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LIQUEFACTION OF NATURAL GAS BY CASCADE REFRIGERATION
AND MULTIPLE EXPANSION

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2 Sheets-Sheet 1

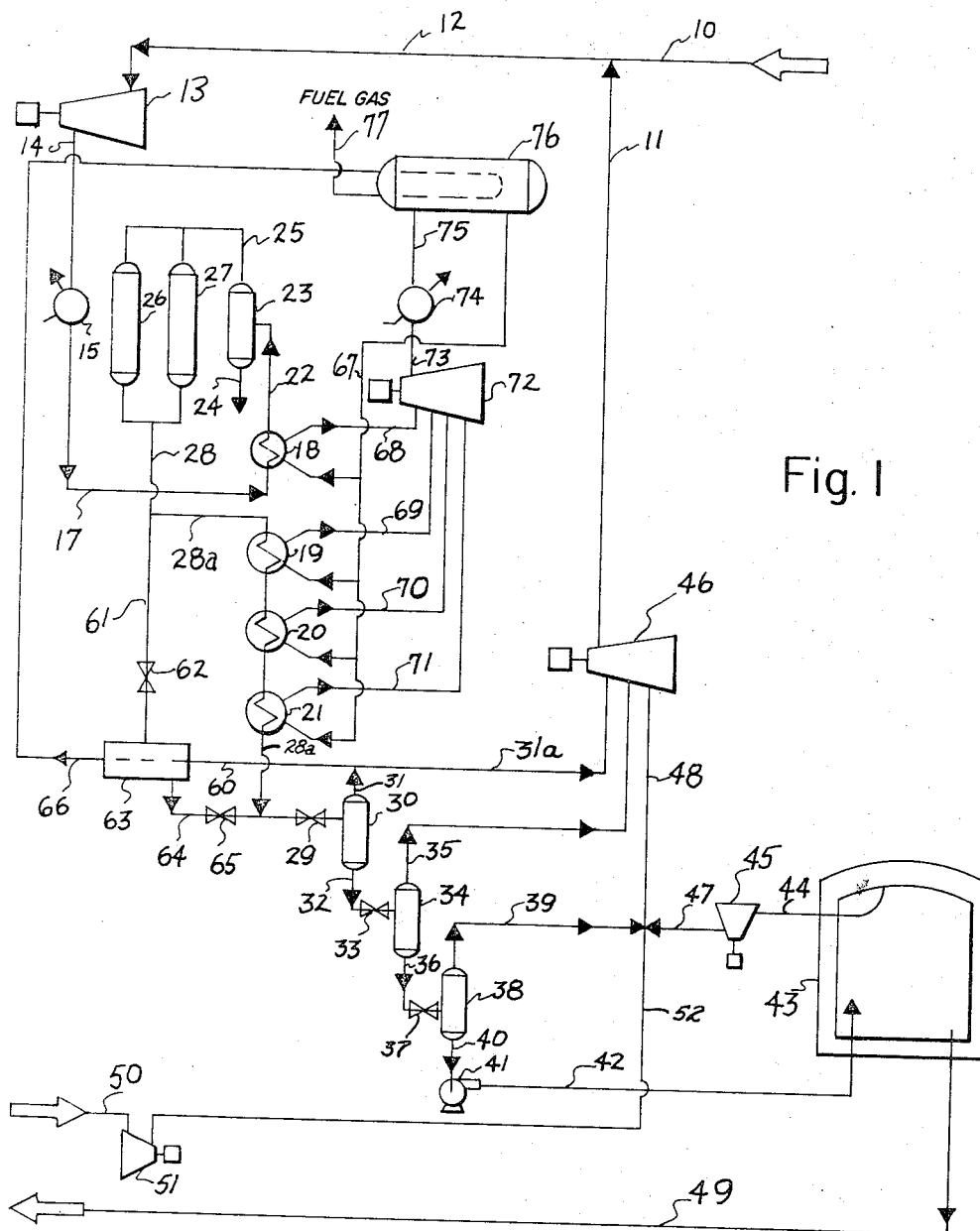


Fig. 1

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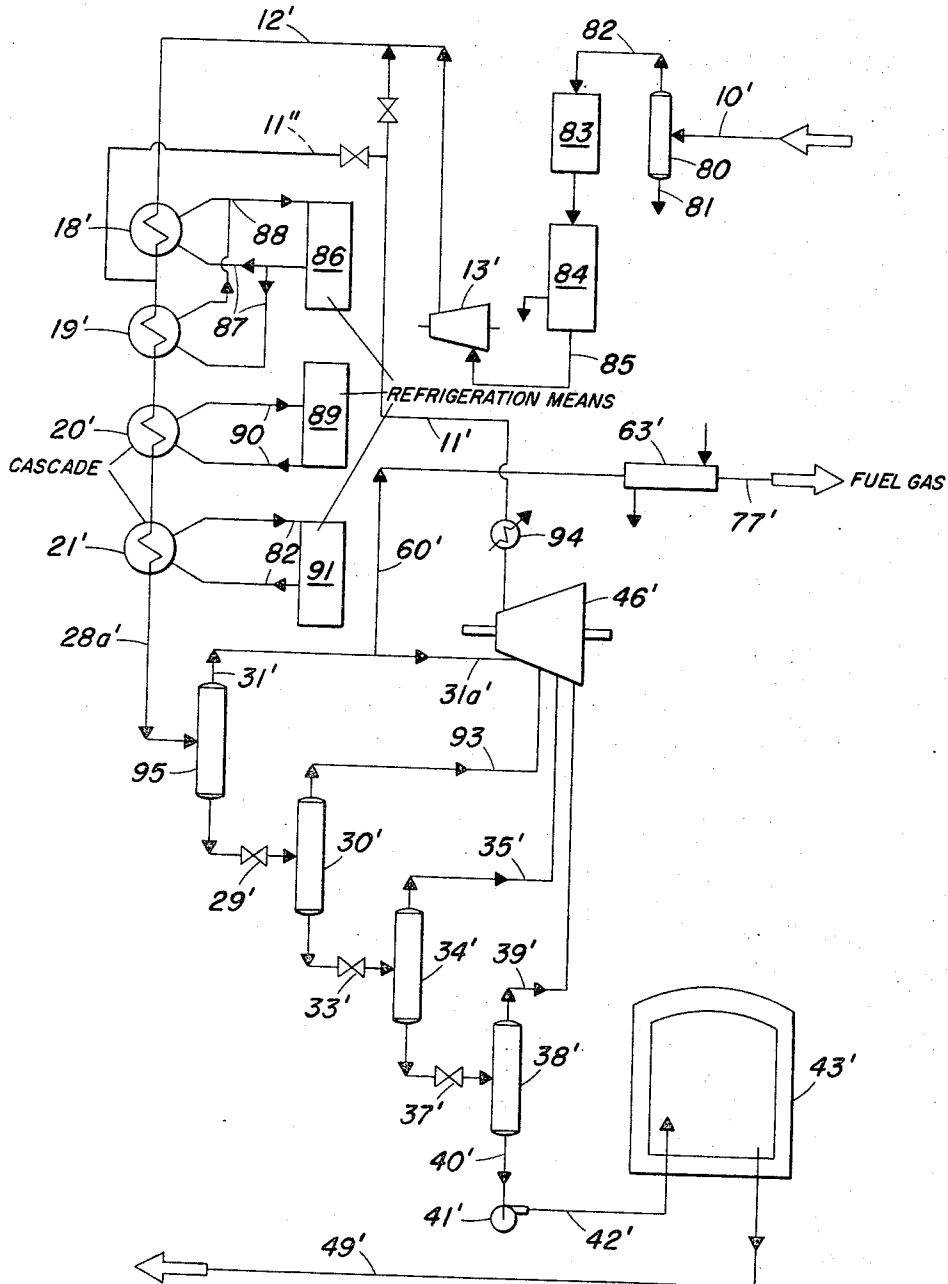


Fig. 2.

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LIQUEFACTION OF NATURAL GAS BY CASCADE REFRIGERATION AND MULTIPLE EXPANSION
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1 Claim. (Cl. 62—23)

The present application constitutes a continuation-in-part of U.S. patent application Ser. No. 358,789, filed Apr. 10, 1964, now abandoned.

This invention relates to a process for the liquefaction of a gas, and more particularly relates to a process and apparatus for the liquefaction of natural gas primarily comprised of methane, and including heavier hydrocarbons such as ethane, propane, butane and the like.

There are many reasons for reducing natural gas to a liquefied state. One of the main reasons for liquefying natural gas is the resultant reduction of the volume of the gas to about $\frac{1}{600}$ of the volume of natural gas in the gaseous state. Such a reduction in volume permits the storage and transportation of liquefied natural gas in containers of more economical and practical design. Additionally, it is desirable to maintain a supply of gas through peak demand periods whereby such periods can be met by liquefied gas held in storage. Another important reason is the transportation of liquefied natural gas from a source of plentiful supply to a distant market where the source and supply may not be efficaciously joined by pipe lines and transportation in the gaseous state would be uneconomical.

The object of this invention is to provide an apparatus and method for the economical and efficient liquefaction of a gas, particularly natural gas, for storage and transportation.

Another object of this invention is to provide a process for the liquefaction of natural gas wherein the natural gas is compressed to a high pressure, preferably above the supercritical pressure, and passed in heat exchange relationship through a refrigeration system, to remove, at relatively high temperatures, the heat as sensible heat, which would be, at lower pressures, latent heat at lower temperatures.

A further object of my invention is to provide a process for liquefying natural gas wherein the gas is compressed to a high pressure, preferably above the supercritical pressures, cooled in a refrigeration system having a single refrigerant to conveniently remove at relatively high temperatures the sensible heat of the natural gas, which would at lower pressures result as latent heat at lower temperature, and thereafter expanding the thus cooled natural gas in a series of stages to produce liquefied natural gas at atmospheric pressure and at a temperature of about -258°F .

A still further object of my invention is to immediately compress the vapors removed from such expansion stages and combine such vapors with the feed introduced into the initial compressor wherein the pressure of the gas is raised to a pressure of from about 600 p.s.i.g. to about 1.3 times the critical pressure thereof.

Still another object of my invention is to provide a method and apparatus for the liquefaction of natural gas wherein the quantity of nitrogen in the process is maintained at a low level, and the liquefied product is substantially free of nitrogen.

Other objects and fuller understanding of my invention may be had by referring to the following description taken in conjunction with the accompanying drawings, in which,

FIGURE 1 is a simplified, schematic flow diagram of a first embodiment of the invention;

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FIGURE 2 is a simplified, schematic flow diagram of a second embodiment of the invention.

The natural gas to be treated in accordance with my invention will be a gas from which a part of the moisture and acid gases, such as carbon dioxide, hydrogen sulfide and the like have been removed in a manner familiar to those skilled in the art. In one embodiment of the invention, the natural gas after such treatment, and a recycle gas stream as hereinafter described, is introduced into a feed gas compressor wherein the pressure of the combined stream is increased to a pressure of from about 600 p.s.i.g. to about 1.3 times the critical pressure of the gas. The compressed gas is thereafter passed through a cooler and a first heat exchanger of a refrigeration system to remove any condensable heavier hydrocarbons. The cooled gas is then passed through the remaining heat exchangers of such refrigeration system to refrigerate the gas to a temperature slightly above the normal boiling point of the refrigerant. In the case of ethane, as the refrigerant, the temperature would be about -125°F . The refrigerant preferred is ethane since it may be supplied by the natural gas. Of course, other refrigerants can be used provided they may be condensed with a cooling medium at a temperature of about 60°F . or less. Only one refrigerant is utilized in the refrigeration system. While a portion of the water has been removed in the gas prior to compression, additional moisture will be contained in the natural gas after cooling, and accordingly, is conveniently removed between the first and second heat exchangers of such refrigeration system.

The natural gas at a temperature slightly above the normal boiling point of the refrigerant is thereafter expanded into a first flash drum wherein a substantial portion of the gas is liquefied during the expansion. The gaseous overhead from the first flash drum, including nitrogen, is withdrawn and a portion thereof withdrawn from the process as fuel gas, after passing such portion through a heat exchanger to recuperate the cold therefrom. The liquefied natural gas from the first flash drum is thereafter successively expanded over a series of flash drums wherein the pressure of the liquefied natural gas is eventually reduced to atmospheric pressure. The liquefied natural gas is withdrawn from the last flash drum and passed to storage and transportation.

The flash gases from each of the flash drums, except the last flash drum, are introduced into intermediate stages of a methane recycle compressor and thereafter combined with the natural gas feed. Such combined gas stream is passed to the feed gas compressor wherein the pressure of the gas is raised to a pressure within the aforementioned pressure range. There is a light reduction in pressure of the liquefied natural gas from the last flash drum into the storage tank, and consequently a portion of the liquefied natural gas will vaporize in the storage tank. Such vaporized portion of the natural gas is also passed to a methane recycle compressor after passing through a centrifugal booster compressor.

In a second embodiment of the invention, natural gas at a pressure above or below the critical pressure, but preferably above that pressure, is initially freed of entrained condensate, acid components and water vapor in the manner well known to the art. Depending on the pressure at which the gas is delivered, it may be further compressed for use in the invention, as described hereinabove, and the recycle gas is then added to the stream. The stream is passed through a plurality of heat exchangers which are controlled to bring about substantially complete liquefaction of the gas.

It is to be noted that the recycle stream, depending on its temperature, may advantageously be added to the main stream after the latter stream has passed through

the first of the heat exchangers, the object being to add the recycle stream at the point where the temperatures of both streams most nearly coincide.

The condensed liquid, still at a high pressure but now below the critical pressure, is passed to a receiver at a lower pressure and a gaseous fraction removed. This fraction is utilized as fuel gas after recovering the cold potential therefrom, as in the first embodiment. Any remaining gas is passed to a methane recompressor.

The temperature of the liquid is then lowered in successive expansion stages, the gases being recompressed as in the first embodiment and recycled as indicated above. The liquefied natural gas is then pumped to storage at about atmospheric pressure.

By bringing about the liquefaction of the gas in the heat exchangers, rather than by expansion as in the first embodiment, the use of more than one refrigerant, or at least more than one refrigeration system, becomes advantageous. Thus, in the second embodiment, it is preferred that the first two heat exchangers employ a cooling medium in one cycle, and the last two heat exchangers, wherein liquefaction occurs, employ a second refrigeration medium, due to the larger cooling load imposed thereon. The heat exchangers may conveniently be arranged in cascade fashion.

Referring to FIGURE 1, which is a schematic flow diagram of an embodiment of the invention, the following describes this embodiment applied to the liquefaction of natural gas. It is understood, however, that the invention is also applicable to the liquefaction of other gases containing hydrocarbons, such as refinery gases and the like.

Lean natural gas, primarily comprised of methane, in line 10, is combined with a recycled gaseous stream, as more fully hereinafter described, in line 11, and passed through line 12 to a feed gas compressor 13. The natural gas is compressed to a pressure of from 600 p.s.i.g. to about 1.3 times the critical pressure of the gas stream. The compressed gas is passed through line 14 to cooler 15 and thence through line 17 to a heat exchanger 18 of a refrigeration system including heat exchangers 18, 19, 20 and 21. The cooled gas is then passed through line 22 into a separator 23. In separator 23, any heavier hydrocarbons which may condense during passage through cooler 15 and heat exchanger 18 are withdrawn from separator 23 through line 24 and the compressed natural gas withdrawn through line 25 and passed to dryers 26 and 27 to remove residual amounts of moisture in the gas. The dryers 27 and 28 are operated in an alternate manner which is well known to those skilled in the art.

The dried gas in line 28 is split and a portion in line 28a passed through the remaining heat exchangers 19, 20 and 21 of the refrigeration system wherein the gas is cooled by a refrigerant which is expanded into the heat exchangers of the refrigeration system.

The refrigeration system utilizes a single refrigerant as will be more fully hereinafter described. An important aspect of my invention is that the natural gas passing through the refrigeration system is cooled, without liquefaction, to a temperature slightly above the normal boiling point of the refrigerant providing the cooling requirements for the refrigeration system. The gas is readily cooled at such high pressures by using a single refrigerant whereby the heat is conveniently removed at relatively higher temperatures as sensible heat, which would, at lower pressures, show up as latent heat at a lower temperature.

The natural gas in line 28a together with another gaseous stream as more fully hereinafter described, is expanded across valve 29 to a lower pressure, such that a substantial portion of the natural gas is liquefied during such expansion and is thereupon introduced into flash drum 30. The liquefied gas in flash drum 30 is withdrawn through line 32, expanded across valve 33 and introduced into a second flash drum 34 whereby the temperature of

the liquefied gas is further reduced. A gaseous phase in flash drum 34 is withdrawn through line 35 while the further cooled liquefied gas is withdrawn through line 36. The liquefied gas in line 36 is thereafter further expanded across valve 37 and introduced into flash drum 38 wherein the liquefied gas is still further cooled. The expansion of the gas in line 28a and the liquefied gas in lines 32 and 36 are to lower pressures, such that the pressure of the liquefied gas in the last flash drum 38 is substantially at atmospheric pressure.

A gaseous phase is withdrawn from flash drum 38 through line 39 while the liquefied natural gas at about atmospheric pressure is withdrawn through line 40. The liquefied natural gas in line 40 is passed by pump 41 through line 42 and introduced into a storage tank, generally indicated as 43, maintained at atmospheric pressure. Since the storage tank 43 is maintained at about atmospheric pressure, the temperature of the liquefied gas about -257° F. If there is a small reduction in the pressure of the liquefied gas during passage to the storage tank 43 from flash drum 38, additional cooling of the liquefied natural gas occurs thereby vaporizing a portion of the liquefied gas. The vapors formed during such additional expansion are withdrawn from the storage tank 43 through line 44, and compressed in a centrifugal compressor 45.

As a distinct feature of my invention, I contemplate taking the flashed gases from the flash drums and the storage tank and compressing such vapors in a recycle compressor 46 and combining such compressed stream with the natural gas feed to the plant. The gaseous phases in lines 31a and 35 are passed to an intermediate stage of the methane compressor 46. The gaseous phase in line 39 is combined with the compressed gas stream in line 47 and passed via line 48 to an initial stage of the methane compressor 46.

Loading the warm tanks of a vessel with the liquefied natural gas from storage tank 43 through line 49 will effect vaporization of a small portion of the liquefied natural gas which is then returned to the plant through line 50, compressed in dockside centrifugal compressor 51 and is combined with the gas in line 48 through line 52 for compression in compressor 46. The now compressed gas is withdrawn from compressor 49 through line 11 to be combined with the natural gas feed to the plant in line 10.

Most natural gases contain nitrogen as a contaminant. It is desirable in the course of liquefaction to remove as much as possible of the nitrogen from the gas since nitrogen in the liquefied natural gas reduces the value thereof, particularly when carrying the gas in bulk transportation. In accordance with equilibrium conditions in flash drum 30, the gaseous phase in line 31 contains a higher percentage of the nitrogen than the liquid phase in line 32, introduced into the process. A portion of such gaseous phase in line 31 is withdrawn through line 60 and is utilized as a fuel gas in compressor drives and other equipment of the plant.

Since the gas in line 60 is at a relatively low temperature, it is desirable to recover the cold potential thereof prior to using such gas as a fuel gas. To recover such potential, a portion of the compressed gas in line 28 is passed through line 61 under the control of valve 62 to heat exchangers 63 and passed in heat exchange relation with the gas in line 60. Once equilibrium has been reached, the amount of nitrogen introduced into the process with the natural gas feed will equal the amount of nitrogen in the fuel gas in line 60, and the liquefied natural gas in storage tank 43. The cooled compressed gas is withdrawn from heat exchanger 63 through line 64 under control valve 65 and combined with the gas leaving heat exchanger 21 prior to expansion through valve 29. The gas in line 60 after passage through heat exchangers 63 is withdrawn through line 66.

As a distinct advantage of my system as compared to prior processes, I am able to utilize one refrigerant, such as ethane, to provide the cooling requirements for refrigeration systems. Liquefied ethane in line 67 is expanded stagewise into heat exchangers 18, 19, 20 and 21 to remove the sensible heat of the compressed gas in line 28. The thus expanded ethane is withdrawn from heat exchangers 18, 19, 20 and 21 through lines 68, 69, 70 and 71, respectively, and is passed to an ethane compressor 72. The gaseous ethane in line 68, 69, and 70 are introduced into intermediate stages of the compressor 72, whereas the gaseous ethane in line 71 is introduced into a first stage thereof. The compressed ethane is passed through line 73 to heat exchangers 74 wherein the ethane is cooled with a cooling medium having a temperature less than about 60° F., whereby the compressed ethane is condensed into the liquid phase. The liquefied ethane is passed through line 75 to ethane receiver 76. The fuel gas in line 66 may be passed through ethane receiver 76 to further recover the cold potential of the fuel gas stream and is subsequently passed through line 77 to the points of use (not shown).

In FIGURE 2 of the drawings, a second embodiment of the invention is shown, wherein process units or lines performing the same or similar functions as the embodiment of FIGURE 1 are designated by prime numerals. Again, the illustration is for liquefaction of natural gas, it being understood that other hydrocarbon gases may be treated similarly.

With reference to FIGURE 2, natural gas in line 10' is delivered at a pressure either above or below the critical pressure of the particular gas composition, and is freed from entrained condensate in drum 80, condensate being removed via line 81. The gas passes in line 82 into suitable apparatus 83, 84 for the removal of acid gas and water vapor, respectively. The cleansed gas, in line 85, is compressed to the desired pressure in compressor 13' and then passes in line 12' to the first heat exchanger 18', being mixed with the recycle gas stream in line 11' either before or after passage through heat exchanger 18', as set forth in detail hereinbelow.

Depending on the gas pressure in line 10', it may not be necessary to employ compressor 13'. It is to be noted that with the refrigeration and liquefaction system preferred for use with this embodiment, it is not as important that the gas be compressed well above its critical pressure, so in many cases it will be expedient to eliminate compressor 13'.

While FIGURE 2 shows four heat exchangers 18', 19', 20' and 21' as in FIGURE 1, the refrigeration means employed are necessarily somewhat different. Thus, as shown in FIGURE 2, a first refrigeration means 86 provides the cooling medium for exchangers 18' and 19', through lines 87 and 88; typically, propylene may be used as the refrigerant at this stage of cooling. Exchangers 20' and 21' are supplied with cooling mediums by refrigeration units 89 and 91, arranged in cascade fashion; these units may use propylene and ethylene, for example. As shown, ethylene would be the preferred coolant. Alternatively, heat exchangers 18', 19' and 20' may be supplied with refrigerants connected in cascade fashion with exchanger 21' supplied independently. The important feature is that the effluent from exchanger 20' be cooled close to or below the condensation temperature of the gas at the prevailing pressure, and that heat exchanger 21' extract the heat of liquefaction so that effluent in the line 28a' is completely condensed. As will be obvious to one skilled in the art, for the case of natural gas the refrigerant employed in exchanger 21' should have a normal boiling point between -110° F. and -155° F. so that the condensation terminates between about -90° F. and -155° F. and at a pressure below the critical pressure for the gas composition being treated.

The liquefied gas is conveyed through line 28a' to receiver 95, wherein a vapor stream 31', of such volume

that it contains the major portion of impurities such as nitrogen or helium contained in the raw gas, is removed.

Depending on the need for fuel gas and composition of vapor stream 31', either all or a part of it is taken off through line 60', cold recuperative exchange 63', and removed through line 77', as in the embodiment of FIGURE 1. Any remaining gas is passed to the intermediate stage of compressor 46' via line 31a'.

The liquid stream from receiving drum 95 is successively flashed to lower pressure through expansion valves 29', 33' and 37', and expansion drums 30', 34' and 38', effluent liquefied natural gas in line 40' being near atmospheric pressure and at near the normal bubble point temperature. As in the embodiment of FIGURE 1, the effluent is pumped by pump 41' into line 42' and conventional storage reservoir 43', from where it may be withdrawn in line 49' for shipment or usage.

Vapor streams 35', 39' and 93 from each of the flash drums 38', 34', 30' enter appropriate pressure stages of recycle compressor 46', wherein they are compressed to just slightly above the pressure maintained in heat exchangers 18'-21'. The discharge from compressor 46' is cooled, if necessary, in heat exchanger 94, wherein a heat exchange medium at ambient temperature is employed. The gas, now in line 11', is joined with the natural gas in line 12' or after any cooler where the temperature of the two streams most nearly correspond to each other. One such alternative arrangement is shown by dotted line 11''.

It is to be noted that the vapor streams 35', 39' and 93 are fed to compressor 46' directly without any heat exchange or other preheating.

The following is an example of my invention pertaining to a specific natural gas wherein the quantity flow in pounds actually represents the ratio of flow in pounds per pound of LNG withdrawn from the process. Such natural gas has the following analysis:

Methane	99.5
Ethane	.1
Nitrogen	.4

With reference to FIGURE 1, 1.119 pounds of natural gas at a temperature of 60° F. in line 10 after being treated to remove acid gases and moisture is combined with 1.2693 pounds of recycled gas at a temperature of -15° F. in line 11. The combined gas stream at a temperature of 16° F. and at a pressure of 600 p.s.i.a. is compressed to 1400 p.s.i.a. in compressor 15 and cooled to a temperature of 55° F. in heat exchanger 15 during passage against a cooling medium at a temperature of 50° F. The compressed gas is further cooled in heat exchanger 18 to a temperature of 5° F. and passed through dryers 26 and 27 wherein residual moisture is removed.

The compressed gas is thereafter serially passed through heat exchangers 19, 20 and 21 and cooled to a temperature of -115° F. The gas withdrawn from heat exchanger 21 is at a pressure of 1360 p.s.i.a. as a result of a pressure drop of 40 p.s.i.a. through the aforementioned units. The gas is then expanded across expansion valve 29 into flash drum 30 to a pressure of 180 p.s.i.a. with a resulting decrease in temperature of -184° F. 1.4115 pounds of liquefied gas is withdrawn from flash drum 30 and expanded across valve 33 to a pressure of 55 p.s.i.a. and introduced into flash drum 34. 1.1458 pounds of liquefied gas now at a temperature of -226° F. is then withdrawn from flash drum 34 and expanded across valve 37 to a pressure of 20 p.s.i.a. and introduced into flash drum 38 wherein the liquefied gas is further cooled to a temperature of -252° F. The liquefied gas in flash drum 38 is withdrawn through line 40 by pump 41 and passed to storage tank 43 maintained at 14.7 p.s.i.a. The additional slight expansion from flash drum 38 and storage tank 43 effects further cooling of the liquefied gas to a temperature of -257° F.

0.8578, 0.2657 and 0.1176 pound of a gaseous phase in lines 31a, 35 and 39 are passed together with 0.0282 pound of gas withdrawn from storage tank 43, to methane recycle compressor 46 and compressed to a pressure of 600 p.s.i.a. and subsequently combined with the feed natural gas. 0.119 pound of flashed gas from flash drum 30 at a temperature of -184° F. is passed through heat exchanger 63 and ethane receiver 76 and withdrawn from the process as fuel gas in line 77. It is understood to those skilled in the art that the above example is with respect to the natural gas of the aforementioned composition and that the temperatures and quantities will vary with the composition of the natural gas.

Gaseous ethane at a temperature of 0° F., -40° F., -80° F. and -120° F. is withdrawn from heat exchangers 18, 19, 20 and 21 through lines 68, 69, 70 and 71 respectively, and compressed to 560 p.s.i.a. in compressor 72 and cooled in heat exchanger 74 to a temperature of 70° F. in heat exchange relation with a cooling medium in and the following claim.

While I have shown and described a preferred embodiment of my invention, I am aware that variations may be made thereto, and therefore, desire a broad interpretation of my invention within the scope of the disclosure herein and the following claim.

What is claimed is:

A method for liquefying a natural gas at elevated pressures comprising:

- (a) cooling the gas in a plurality of stages by expanding a refrigerant in an indirect heat transfer relation with the gas, liquefaction of the gas being effected in the last stage;
- (b) expanding the liquefied gas in a plurality of stages to atmospheric pressure; a gaseous fraction being produced in the course of said expansion;
- (c) compressing a gaseous fraction from each stage and recycling the compressed gaseous fraction to

said cooling stages, the compressed gas fractions being recycled to and admixed with the natural gas at the cooling stage having a temperature most nearly corresponding to the temperature of the compressed gas fractions; said recycled gas and natural gas being liquefied in admixture with each other in passing through the cooling stages;

- (d) separating a portion of a gas fraction removed from the first expansion stage and passing said portion of gas fraction to fuel gas consumption and
- (e) in the said plural stages of refrigeration, providing a cooling close to the condensation temperature of the gas at the prevailing pressure and a subsequent cooling to extract the heat of liquefaction, respectively, in the order of natural gas flow, supplying the said cooling close to the condensation temperature with a refrigerant connected in cascade fashion with the refrigerant supplying cooling to extract the heat of liquefaction.

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