned mixture as feed into the fractionating column, effecting a separation of said mixture into a nitrogen fraction and hydrocarbon fraction, withdrawing nitrogen as overhead from the column, passing said overhead nitrogen in heat exchange relation with the compressed mixture for subcooling same as aforesaid, work expanding the exiting nitrogen and further cooling same, withdrawing low boiling hydrocarbon in substantially pure liquid form from the lower portion of the column, separating a portion of said liquid hydrocarbon, passing the work expanded nitrogen in heat exchange relation with such portion of liquid hydrocarbon for subcooling same, throttling said subcooled portion of liquid hydrocarbon and further reducing the temperature and pressure thereof, and employing such throttled hydrocarbon to provide condensing duty in the upper portion of the column required to effect the separation of said mixture therein.

Preferably, the compressed feed mixture, after initial cooling thereof, is passed in heat exchange relation along the lower portion of the fractionating column to effect a more efficient distillation in the column, and most desirably the cooled compressed feed gas mixture is first passed transversely across the bottom of the column, and then upwardly along the lower portion of the column, to provide reboil heat for the column.

In preferred practice, the overhead nitrogen withdrawn from the column is passed several times in heat exchange relation with the compressed feed mixture for cooling same, for example, according to a preferred embodiment, after the first passage of such nitrogen in heat exchange relation with the compressed feed mixture, exiting nitrogen is work expanded to an intermediate pressure to further cool same, and the work expanded nitrogen is again recycled in another pass in heat exchange relation with the compressed feed mixture, following which the exiting nitrogen is finally work expanded to a pressure close to atmospheric pressure, and further cooling the nitrogen prior to passage thereof in heat exchange relation with a portion of the liquid hydrocarbon for subcooling same to permit the latter to provide the above-noted condensing duty for the column.

According to one embodiment, the subcooled liquid hydrocarbon, e.g., methane, is throttled down to a pressure close to atmospheric, and is passed through the coils of a condenser in the upper portion of the column, the liquid methane vaporizing in such coils and providing condensing duty in the upper portion of the column, and the vaporized methane is passed in heat exchange relation with that portion of the liquid hydrocarbon or methane withdrawn from the lower portion of the column and diverted to provide the above-noted condensing duty.

According to another modification, the portion of liquid hydrocarbon, e.g., methane, withdrawn from the lower portion of the column and employed for condensing duty, is initially subcooled by heat exchange relation with work expanded overhead nitrogen vapors, is throttled to an intermediate pressure and a corresponding reduced temperature, and is brought in heat exchange relation in a condenser with contents of an intermediate portion of the column, and a portion of the initially subcooled liquid hydrocarbon vapor is diverted and further subcooled in heat exchange relation with expanded overhead nitrogen vapors and throttled to a low pressure approximately atmospheric and further cooled. The last-mentioned throttled hydrocarbon, e.g., methane, is then vaporized in a second condenser in heat exchange relation with contents of the upper portion of the column and the vaporized hydrocarbon or methane from both condensers is used to aid in subcooling the compressed liquid hydrocarbon initially withdrawn from the lower portion of the column and employed for the condensing duty described above. This modification provides increased efficiency of operation of the column.

In each of the embodiments of the invention process and system, preferably substantially pure low boiling hydrocarbon, e.g., methane, separated in the column is passed in heat exchange relation with compressed feed mixture for initially cooling same, and the low boiling hydrocarbon or methane is compressed prior to passage in heat exchange relation with the compressed feed mixture, to deliver low boiling hydrocarbon or methane at a pressure close to that of the compressed feed mixture. As an illustration, where a compressed feed mixture of 1,000 p.s.i. is employed, substantially pure liquid hydrocarbon, e.g., methane, product at a pressure of about 800 p.s.i. can be produced.

Also, in preferred practice waste nitrogen from the system is passed in heat exchange relation with compressed feed mixture for cooling same.

The invention will be understood more clearly by the description below of certain preferred embodiments of the invention taken in connection with the accompanying drawing wherein:

FIG. 1 is a schematic representation of one preferred form of the invention system for separating substantially pure low boiling hydrocarbon, e.g., methane, from a mixture of such hydrocarbon and nitrogen; and

FIG. 2 is a schematic representation of another preferred modification of the system of FIG. 1.

Referring to FIG. 1 of the drawing, a compressed natural gas feed mixture at **10** containing 85% methane and 15% nitrogen at a pressure of 1,000 p.s.i. (absolute) and at about ambient temperature, is cooled to a temperature of about 189° K., by passage of such compressed feed gas through coil **12** of a main heat exchanger **14** in countercurrent heat exchange relation with coil product and waste gas streams, as will be described more fully hereinafter.

The resulting compressed feed mixture at **16** is throttled at **18** to a reduced pressure of about 600 p.s.i. and close to its saturation temperature. The throttled feed mixture at **20** is passed through coil **22** of a reboiler **24** in the bottom of a fractionating column **26**, in indirect heat exchange relation with the boiling liquid contents **28** in the bottom of the column.

The compressed feed mixture passing through the reboiler **24** is cooled to a temperature of about 172° K., and providing reboil heat to the bottom of the column. The resulting compressed feed mixture at **30** is introduced into the lower end of one or more passages **32** of heat exchange unit **34** in the lower portion of the fractionating column, and the mixture passing upwardly through the heat exchange passages **32** in indirect heat exchange relation with the contents of column **26**, provides additional heat for reboiling in the lower or enriching section **36** of the column. The compressed feed mixture passing through the heat exchange passages **32** is usually in compressed liquid form and is cooled therein, and the exiting compressed liquid at **38** and at a pressure of 600 p.s.i. and at a temperature of 152° K. is subcooled by passage through coil **40** of a heat exchanger **42** in indirect heat exchange relation with waste overhead nitrogen from column **26**, passing through coils **44** and **46** of the heat exchanger. The compressed feed mixture at **48** now at a reduced temperature of 147° K., is throttled at **50**, to about 300 p.s.i., the pressure within column **26**, and reduced in temperature to bubble point temperature, and the throttled mixture is then introduced as feed at an intermediate point indicated at **52**, into the column.

The mixture in the column **26** is separated into a low boiling fraction, namely, nitrogen, and a higher boiling fraction, that is, methane, the nitrogen being withdrawn as overhead at **54** from the top of the column, and substantially pure liquid methane is withdrawn at **56** from the bottom of the column.

A portion **58**, e.g., about 25%, of the liquid methane product withdrawn at **56** from the column, at a pressure of about 300 p.s.i. and a temperature of about 165° K. is passed through a coil **60** of the heat exchanger **62** in countercurrent heat exchange relation with vaporized methane passing through coil **64** and waste nitrogen passing through coil **66**, and the resulting subcooled liquid methane at **68**, cooled to a temperature of about 119° K. is then thottled at **70** to a pressure of 21 p.s.i. and a temperature of about 115° K., and is introduced into a vapor-liquid separator **72**. The liquid methane at **74** in separator **72** is at a sufficiently low temperature so that it can be employed for providing condensing duty in the upper portion or stripping section **76** of column **26**.

Accordingly, liquid methane at 115° K. is introduced at **78** into the coils **80** of a condenser **82** positioned in the top of the fractionating column for condensation of nitrogen and production of reflux at the top of the column, which descends over the trays **84** in the upper portion of the column in countercurrent heat exchange relation with ascending vapors passing upwardly in contact with the descending liquid on the column trays **84**, to provide equilibrium between vapor and liquid in the upper portion of the column. It is noted that the compressed feed mixture passing through heat exchange passages **32** in the lower portion of the column and providing reboil duty therein, is brought into heat exchange relation along the length of the lower portion of the fractionating column and effecting a nonadiabatic differential distillation therein. In this manner, substantially greater efficiency is achieved and equilibrium between liquid and vapor is obtained incrementally along the column, whereby equilibrium is much more closely approached throughout the column, thus substantially increasing the efficiency of separation in column **26**.

The methane passing through coils **80** of condenser **82** is vaporized, and the exiting vaporized methane at **86** is returned to the vapor-liquid separator **72**, and overhead methane vapor withdrawn from the separator at **88**, at a pressure of about 20 p.s.i. and a temperature of about 115° K. is conducted through coil **64** of the heat exchanger **62** in heat exchange relation with compressed liquid methane at **60**, for cooling same. The exiting methane vapor at **90**, heated to a temperature of about 162° K. is then passed through coil **92** of the main heat exchanger **14** and is delivered as output at **94**, at about 16 p.s.i. and at ambient temperature.

The remaining or usually major portion of the liquid methane withdrawn at **56** from the column is pumped by means of a liquid pump **96** to delivery pressure of about 800 p.s.i., and the compressed liquid at **98** is passed through a coil **100** of the main heat exchanger **14** in countercurrent heat exchange relation with incoming compressed feed mixture at **12** for cooling same, and the exiting compressed liquid methane product at **102** is provided at about 800 p.s.i. pressure and about ambient temperature. If desired, the minor portion of methane output at **94** can be compressed or pumped to a pressure of about 800 p.s.i. and mixed with the major product output portion at **102**.

The portion of the liquid methane which is diverted at **58** and employed to provide condensing duty in the column, can vary depending upon the composition of the feed mixture. Thus, for example, from about 10 to about 50% of the total methane liquid withdrawn at **56** can be so diverted at **58**, and usually a minor portion of about 20 to about 30% of such liquid methane is thus employed for condensing duty in the column.

The nitrogen vapor overhead fraction withdrawn at **54** from the top of the column, at a pressure of 300 p.s.i. and a temperature of 117° K., is passed through coil **44** of heat exchanger **42** in heat exchange relation with liquid feed mixture at **40** for cooling same, and the warmed nitrogen exiting vapor at **104**, at a temperature of 151° K. is introduced into a work expander or turbine **106**, the nitrogen being expanded therein to a discharge pressure of 80 p.s.i. and a reduced temperature of 108° K. The work expanded nitrogen at **108** is then recycled through coil **46** of heat exchanger **42** again in heat exchange relation with liquid feed mixture at **40** to provide additional cooling therefor, and the exiting nitrogen at **110**, at a pressure of 75 p.s.i. and heated to a temperature of 152° K. is introduced into a second work expander or turbine **112**, reducing the pressure of the discharged nitrogen at **114**, to a pressure of about 20 p.s.i. and a reduced temperature of 111° K. The nitrogen at **114** is then conducted through coil **66** of the heat exchanger **62** to provide additional cooling for the compressed liquid methane at **60**, and the exiting nitrogen at **116** now at a pressure of about 17 p.s.i. and warmed to about 163° K. is conducted through coil **118** of the main heat exchanger **14** to cool compressed feed mixture at **12**, and is discharged as waste at **120**.

In the system as described above and illustrated in FIG. 1, it is noted that the turbines or work expanders **106** and **112** for work expansion and cooling of the overhead nitrogen provide the requisite refrigeration for the system. It is also noted that the delivery pressure of the substantially pure methane at **102** is about 800 p.s.i., somewhat below the feed pressure of about 1,000 p.s.i.

In FIG. 2 of the drawing there is shown a modification of the process and system described above and illustrated in FIG. 1, the system of FIG. 2 having a somewhat greater efficiency than that of FIG. 1. Referring to FIG. 2, the compressed feed gas mixture at **122**, e.g., a compressed natural gas feed containing 85% methane and 15% nitrogen at a pressure of 1,000 p.s.i., is conducted to a work expander **124**, the discharged work expanded feed mixture at **126** being reduced to a pressure of about 825 p.s.i. A portion, e.g., about 25%, of such work expanded and compressed feed gas mixture is divided at **128** from the main stream **126**, and is throttled at **130** to a reduced pressure of about 625 p.s.i. and reduced temperature, and is then conducted via coil **130′** of a heat exchanger **132** in cocurrent heat exchange relation with cold product and waste gas streams, as will be described more fully hereinafter, and the cooled compressed gas mixture is discharged at **134** at about 600 p.s.i. pressure. The remaining portion **135** of work expanded feed mixture at **126**, at a pressure of 825 p.s.i. is then divided into two streams, one of which at **136** is conducted through coil **138** of a heat exchanger **140** in heat exchange relation with cold high pressure methane product stream passing through coil **142**, and is discharged at **144**. The second stream **146** of work expanded feed mixture at 825 p.s.i., is introduced into coil **148** of a heat exchanger **150** in counter-current heat exchange relation with cold nitrogen waste gas passing through coil **152**, and the cooled compressed feed mixture at **154** is then introduced at **156** into a suitable intermediate portion of coil **138** of the heat exchanger **140**, for admixture with the first stream **136** of the compressed feed mixture. The resulting cooled compressed feed mixture at **144** is then throttled at **158** to a pressure of about 600 p.s.i. and reduced temperature, and the throttled feed mixture at **160** is then combined with the initially divided portion of the cooled compressed feed mixture at **134**, and the combined feed mixture **162** is introduced into coil **22** of the reboiler **24** in the lower portion of the fractionating column **26′**, to supply reboil heat thereto.

The same operational steps and elements as are employed in FIG. 1 for further cooling the exiting compressed feed mixture at **30**, prior to introducing such compressed feed mixture into the column at **52**, are employed in the system of FIG. 2, and the same numerals are employed in FIG. 2 corresponding to the same elements in FIG. 1 for this purpose.

In FIG. 2, a portion **164** of the liquid methane product withdrawn at **56** from the column, at a pressure of about 300 p.s.i., is cooled by passage through coil **166** of a heat exchanger **168**, in countercurrent heat exchange relation with cold product and waste gas streams described more fully below, and the exiting cold methane product is divided into a first stream **170** and a second stream **172**. The first stream **170** is throttled at **174** down to an intermediate pressure of about 67 p.s.i. and at an intermediate reduced temperature of about 133° K., and the throttled liquid methane at **176** is then passed through coils **178** of a condenser **180** in countercurrent heat exchange relation with a vapor mixture withdrawn at **182** from an intermediate portion of the upper or stripping section **184** of the column, and conducted to condenser **180**, such vapor being condensed therein and the condensed liquid discharged from the bottom of condenser **180** then being returned at **185** and reintroduced into the column at **186**. The methane heated and vaporized in coils **178** of condenser **180** is then introduced via **188** into coil **190** of heat exchanger **168** for cooling compressed liquid methane at **166**, and the exiting methane is then conducted through coil **192** of heat exchanger **132** for cooling compressed feed mixture at **130′**, and is discharged at **194**.

The second stream **172** of cooled liquid methane exiting coil **166** of heat exchanger **168** is further cooled by passage through coil **196** of heat exchanger **198** in countercurrent heat exchange relation with product and waste streams as described below, and the further cooled liquid methane at **200** is throttled at **202** to a reduced pressure of 21 p.s.i. and a reduced temperature of 115° K., and is introduced into the vapor-liquid separator **204**. Liquid methane from the separator is conducted at **206** through coils **208** of a second condenser **210** to condense essentially nitrogen vapor conducted via line **212** from the top of column **26′** to the condenser, the condensed liquid nitrogen being withdrawn at **214** from the bottom of condenser **210** and reintroduced at **216** as reflux to the top of the column. It will be noted that as in the case of the system of FIG. 1, the methane stream at **200** is throttled at **202** to a pressure and a corresponding reduced temperature such that the resulting liquid methane can be employed to condense essentially nitrogen vapor from the top of the column, to provide adequate reflux for the column. Also, of course, the methane stream at **170** is throttled at **174** to an intermediate pressure and a reduced temperature such that the methane at **178** is at a temperature sufficiently low to condense the vapor mixture withdrawn at **182** from an intermediate section of the upper column portion **184**.

The vaporized methane from condenser **210** returns at **218** to the vapor-liquid separator **204**, and such vapor withdrawn from the separator at **220** is first passed through coil **222** of heat exchanger **198** for cooling compressed liquid methane at **196**, and the exiting methane vapor is then passed through coil **224** of heat exchanger **168** for cooling the compressed liquid methane stream at **166**, and the thus further heated methane stream is finally conducted through coil **226** of the heat exchanger **132** to aid in cooling compressed feed mixture at **130'**, and the exiting methane stream at **228** is then compressed at **230** to a pressure of about 63 p.s.i., and the discharge stream at **232** is combined with the methane stream **194**, and such combined stream **234** is then further compressed at **236** to product pressure of 800 p.s.i., and the compressed methane discharge at **238** is then combined with that portion of the liquid methane product withdrawn at **56** and pumped at **96** to discharge pressure of 800 p.s.i. and which was passed through coil **142** of the heat exchanger **140** and discharged at **240**, thus providing a single combined methane product output of 800 p.s.i. at **242**.

The nitrogen overhead fraction withdrawn at **54** from the top of the column in FIG. 2 is employed similarly as described above and illustrated in FIG. 1, for subcooling compressed feed gas mixture, and to provide refrigeration by expansion thereof, and the same elements are employed for this purpose in FIG. 2 as in the case of FIG. 1, with the same numerals designating corresponding elements.

In FIG. 2 the final work expanded nitrogen discharge at **114** is passed through coil **244** of the heat exchanger **198** and then through coil **246** of heat exchanger **168** for cooling compressed liquid methane streams **196** and **166**, respectively, and the exiting heated nitrogen is then passed through coil **248** of heat exchanger **132** for cooling a portion of the compressed feed gas mixture at **130'**, and the exiting further heated nitrogen is then passed through coil **152** of heat exchanger **150** for cooling that portion of the compressed feed gas mixture passing through coil **148** of the heat exchanger, and the exiting nitrogen is discharged at **250** as waste nitrogen at about atmospheric pressure and close to ambient temperature.

The system described above and illustrated in FIG. 2 is basically the same as that of FIG. 1, but differs in the expansion and partial reduction in pressure of the compressed feed gas mixture and its division into several streams to provide more efficient cooling of the feed gas mixture prior to introduction into the reboiler **24**, and also provides for two-stage reduction in pressure of the portion of liquid methane product at **164** which is employed for providing condensing duty and reflux in the column, so that condensing duty can be provided for the vapor mixture of intermediate composition at **182** in the upper portion of the column, and for condensing a substantially nitrogen-containing vapor withdrawn at **212** from the upper end of the column.

It will be understood in the systems illustrated in FIGS. 1 and 2 above, that work expansion of the warmed overhead nitrogen at **104** for recycling through the heat exchanger **42** can take place in two or more stages, and under these conditions, depending upon the final work expanded pressure of the nitrogen, the final work expander **112** may or may not be required.

In unit **34** the heat exchange passages or constructions **32** for passage of the compressed feed mixture in heat exchange relation along the lower portion of the column **26**, can be in the form of a unitary plate-fin heat exchanger (not shown) arranged in indirect heat exchange relation with channels bearing the liquid-vapor mixture being separated in the column **26**. Such channels may be constructed in a manner of a series of perforated fins, or plates, producing the effect of distillation column trays. This is a known type of heat exchanger arrangement described in International Advances in Cryogenics, volume 10, 1965. A heat exchanger arrangement or construction of this type is also disclosed in the copending application, Ser. No. 572,135, filed Aug. 12, 1966, of James D. Yearout, and which is incorporated herein by reference. Since such heat exchanger arrangements or constructions per se form no part of the present invention, they are not shown herein. Although such a plate-fin type of heat exchanger arrangement is preferably employed, any other suitable form of heat exchanger apparatus can be employed to bring the compressed feed mixture in passages **32** in indirect heat exchange relation with the adjacent contents of the column, preferably so as to effect the above-described nonadiabatic differential distillation in the fractionating column.

It will be understood that the systems described above, including the temperatures and pressures set forth, are only illustrative and are not intended as limitative of the invention.

In FIG. 2, although the condensers **180** and **210** are shown to be separated and external from the column **26'**, if desired, it will be understood that these condensers can be incorporated into the column itself, as in the case of condenser **82** in FIG. 1, at the appropriate positions in the upper portion of the column.

It will be noted also, that in the system of FIG. 2 compressors **230** and **236** for compressing the recycled vapor methane streams **228** and **194** to product pressure, can be operated from a suitable motor, or a portion of such energy can be supplied by suitable connections from expanders **106**, **112** and **124**.

From the foregoing, it is seen that the invention provides a novel method and system for separating the components of a feed mixture containing nitrogen and low-boiling hydrocarbons such as methane, ethane and propane, wherein the feed mixture to be separated is at a pressure higher that the critical pressure of the components, it is desired to recover the hydrocarbon or hydrocarbons at a pressure approximately or close to the feed pressure, such system employing the overhead nitrogen separated in the column for providing sufficient subcooling of compressed liquid feed mixture, and for providing sufficient refrigeration to cool a portion of the liquid hydrocarbon product sufficiently to permit such cooled portion of liquid hydrocarbon product to be employed so as to provide adequate condensing duty for the column, and wherein nonadiabatic differential distillation is applied to provide maximum efficiency. Feed mixtures which can be advantageously separated to provide substantially pure hydrocarbon product at high pressure according to the invention, include compositions containing from about 10 to about 60% nitrogen and about 40 to about 90% methane. As previously noted, and as applied in the systems described above and illustrated in the drawings, the system is particularly advantageous for separating pure methane from natural gas mixtures containing about 85% methane and about 15% nitrogen.

While I have described particular embodiments of my invention for the purpose of illustration, it should be understood that various additional modifications and adaptations thereof may be made within the spirit of the invention, and within the scope of the appended claims.

I claim:

1. A process for the separation of a mixture of gases consisting essentially of a low boiling hydrocarbon and nitrogen, which comprises the steps of cooling a compressed feed mixture of said gases, passing said cooled compressed feed mixture in heat exchange relation with the lower portion of a fractionating column to provide reboil heat thereto by passing said feed mixture transversely through a reboiling zone in the lower portion of the column in heat exchange relation with said lower portion of the column, passing said said compressed feed mixture in heat exchange relation along a substantial length of the lower fractionating portion of said fractionating column, said column operating at a pressure substantially lower than the pressure of said compressed gas mixture, subcooling the exiting compressed feed gas mixture, reducing the pressure of said subcooled compressed mixture approximately to the pressure in said column, introducing said last-mentioned mixture as feed into said fractionating column, effecting a separation of said mixture in said column into a nitrogen fraction and a hydrocarbon fraction, withdrawing nitrogen as overhead from said column, passing said overheat nitrogen in heat exchange relation with said compressed feed mixture for subcooling same as aforesaid, work expanding the exiting nitrogen and further cooling same, withdrawing said low-boiling hydrocarbon in substantially pure liquid form from the lower portion of said column, separating a portion of said liquid hydrocarbon, passing said work expanded nitrogen in heat exchange relation solely externally of said column with said portion of liquid hydrocarbon for subcooling same, throttling said subcooled portion of liquid hydrocarbon further reducing the temperature and pressure thereof, and employing solely said throttled hydrocarbon to provide condensing duty in the upper portion of said column required to effect said separation of said mixture therein.

2. A process as defined in claim **1**, wherein said low-boiling hydrocarbon is methane.

3. A process as defined in claim **1**, wherein said cooled compressed feed gas mixture is throttled to reduced pressure and temperature prior to passage of said feed mixture into heat exchange relation with the lower portion of said column.

4. A process as defined in claim **1**, wherein said overhead nitrogen, following passage thereof in heat exchange relation with said compressed feed mixture for subcooling same, is first work expanded to an intermediate pressure, and said last-mentioned work expanded nitrogen is recycled in heat exchange relation with said compressed feed gas mixture, and the resulting exiting nitrogen is again work expanded to a pressure close to atmospheric pressure for said further cooling of said nitrogen prior to passage of said expanded nitrogen in heat exchange relation with said liquid hydrocarbon.

5. A process as defined in claim **1**, wherein said subcooled portion of liquid hydrocarbon is throttled to a reduced pressure such as to provide a sufficiently low temperature of said throttled hydrocarbon to permit same to condense nitrogen vapors in the upper portion of said column, and including passing said throttled liquid hydrocarbon through a condensing zone in the top of said column in heat exchange relation with the nitrogen at the top of the column for condensing same and providing reflux in the column, and passing the exiting vaporized hydrocarbon in heat exchange relation with said separated portion of compressed liquid hydrocarbon for subcooling same.

6. A process as defined in claim **1**, including dividing said subcooled portion of compressed liquid hydrocarbon into a first and second stream, throttling said first stream of compressed liquid hydrocarbon to an intermediate pressure and a reduced temperature sufficient to condense vapor contents in an intermediate portion of the upper section of said column, passing said throttled first stream of liquid hydrocarbon into heat exchange relation with said intermediate vapor contents of said column, for condensing same, further subcooling said subcooled second stream of compressed liquid hydrocarbon, throttling said further subcooled second stream of compressed liquid hydrocarbon to a reduced pressure and a reduced temperature sufficiently low to condense nitrogen vapors in the upper end of said column, passing said throttled second stream of liquid hydrocarbon into heat exchange relation with said nitrogen vapors to condense said nitrogen vapors, thereby providing condensing duty and reflux in the upper portion of said column, and passing the vaporized first and second hydrocarbon streams into heat exchange relation with said separated portion of compressed liquid tion with said separated portion of compressed liquid hydrocarbon for subcooling same as aforesaid.

7. A process as defined in claim **1**, wherein said feed gas mixture consists essentially of about 85% methane and about 15% nitrogen, said nitrogen-methane feed mixture being initially compressed to about 1,000 p.s.i., and which includes delivering the methane withdrawn from the lower portion of the column as product at pressure of about 800 p.s.i.

8. A process as defined in claim **1**, wherein said low-boiling hydrocarbon is methane, and including passing the expanded nitrogen exiting from heat exchange relation with said liquid methane, in heat exchange relation with said compressed feed gas mixture, pumping the major portion of liquid methane withdrawn from the lower portion of the column up to desired delivery pressure, and passing said pumped liquid hydrocarbon in heat exchange relation with said feed gas mixture for cooling same, passing the remaining minor portion of throttled methane exiting from said condensing duty, into heat exchange relation with said feed gas mixture for cooling same.

9. A process as defined in claim **1**, wherein said low-boiling hydrocarbon is methane, and wherein said overhead nitrogen, following passage thereof in heat exchange relation with said compressed feed mixture for subcooling same, is first work expanded to an intermediate pressure, and said last-mentioned work expanded nitrogen is recycled in heat exchange relation with said compressed feed gas mixture, and the resulting exiting nitrogen is again work expanded to a pressure close to atmospheric pressure for said further cooling of said nitrogen prior to passage of said expanded nitrogen in heat exchange relation with said portion of liquid methane, and wherein said subcooled portion of liquid methane is throttled to a reduced pressure such as to provide a sufficiently low temperature of said throttled methane to permit same to condense nitrogen vapors in the upper portion of said column, and including passing solely said throttled liquid methane through a condensing zone in the top of said column in heat exchange relation with the nitrogen at the top of the column for condensing same and providing reflux in the column, and passing the exiting vaporized methane in heat exchange relation with said separated portion of compressed liquid methane for subcooling same.

10. A process as defined in claim **1**, including work expanding said feed gas mixture to an intermediate pressure, and dividing said expanded feed gas into a first and second portion, throttling said first portion and passing same in heat exchange relation with said work expanded nitrogen for cooling said first portion of said feed mixture, passing said second portion of work expanded feed mixture into heat exchange relation with said work expanded nitrogen and with liquid hydrocarbon withdrawn from the lower portion of said column, for cooling said second portion of compressed feed mixture, throttling said exiting and cooled second portion of feed mixture approximately to the pressure of said cooled throttled first portion of feed mixture, and combining said cooled and throttled first and second portions of feed mixture prior to passage of said feed mixture in heat exchange relation with the lower portion of said column.

11. A process as defined in claim **10**, wherein said throttled hydrocarbon exiting from said condensing duty is passed in heat exchange relation with said first throttled portion of feed mixture for cooling same.

12. A system for the separation of the components of a mixture of gases, which comprises a fractionating column, means for cooling a compressed feed gas mixture, a reboiler in the lower portion of said column, means for conducting said cooled compressed feed gas mixture transversely through said reboiler, passage means along a substantial length of the lower portion of said column, means for conducting the exiting compressed feed gas mixture from said reboiler into said passage means, first subcooling means for subcooling a compressed feed mixture withdrawn from said passage means, means for reducing the pressure of said subcooled gas mixture, means for introducing the resulting gas mixture as feed in liquid form into the fractionating column intermediate the ends thereof, and to effect a separation of said mixture in said column, into a fluid at the upper end of said column and a substantially pure liquid at the bottom of said column, means for conducting fluid from the upper end of said fractionating column into said first subcooling means in heat exchange relation with compressed feed gas for subcooling same, means for work expanding said fluid exiting said subcooling means, means for withdrawing liquid product from the bottom of said fractionating column, means for separating a portion of said last-mentioned liquid, second subcooling means for passing said work expanded fluid in heat exchange relation externally of said column with said separated portion of liquid product withdrawn from the bottom of said column, for subcooling same, means for throttling said subcooled portion of liquid exiting said last-mentioned subcooling means, and means for bringing solely said throttled portion of liquid in heat exchange relation with contents of the upper portion of said column to provide condensing duty therein and reflux for said column required to effect said separation of the feed gas mixture therein.

13. A system as defined in claim **12**, said work expanding means for said fluid including means for work expanding said fluid exiting said first subcooling means, to an intermediate pressure, means for recycling said work expanded fluid of intermediate pressure through said first subcooling means in heat exchange relation with said compressed feed gas mixture, and means for work expanding the exiting fluid of intermediate pressure, to a further reduced pressure and temperature prior to passage of said work expanded fluid in heat exchange relation with said separated portion of liquid product.

14. A system as defined in claim **12**, wherein said last-mentioned means includes a condenser associated with the upper portion of said column, means for passing said throttled portion of liquid in heat exchange relation in said condenser with vapor contents of the upper portion of said column for condensing said vapors, and means for passing exiting vaporized fluid from heat exchange relation with said column contents in said condenser, into said subcooling means for subcooling said separated portion of liquid product.

15. A system as defined in claim **12**, said last-mentioned means including a first and second condenser, said first condenser being associated with an intermediate section in the upper portion of said column, and said second condenser being associated with the upper end of said column, means communicating said first condenser with vapor mixture from said intermediate section of said column, and means communicating said second condenser with vapors from the upper end of said column, means for throttling a first portion of liquid product exiting said second subcooling means to an intermediate pressure and means for passing said last-mentioned throttled liquid into heat exchange relation with the contents of said first condenser for condensing same and vaporizing said liquid, means for separating a second portion of the exiting liquid from said second subcooling means, a third subcooling means for further cooling said last-mentioned second portion of liquid, means for throttling said further cooled second portion of liquid to a substantially reduced pressure, means for conducting said last-mentioned throttled liquid in heat exchange relation with the contents of said second condenser for condensing same, and vaporizing said liquid, and means for passing the vaporized liquid exiting said first and second condensers through said second and third subcooling means in heat exchange relation with compressed liquid product for subcooling same.

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