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Juncker et al.

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[54] **SINGLE-STAGE FRACTIONATION OF NATURAL GAS CONTAINING NITROGEN**

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[30] **Foreign Application Priority Data**

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[52] **U.S. Cl.** **62/40,** 62/28, 62/39, 62/41

[51] **Int. Cl.** **F25j 1/02,** F25j 3/02

[58] **Field of Search** 62/22, 23, 24, 27, 28, 62/39, 41, 40, 38

[56] **References Cited**

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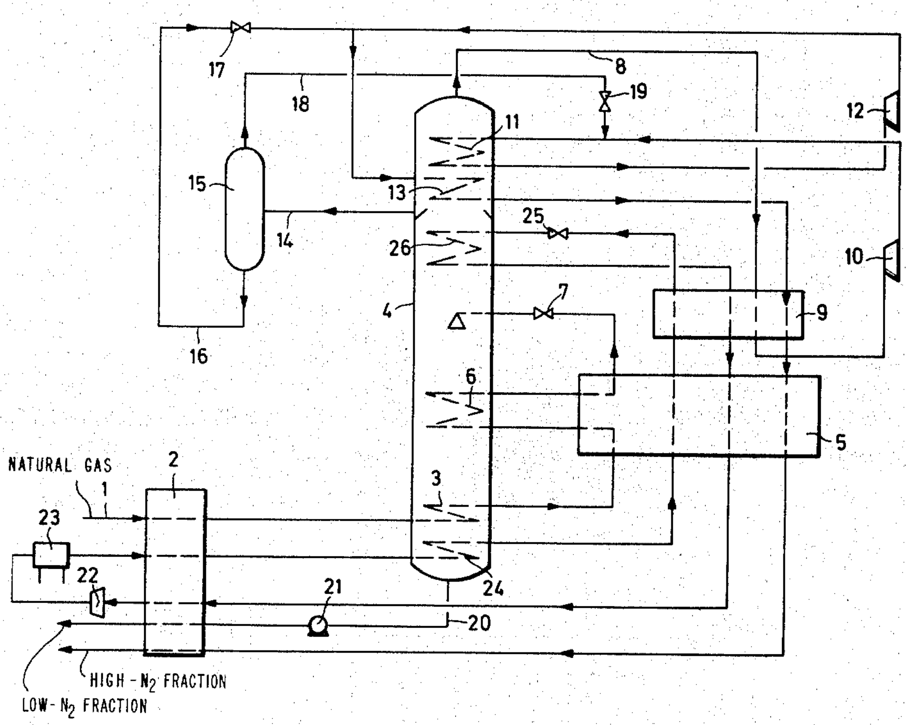
[57] **ABSTRACT**

Separation of natural gas containing nitrogen into a low-nitrogen fraction and a high-nitrogen fraction is achieved in a single distillation column in which reflux is provided by expanding the high-nitrogen fraction with the performance of work and using the resulting refrigeration to condense vapor in the upper section of the column while additional reflux is provided by vaporizing a recycle medium in heat exchange relation with vapor in the column. Incoming natural gas is passed in heat exchange relation with liquid in the column bottom, further cooled and expanded into the middle section of the column. A helium-rich fraction may be separated from the high-nitrogen fraction.

8 Claims, 2 Drawing Figures



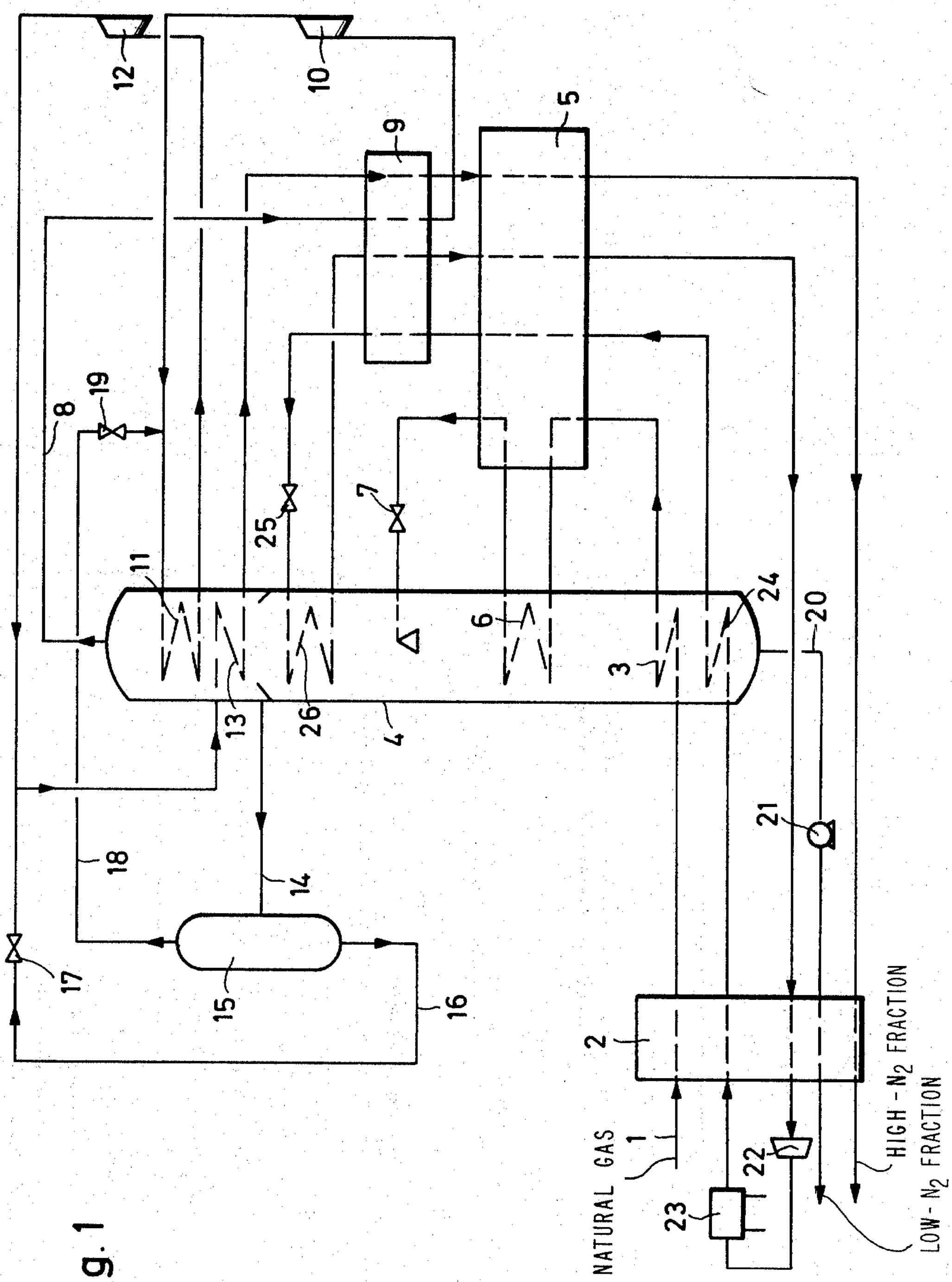


Fig. 1

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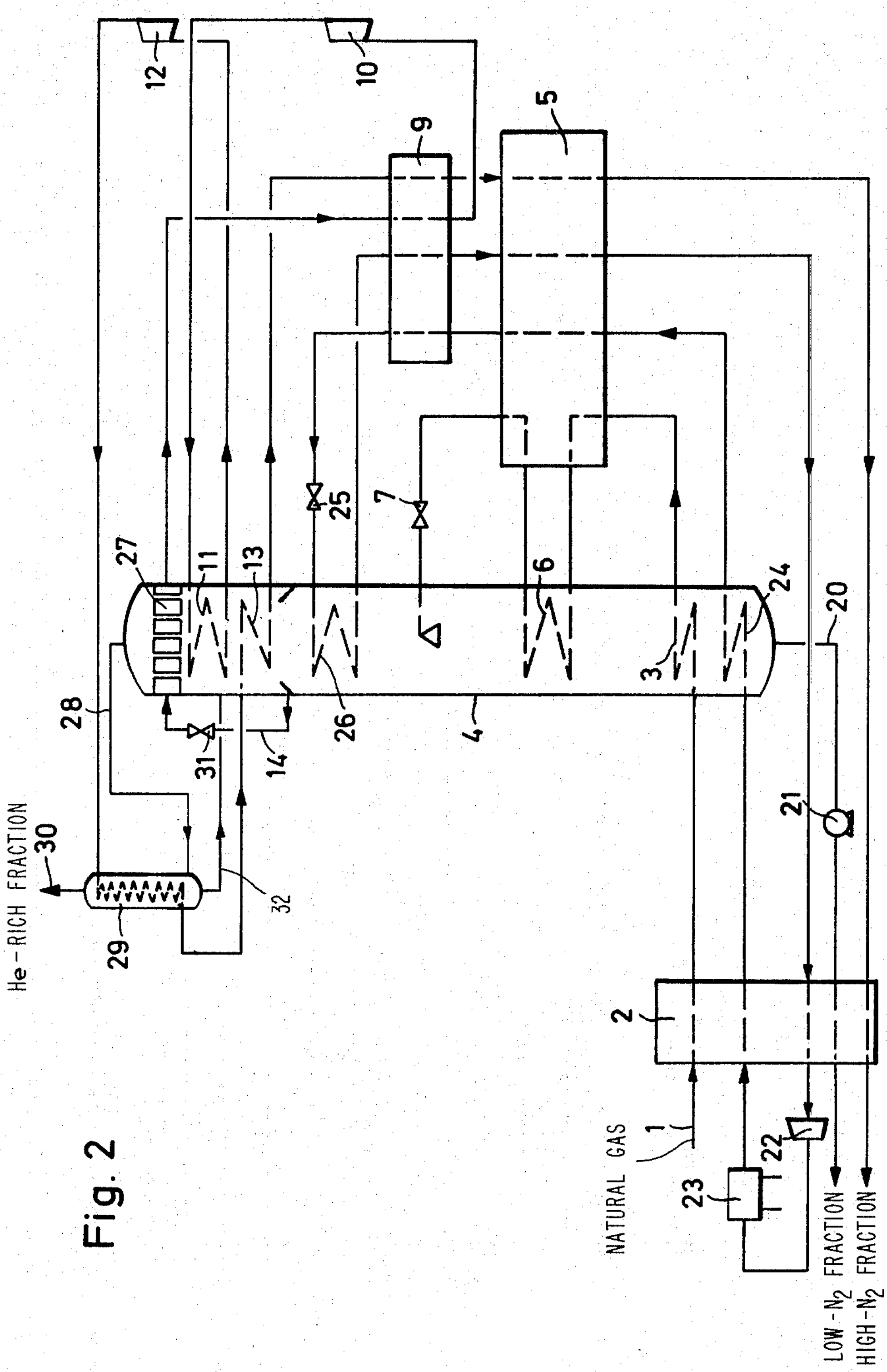
He-RICH FRACTION
30
29
32
28
31
14
27
11
13
26
25
12
10
9
5
7
6
4
3
24
20
21
2
1
NATURAL GAS
22
23
LOW-N_2 FRACTION
HIGH-N_2 FRACTION

Fig. 2

SINGLE-STAGE FRACTIONATION OF NATURAL GAS CONTAINING NITROGEN

BACKGROUND OF THE INVENTION

This invention relates to a process for separating natural gas containing nitrogen into a low-nitrogen and a high-nitrogen fraction by low-temperature distillation in a single column.

Depending on the location of the well, natural gas contains different percentages of nitrogen, carbon dioxide, and heavy hydrocarbons. If the contents of non-combustible components are substantial, it becomes necessary or expedient to separate the non-combustible components prior to transportation or utilization. The customary method for removal of nitrogen is the distillation of natural gas at low temperatures. However, this method is subject to difficulties due to the carbon dioxide in the natural gas.

Carbon dioxide has a triple point of −56.6°C and a very low solubility at low temperatures. Therefore, depending on the method, the carbon dioxide may become solid at low temperatures. In known processes, carbon dioxide is therefore separated before the natural gas enters the low-temperature plant. This separation is costly since, for instance, special carbon dioxide adsorbers with regeneration facilities must be provided, and additional energy is required.

The process of copending application Ser. No. 849,439, filed Aug. 12, 1969, by Martin Streich, avoids carbon dioxide removal by separating the natural gas containing nitrogen into a high-nitrogen and a low-nitrogen fraction prior to two-stage distillation as such. This process is advantageous if the natural gas contains little nitrogen, i.e. up to approximately 20 percent. However, with this process too, the low-pressure column must remain free of carbon dioxide and low in heavy hydrocarbons.

It is the object of this invention to provide a process for the separation of natural gas containing nitrogen into a low-nitrogen and a high-nitrogen fraction by single-stage distillation without the aforesaid shortcomings and limitations regarding carbon dioxide removal.

SUMMARY OF THE INVENTION

According to this invention, the reflux required for the distillation is produced by work-performing expansion of at least the greater part of the high-nitrogen fraction and using the refrigeration produced by this expansion and by vaporization of a recycle medium to condense vapor in the column.

Advantageously, the pressure for this process is between 10 and 35 ata (atmospheres absolute) and a pressure of approximately 28 ata is preferred. Methane is a preferred recycle medium. The process becomes particularly favorable if the high-nitrogen fraction is expanded in several stages with inter-stage heating. In the known manner, the natural gas entering the plant at high pressure is utilized to heat the column sump before it is expanded into the middle section of the distillation column. In an advantageous form of this process, the natural gas is also brought into heat exchange relation with vapor in the lower section of the column before being expanded into the column; thus, the reflux conditions in the column are improved.

The process of the invention is very well suited for recovering a helium-rich fraction from natural gas containing a worthwhile percentage, say over 0.1 percent, of helium. For this purpose, the head product of the distillation, which contains helium, is enriched in two nitrogen condensing stages; in the first stage fractional condensation is carried out by heat exchange with high-nitrogen fraction boiling at high pressure, and in the second stage with cold gaseous high-nitrogen fraction at approximately atmospheric pressure from the work-performing expansion of the high-nitrogen fraction. Refrigeration for the second stage can also be supplied by nitrogen evaporating at low pressure. If necessary, the low-nitrogen liquid fraction from the sump of the distillation column may be pumped to higher pressure before being vaporized and heated to ambient temperature.

BRIEF DESCRIPTION OF THE DRAWINGS

Further advantages of the invention will become apparent from the description of the accompanying drawings of which:

FIG. **1** is a flow diagram of an embodiment of the invention utilizing two-stage expansion of the high-nitrogen fraction; and

FIG. **2** is similar to FIG. **1** with additional features for helium recovery.

DESCRIPTION OF THE PREFERRED EMBODIMENTS

In the process shown in FIG. **1**, natural gas enters the low-temperature plant through line **1** after removal of moisture in a drier (not shown). The natural gas contains about 43% by volume of nitrogen and 56% by volume of methane, the remainder consisting of heavy hydrocarbons 0.4%m helium 0.4% and carbon dioxide 0.2%. The gas pressure is 55 ata.

In heat exchanger **2**, the natural gas is cooled by counter-current flow to the separation fractions. Then it flows through vaporizer **3** in the bottom of distillation column **4** and thus it provides part of the vapor upflow required in column **4**.

The natural gas is then further cooled by flowing through subcooler **5** and coil **6** in the lower section of column **4** where it produces more vapor upflow and thus improves the rectification conditions. Finally, the subcooled natural gas is expanded through expansion valve **7** into the middle section of column **4** to a pressure of 28 ata.

In distillation column **4**, the natural gas is separated into a high-nitrogen mixture at the head of column **4** and a low-nitrogen mixture at the bottom of column **4**. At the head of column **4**, the high-nitrogen mixture is withdrawn in gaseous state through line **8**, somewhat heated in heat exchanger **9**, and expanded in work-performing turbine **10** to a medium pressure of about 6 ata. Then it is reheated in heat exchanger **11** against condensing vapors of the high-nitrogen mixture, further expanded in turbine **12** to about atmospheric pressure and once again heated in heat exchanger **13** against condensing vapors of the high-nitrogen mixture. After further heating in heat exchangers **9**, **5**, and **2**, the high-nitrogen fraction is finally available at the plant outlet at ambient temperature.

Liquid high-nitrogen mixture from the column head can be expanded through line **14** into storage vessel **15** where it is separated into a liquid and a gaseous phase. The gaseous phase is withdrawn through line **18** and fed through valve **19** into the discharge line of turbine

10. The liquid phase is withdrawn through line **16** and expanded through expansion valve **17** into the discharge line of turbine **12**. This is necessary only in case of a sudden need for refrigeration in column **4**, for instance in case of a sudden rise in pressure in column **4**. The liquid from storage vessel **15** will then supply the necessary additional refrigeration.

The low-nitrogen mixture is withdrawn from the sump of column **4** through line **20**, pressurized by pump **21** to the inlet pressure of **55** ata of the natural gas and heated in heat exchanger **2** to ambient temperature.

If the average nitrogen content of the natural gas is about 40 to 50percent by volume, the refrigeration supplied by expansion of the gas in turbines **10, 12** is not enough to produce sufficient reflux for the complete fractionation of the natural gas. Therefore, a recycle medium is provided to improve the reflux conditions. The preferred recycle medium or gas is methane; however a mixture of nitrogen and light hydrocarbons may also be used. In compressor **22**, the recycle gas is compressed from a pressure of about 2 ata to a pressure which is higher than the pressure of distillation column **4**.

Since, in the present example, the recycle medium is also to take refrigeration from the returning low-nitrogen fraction in line **20**, it is compressed to 55 ata. After flowing through after-cooler **23**, it is cooled in heat exchanger **2** against itself and the returning fractions. Then, in heat exchanger **24**, the recycle gas heats the liquid in the column sump, and in heat exchangers **5** and **9** it is subcooled. After expansion to 2 ata in expansion valve **25**, it evaporates in heat exchanger **236**, at least to a great extent. Consequently, with heat exchanger **26**, the recycle medium also produces reflux in column **4**. In heat exchangers **9, 5** and **2**, the recycle gas is reheated to approximately ambient temperature. Since expansion of the high-nitrogen fraction in work-performing turbines **10** and **12** supplies a lot of refrigeration, the recycle gas may be discharged from the low-temperature plant at a relatively low temperature. In compressor **22**, the recycle gas may then be compressed cold, i.e., with less energy consumption. Preferably, the recycle gas is fed into compressor **22** at a temperature of about −35°C since, at that temperature, it is not necessary to use special materials for compressor **22**.

FIG. **2** illustrates the process with the additional recovery of helium. Identical plant components have been marked with identical reference numerals from FIG. **1** and the process will be described only insofar as it differs from the process shown in FIG. **1**. As compared to FIG. **1**, the head of distillation column **4** is provided with an additional condenser **27** where the high-nitrogen vapor mixture rising from heat exchanger **11** is extensively liquefied. The gas withdrawn above condenser **27** through line **28** already contains 5 to 10 percent by volume of helium. In heat exchanger **29**, this vapor mixture is further concentrated by partial condensation to 80 to 90 percent by volume of helium. The helium vapor concentrate is withdrawn through line **30**, while the nitrogen condensate is returned to column **4** by line **32**.

The concentration of helium is accomplished in two stages. The refrigeration for first-stage condenser **27** is supplied by evaporating the high-nitrogen liquid mixture which is withdrawn from column **4** through line **14**, expanded to 24 ata in expansion valve **31** and discharged into condenser **27**. The resulting high-nitrogen vapor mixture is then conducted through turbines **10** and **12** as already described for FIG. **1**. The refrigeration for the concentration second-stage heat exchanger **29** is supplied by the cold high-nitrogen vapor mixture from turbine **12**, which is subsequently heated to ambient temperature as described for FIG. **1**. The refrigeration for the second concentration stage may also be supplied by high-nitrogen liquid mixture evaporating at low pressure.

Heat exchangers **24, 3, 6, 26, 13** and **11** may also be located outside column **4** as plate heat exchangers.

The process of this invention avoids very low temperatures for the natural gas and for the low-nitrogen product fraction. Furthermore, the natural gas evaporates at high pressure. Thus, it is possible to tolerate higher percentages of heavy hydrocarbons in the natural gas without entailing deposits and plugging. Conditions are similar where carbon dioxide is concerned. The carbon dioxide passes through the plant without being deposited anywhere. The costly pre-purification of the natural gas is thus avoided.

What is claimed is:

1. A process for the separation of natural gas containing nitrogen into a low-nitrogen fraction and a high-nitrogen fraction in a single distillation zone which comprises expanding at least the greater part of said high-nitrogen fraction with the performance of work, using the resulting refrigeration to condense vapor of the upper section of said distillation zone to provide reflux in said distillation zone, and providing additional reflux below the aforesaid reflux by vaporizing a closed recycle liquefied gas in indirect heat exchange relation with vapor of said distillation zone after said recycle gas has been passed in indirect heat exchange relation with liquid while the said liquid is at the bottom of said distillation zone.

2. The process of claim **1** wherein the distillation zone is maintained at a pressure in the range of about 10 to 35 ata.

3. The process of claim **1** wherein the nitrogen content of the natural gas is over 20 percent by volume and the expansion of high-nitrogen fraction with the performance of work is conducted in several stages with the resulting refrigeration of each stage being used to condense vapor at different levels of the upper section of the distillation zone.

4. The process of claim **1** wherein the natural gas containing nitrogen is passed in indirect heat exchange relation with liquid at the bottom of the distillation zone, then with the expanded high-nitrogen fraction and again with liquid at an intermediate level of said distillation zone, is expanded to a pressure in the range of about 10 to 35 ata and is discharged into the middle section of said distillation zone.

5. The process of claim **1** wherein the recycle liquefied gas is methane, the nitrogen content of the natural gas is at least 40 percent by volume, and the distillation zone is maintained at a pressure of about 28 ata.

6. The process of claim **1** wherein the natural gas containing nitrogen also contains helium and a helium-rich fraction is recovered by partially condensing nitrogen in the vapor withdrawn from the top of the distillation zone in two condensation stages, for refrigeration for the first of said stages being provided by evaporating at reduced high pressure liquid withdrawn from the top of said distillation zone and the refrigeration for the second of said stages being provided by the high-nitrogen fraction expanded with the performance of work to nearly atmospheric pressure.

7. The process of claim 4 wherein the nitrogen content of the natural gas is over 20 percent by volume, the expansion of high-nitrogen fraction with the performance of work is conducted in two stages, the discharge pressure of the second of said stages being nearly atmospheric, and the resulting refrigeration of each of said stages is used to condence vapor at different levels of the upper section of the distillation zone.

8. The process of claim 7 wherein the natural gas containing nitrogen also contains helium and a helium-rich fraction is recovered by partially condensing nitrogen in the vapor withdrawn from the top of the distillation zone in two condensation stages, the refrigeration for the first of said stages being provided by evaporating at reduced high pressure liquid withdrawn from the top of said distillation zone and the refrigeration for the second of said stages being provided by the high-nitrogen fraction expanded with the performance of work to nearly atmospheric pressure.

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