

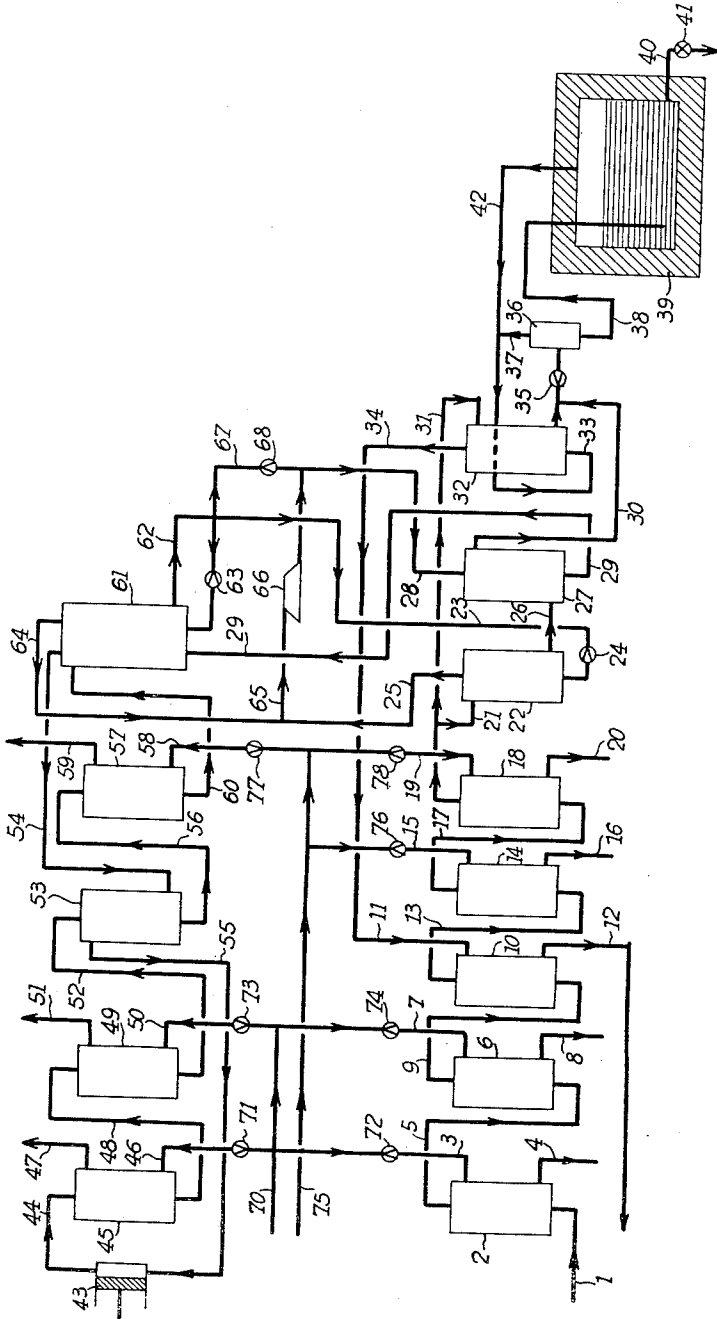
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PROCESS FOR THE LIQUEFACTION OF A GAS

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PROCESS FOR THE LIQUEFACTION OF A GAS
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The present invention relates to a process for the liquefaction of a gas, more especially of methane or of gases having a high methane content, for the purpose of its storage and its transport in the liquid state under low pressure.

As is known, the conveyance of gases from the place at which they are collected or manufactured to the places at which they are used through pipelines or in cylinders under high pressure becomes costly when the distance to be covered is fairly great. An economic method proposed for such conveyance consists in liquefying the gas at its source, in conveying it in the liquid state in vehicles or on ships provided with thermally insulated containers and then in vapourising it at its arrival at the place of use.

It has already been proposed to effect the liquefaction of gases which are somewhat difficult to liquefy, such as methane and even hydrogen and helium, by first using one or more auxiliary cooling fluids in accordance with the so-called "cascade" method, and then effecting a final cooling of a fraction of the gas to be liquefied by expansion with external work. The gas is thus cooled under a relatively high pressure by heat exchange with at least one cooling fluid, an auxiliary fraction of this gas is expanded with external work, and the gas is re-cooled under high pressure by heat exchange with the expanded auxiliary fraction, whereafter the gas is expanded to its storage pressure and the liquid obtained is collected.

The invention consists in an improvement in the aforesaid method, by means of which it is possible to obtain a high production of cold by the expansion with external work and a final very intensive cooling of the gas under pressure, so that the quantity of gas remaining in the gaseous state after the expansion to storage pressure remains small.

This improvement also makes it possible to effect the liquefaction of the gas with a relatively small expenditure of energy and high regularity of operation.

The process of the invention is characterised in that at least a part of the auxiliary fraction of the gas is obtained by expansion of a part of the cooled gas to a pressure intermediate between its high initial pressure and its storage pressure, followed by warming up of this part in heat exchange with the gas cooled under high pressure.

A particularly advantageous application of this process resides in the liquefaction of methane or of gases of high methane content, such as natural gas. In this case, the methane under relatively high pressure is liquefied by heat exchange with a cooling fluid. The liquid methane is sub-cooled by heat exchange with the expanded auxiliary fraction with external work before being expanded to the storage pressure. The auxiliary gas fraction is evaporated and then warmed up in heat exchange with the liquid methane under high pressure before reaching the expansion machine. The expenditure of energy in relation to the quantity of liquefied methane is relatively small.

The gas to be liquefied, more especially natural gas, generally contains impurities of low volatility, such as C_2 and C_3 hydrocarbons (ethane and propane), which might condense in the expansion machine and damage it or at least interfere with its operation.

In accordance with one modified embodiment of the invention, this disadvantage is avoided by effecting the ex-

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pansion with external work on gas freed from its impurities of low volatility, which flows through a closed cycle and performs the function of an additional cooling fluid. The expanded purified gas is thereafter used to complete the cooling of the impure gas which has previously been liquefied by heat exchange with one or more other cooling fluids, by indirect heat exchange with the latter, and it is then warmed up, re-compressed and returned into the cycle.

The methane to be liquefied is often available under variable pressure and with a variable content of more-volatile impurities (nitrogen) or less-volatile impurities (hydrocarbons). It is therefore necessary constantly to adapt the production of cold in the liquefying installation to these variations. In accordance with one embodiment of the invention, this result is obtained by liquefying and cooling the methane to a substantially fixed temperature level, approximately $-100^\circ C.$, by successive heat exchanges with cooling fluids, and then cooling from $-100^\circ C.$ to a temperature in the neighbourhood of the storage temperature under low pressure with the aid of the cold produced by the expansion of a methane fraction with external work. The cold production is thus readily adjusted as a function of the requirements by controlling the rate of flow of the fraction passing through the expansion machine.

In addition, in accordance with a further variant of the invention, it is possible to cool the methane expanded with external work to a temperature level even lower than the temperature at which it leaves the expansion machine by adding thereto a fraction of liquid methane which evaporates in contact therewith.

The cooling fluids which are preferred for the cooling of the methane are propane or propylene, and ethane or ethylene. However, there may be employed instead of these or simultaneously therewith other cooling fluids such as ammonia, fluorinated hydrocarbons known under the name "Freons," carbon dioxide, etc.

There will hereinafter be described by way of non-limiting example with reference to the accompanying drawing an installation for the liquefaction of natural gas employing the process of the invention. This installation is suitable more especially for the liquefaction of natural gas at high rates of flow greater than 10,000 cubic metres per day, measured under normal conditions.

The natural gas to be liquefied has approximately the following average composition:

	Percent
Methane	88.2
Nitrogen	0.8
C_2 hydrocarbons (ethane)	9.5
C_3 hydrocarbons (propane)	1.5

It is available at $21^\circ C.$ under an absolute pressure of 41.8 kg./cm.². This gas is passed through the duct 1 to the heat exchangers 2 and 6, in which it is successively cooled in counter-current by liquid propane in the course of evaporation under pressures of approximately 4.5 kg./cm.² and 1 kg./cm.² respectively, which arrives through the ducts 3 and 7 and is discharged in vaporous form through the ducts 4 and 8. Having thus been cooled to about $-32^\circ C.$, the natural gas undergoes an auxiliary cooling in a heat exchanger 10 with the aid of methane vapours under low pressure, which arrive through 11 and are discharged at 12. It then passes successively through the ducts 13 and 17 into the heat exchangers 14 and 18, in which it is liquefied in counter-current with liquid ethylene undergoing evaporation under pressures of about 4.5 kg./cm.² and 1 kg./cm.² respectively, which arrives through the ducts 15 and 19 and is discharged through the ducts 16 and 20. The liquefied natural gas is then divided into two fractions. The first fraction, which is the

larger, passes through the ducts 21 and 26 respectively into the heat exchangers 22 and 27, through which it passes in counter-current with two pure methane fractions undergoing evaporation, one in the liquid state under a pressure of about 6 kg./cm.², and the other already partially evaporated and under a pressure of about 1 kg./cm.². The said first fraction leaves by way of the duct 30 in the form of liquid sub-cooled at about -150° C. The second fraction, which constitutes a few percent of the total, is also sub-cooled at -150° C., by heat exchange in counter-current with methane vapours under low pressure in the exchanger 32, whereafter it is combined with the first fraction. The liquid methane is then expanded to about 1 kg./cm.² in the valve 35 and sent to the separator 36. The vapours formed in the course of the expansion are combined by the tube 37 with the vapours coming from the storage vessel and are sent with the latter through the duct 33 to the exchanger 32. The liquid, which is then at about -160° C., finally passes through the ducts 38 into the storage tank 39. The said tank comprises, for example, a metal inner jacket surrounded by a layer of heat-insulating material of sufficient effectiveness and thickness to reduce to a fairly low degree the losses of liquid methane by evaporation. For the purpose of enabling the inner jacket to undergo expansions or contractions in the course of its cooling or heating, without any excessive stresses being set up, it may advantageously be provided with the system of expansion joints according to French Patent No. 1,230,506 of the February 20, 1959.

The expansion with external work takes place on a fraction of pure methane which passes through the closed cycle hereinafter described.

The warmed up pure methane under low pressure, the origin of which will hereinafter be indicated, is brought to a pressure of about 30 kg./cm.² by the compressor 43. It thereafter passes successively through the ducts 44 and 48 into the heat exchangers 45 and 49, in which it is cooled to about -30° C. in counter-current with liquid propane undergoing evaporation under pressures of about 4.5 kg./cm.² and 1 kg./cm.² respectively. It then passes through the duct 52 into the exchanger 53 in counter-current with cold gaseous methane under low pressure, which cools it to -80° C. It is thereafter passed through the duct 56 into the exchanger 57, in which it is liquefied in counter-current heat exchange with boiling ethylene under a pressure in the neighbourhood of atmospheric pressure, which enters at 58 and leaves at 59. It is thereafter sub-cooled in the exchanger 61 to about -130° C. in heat exchange, on the one hand with a liquid methane fraction expanded to 6 kg./cm.² in the valve 63, and on the other hand, with gaseous methane under a pressure of about 1 kg./cm.² which arrives through the duct 29 and is discharged at 54.

The pure liquid methane leaving the exchanger 61 through 62 is then divided into three parts. A first part, expanded to 6 kg./cm.² in the valve 63, is evaporated and warmed up to about -100° C. in the exchanger 61 and is then passed through the ducts 64 and 65 to an expansion turbine 66. A second part passes through the duct 23 to an expansion valve 24, evaporates and is warmed up to -100° C. in the exchanger 22 in counter-current with the natural gas undergoing liquefaction, whereafter it returns through the ducts 25 and 65 to the turbine 66. The first two combined fractions expand in the said turbine to about 1 kg./cm.² and cool to -145° C. A third portion of the liquid methane is then added thereto through the duct 67 and the expansion valve 68 and partially evaporates, thus lowering the temperature of the whole to -158° C. The combined three portions then pass into the heat exchanger 27, in which they effect an extensive sub-cooling of the liquid natural gas under pressure. They leave the said exchanger after having been warmed up to about -135° C. and pass through the duct 29 into the exchanger 61 and then through the duct 54 into the exchanger 53, in which they are warmed up to

about -40° C., whereafter they return to the compressor 43 through the duct 55.

The liquid propane enters the installation under a pressure of about 12 kg./cm.² through the duct 70. It is expanded in the valves 71 and 72 to 4.5 kg./cm.² and in the valves 73 and 74 to about 1 kg./cm.² before being evaporated in heat exchange with the natural gas and with the pure methane under pressure as hereinbefore stated.

The liquid ethylene enters the installation by way of the duct 75 under a pressure of about 18 kg./cm.². It is expanded in the valve 76 to 4.5 kg./cm.², and in the valves 77 and 78 to about 1 kg./cm.², before being evaporated in the manner already indicated.

The evaporated propane and ethylene are preferably re-compressed and then returned into the cycle with the aid of compressors and pipes, which have been omitted from the drawing for the sake of clarity.

When applied to the treatment of large volumes of natural gas, the installation makes it possible to liquefy almost 95% of the treated natural gas, the loss of liquefied gas by evaporation in the course of the expansion of the liquid under pressure thus being reduced to about 5%.

What we claim is:

1. A process for liquefying a gas to be stored in the liquid state under a low pressure, comprising the steps of:
 - (a) cooling said gas under a relatively high pressure by heat exchange with at least a cooling fluid,
 - (b) cooling and liquefying an auxiliary gaseous stream under a high pressure, expanding it through a valve to an intermediate pressure, vaporizing and warming up at least part of said valve expanded auxiliary stream by heat exchange with the whole of said liquefied auxiliary stream still under a high pressure, and expanding with external work said warmed up auxiliary gaseous stream, and
 - (c) further cooling said gas under a relatively high pressure by heat exchange with said auxiliary gaseous stream expanded with external work, expanding said gas to its storage pressure, and collecting the liquefied gas obtained.
2. A process according to claim 1, wherein the gas to be liquefied is a gaseous mixture, the auxiliary gaseous stream is a stream of a main component of said gaseous mixture, and said auxiliary gaseous stream warmed up by heat exchange with the gas under a relatively high pressure to be liquefied is compressed again and recycled to the cooling and expansion steps.
3. A process according to claim 1, wherein the gas to be liquefied is natural gas, wherein the natural gas under pressure is cooled down to about -100° C. by successive heat exchanges with cooling fluids, then from about -100° C. down to a temperature near its storage temperature under a low pressure by heat exchange with a methane stream previously at least in part expanded with external work.
4. A process according to claim 3, wherein the part of the methane stream expanded with external work is further cooled by the vaporisation in contact therewith of another part of said methane stream, previously liquefied.
5. A process according to claim 3, wherein the cooling fluids are propane and ethane.
6. A process according to claim 3, wherein the cooling fluids are vaporised and warmed up at two different pressure levels by heat exchange with the natural gas under pressure.

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