United States Patent [19]

Becker

[11] **3,707,849**

[45] **Jan. 2, 1973**

[54] **FRACTIONATION OF AIR BY USING REFRIGERATION FROM OPEN CYCLE EVAPORATION OF EXTERNAL CRYOGENIC LIQUID**

[75] Inventor: **Rudolf Becker**, Munich, Germany

[73] Assignee: **Linde Aktiengesellschaft Zentrale Patentabteilung**, Hollriegelskreuth, Germany

[22] Filed: **March 3, 1970**

[21] Appl. No.: **16,077**

[30] **Foreign Application Priority Data**

March 7, 1969 Germany.....P 19 11 765.8

[52] **U.S. Cl.** **62/40**, 62/29, 62/41, 62/13

[51] **Int. Cl.**.....**F25j 1/02**, F25j 3/04

[58] **Field of Search**.....62/911, 28, 29, 40, 13

[56] **References Cited**

UNITED STATES PATENTS

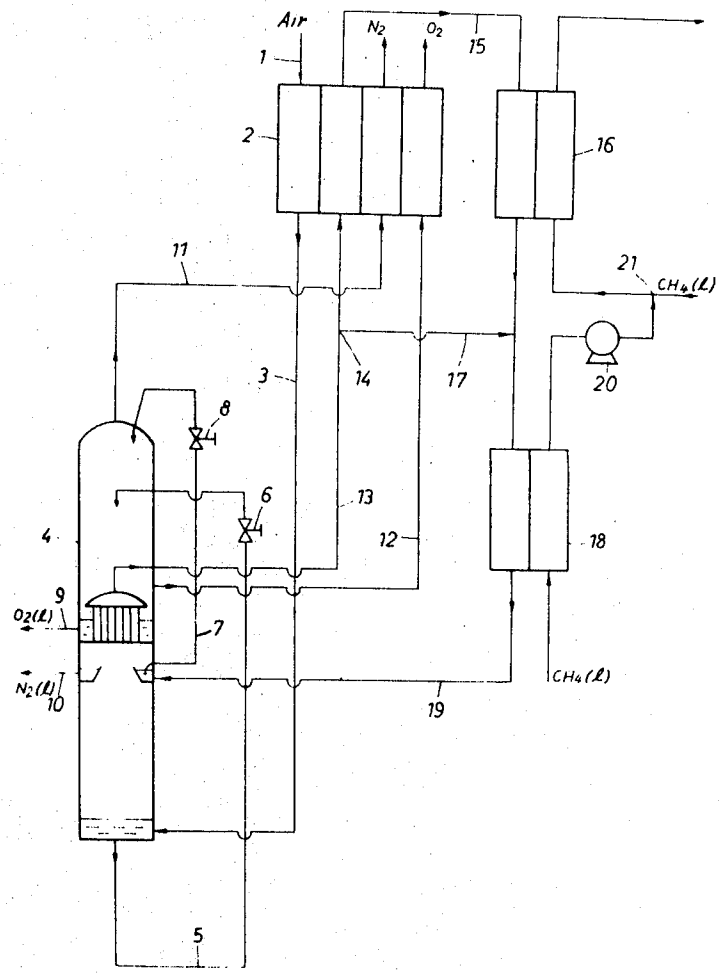
3,339,370	9/1967	Streich	62/40
3,183,677	5/1965	Tafreshi	62/40
3,058,314	10/1962	Gardner	62/40
3,216,206	11/1965	Kessler	62/13
2,685,181	8/1954	Schlitt	62/40
3,107,992	10/1963	Sellmaier	62/40
3,492,828	2/1970	Ruckborn	62/29

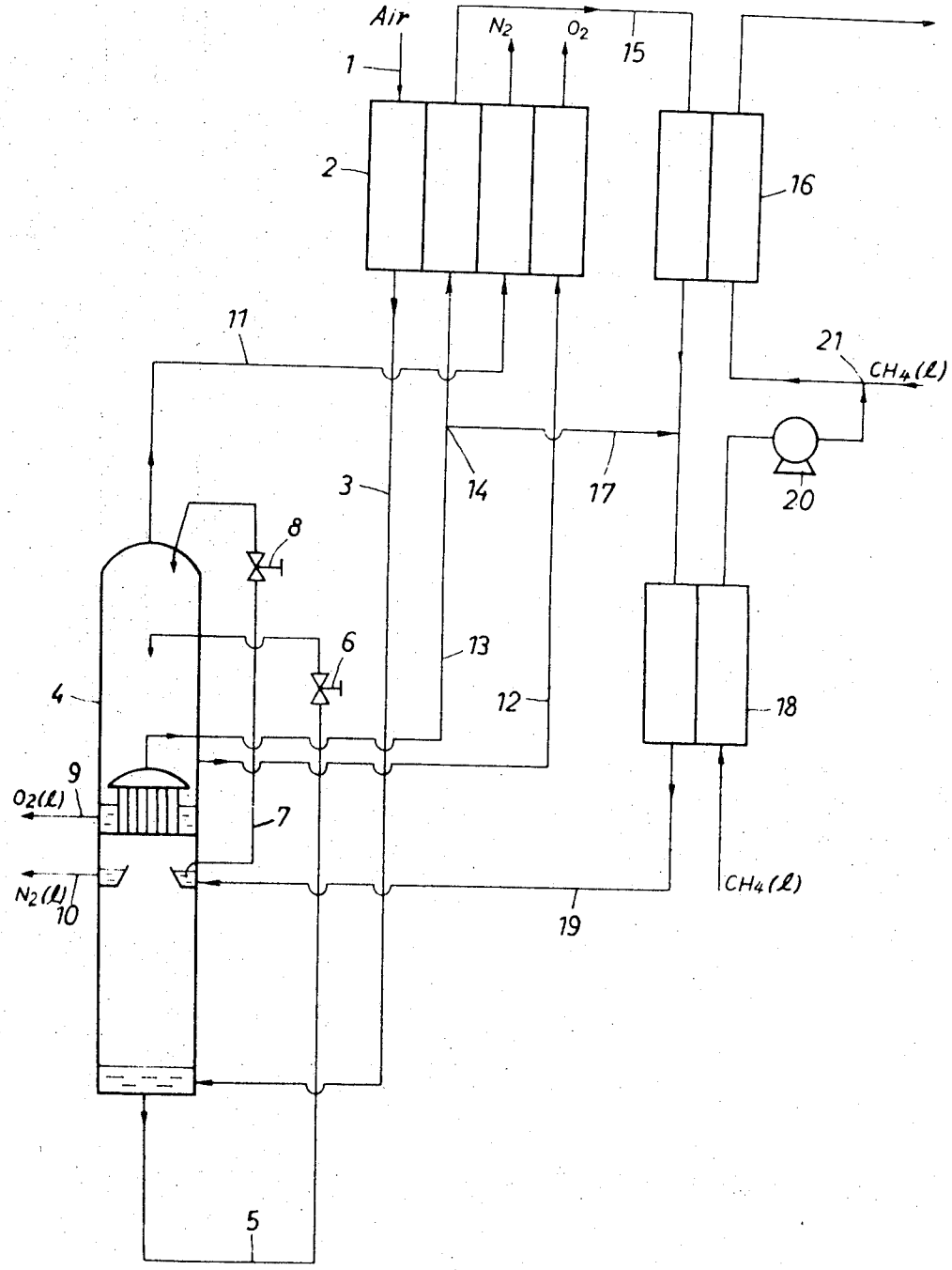
Primary Examiner—Norman Yudkoff
Assistant Examiner—Arthur F. Purcell
Attorney—Millen, Raptes & White

[57] **ABSTRACT**

A system for the low temperature fractionation of air is provided wherein gaseous nitrogen is withdrawn from the rectification column and passed in indirect heat exchange relationship with a vaporizing external refrigerant such as methane or natural gas. The resultant liquified nitrogen is recycled to the column and acts as reflux liquid therein. To provide make-up refrigeration for liquid products that are removed from the system without passing through the main heat exchanger, a portion of the withdrawn nitrogen is heated in the main heat exchanger, thereby cooling the incoming air. The warmed nitrogen is then pre-cooled and subsequently liquefied by the external refrigerant. The nitrogen recycle steps are conducted at approximately the same pressure of nitrogen thereby eliminating the need for compressor and expansion valves on said recycle line.

2 Claims, 1 Drawing Figure





INVENTOR
RUDOLF BECKER
Millen, Raptes & White
by J. William Millen
ATTORNEY

FRACTIONATION OF AIR BY USING REFRIGERATION FROM OPEN CYCLE EVAPORATION OF EXTERNAL CRYOGENIC LIQUID

BACKGROUND OF THE INVENTION

This invention relates to an improved process and apparatus for the low-temperature fractionation of air wherein the refrigeration necessary for the separation and optional liquefaction of products is supplied by the evaporation of an external refrigerant, and wherein the heat transfer of the cold refrigerant to the air is conducted via an intermediate auxiliary gas.

It is old [DAS (German Published Application) No. 1,051,299] to cool air prior to fractionation by heat exchange between the air and a colder liquefied gas. In this process, the heat necessary for evaporating the liquefied gas is obtained from the air, thereby resulting in a substantial cooling of same. In this conventional process, a separate, closed heat transfer cycle is incorporated between the evaporating refrigerant (methane) and the air fractionation plant, the latter being provided with a single or double column. This heat transfer cycle serves the purpose of withdrawing heat from the air to be separated and transferring this heat to the liquid methane to be vaporized.

In this closed heat transfer cycle an auxiliary gas is subjected to two different pressure stages, by a compressor, and the lower pressure stage by an expansion valve. The lower pressure stage is employed for heat exchange with the air and the higher pressure stage is used for heat exchange with the methane. Since such a conventional auxiliary gas cycle is in effect designed according to the principle of refrigerating machine, it must be provided with both a compressor and an expansion valve. Accordingly, the overall fractionation plant is not only made more expensive, but it also becomes considerably more susceptible to operating difficulties.

To avoid these disadvantages, it is also conventional (DAS No. 1,250,460) to effect heat transfer between the refrigerant and the air to be separated without the interposition of an auxiliary gas cycle. However, in this known process it is necessary to transfer the heat to the refrigerant (also methane in this case) at two different zones of the process: (a) to the vapor at the head of the rectification column to provide reflux liquid; and (b) to the air to be separated prior to its introduction into the rectification column. In this known process, it is necessary to allow the methane, in heat exchange with the vapors in the head of the rectification column, to evaporate under subatmospheric pressure so that the air rectification column can be operated under a pressure which is not excessive. In addition, from the standpoint of thermodynamic efficiency, it is necessary to bring the refrigerant not under subatmospheric pressure into heat exchange with the incoming air prior to compression of the latter. However, this results in the disadvantage that, during the subsequent air compression step (indispensable for the fractionation process), a very cold gas is fed to the compressors, thereby entailing difficulties in handling as well as the danger of premature condensation in the compressors.

SUMMARY OF THE INVENTION

It is thus an object of the present invention to provide a process and apparatus substantially eliminating the disadvantages of the above-mentioned conventional processes.

Another object is to combine an air fractionation process with the evaporation of a liquefied refrigerant in a thermodynamically efficient relationship.

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

These objects are attained by withdrawing nitrogen vapor from the air fractionation plant, passing it in indirect heat exchange with an external evaporating cryogenic liquid, thereby liquefying the nitrogen, and passing resultant liquid nitrogen to the fractionation plant for use as liquid reflux. Thus, the nitrogen withdrawn from the fractionation plant at column pressure is employed as the auxiliary fluid.

In a preferred embodiment, a portion of the withdrawn nitrogen is warmed, countercurrently to unfractionated air, to room temperature. Then this warmed portion is cooled and liquefied by heat exchange against the external cryogenic fluid evaporating under a subatmospheric pressure. This liquefied nitrogen portion is also subsequently recycled to the air fractionation plant at approximately the same pressure.

DETAILED DISCUSSION OF THE INVENTION

The process of this invention exhibits the advantage that it does not require a separate, inherently closed cycle of an auxiliary gas for the transfer of heat between the refrigerant and the air separation stage. Furthermore, it is not necessary for the nitrogen withdrawn from the air separation stage to pass through two different pressure stages, as in the above-mentioned prior art process, but rather it can be recycled to the air fractionating column at approximately the same pressure.

According to a preferred embodiment of the invention, the nitrogen withdrawn from the air separation column can be liquefied at a higher elevation than the recycle pipe to the column. In this way, it is unnecessary to provide a recycle pump. If such a piping arrangement is impractical, then a small pump would be necessary.

To conduct the process, gaseous nitrogen is withdrawn from either the head of the high-pressure column of a double-column apparatus, or from the head of a single-column apparatus, and brought into heat exchange with a liquid capable of boiling at a lower temperature than that at which nitrogen can condense, e.g., preferably, liquid methane or liquid natural gas. In this connection, it is especially advantageous to condense the nitrogen at a pressure of 4–8 atmospheres absolute, in heat transfer relationship with vaporizing methane at a pressure of 0.12 – 0.3 atmospheres absolute. The liquefied nitrogen is then recycled into the head of the high-pressure column of the double column apparatus or into the head of a single-column apparatus.

By the process of this invention, it is possible to produce liquid oxygen, liquid nitrogen, or liquid argon. The quantity of the product or products withdrawn in

the liquid phase must be compensated for in the heat balance of the main heat exchange, the latter being conducted in regenerators or reversing exchangers. Accordingly, a special embodiment of this invention compensates for this amount of cold gas missing from the main heat exchange by branching off from the auxiliary nitrogen to be liquefied and recycled, a stream which is then warmed up to room temperature in heat exchange with unfractionated air. For this purpose, there is provided either a special cross section in a reversing heat exchanger, or pipe coils embedded in regenerators. This warmed nitrogen is thereafter preferably cooled against liquid methane at a pressure of about 1 to 100 atmospheres absolute. Then, the thus re-cooled nitrogen is admixed to the other portion of the nitrogen, and both combined streams are liquefied in heat exchange with methane boiling at subatmospheric pressure. The resultant liquid nitrogen is then recycled to the air separation. The following numerical example will serve to demonstrate these relationships:

For the production of 1 Nm^3 of liquid oxygen, there are required 1.4 – 2 Nm^3 of liquid nitrogen at 5 atmospheres absolute. Correspondingly, for the production of 1 Nm^3 of liquid nitrogen, there are required 1.1 – 1.6 Nm^3 of liquid nitrogen at 5 atmospheres absolute. For liquefying 1 Nm^3 of nitrogen under 5 atmospheres absolute, it is necessary to evaporate 0.6 – 1 Nm^3 of methane under subatmospheric pressure. To achieve the necessary heat balance in the regenerator or reversing exchanger, the process of this invention requires an amount of auxiliary nitrogen which is only a few percent, e.g., 5 to 15 percent larger than the sum of the amounts of liquid oxygen and liquid nitrogen which are withdrawn as the products.

Thus, by the process of this invention, liquid oxygen, liquid nitrogen, or liquid argon can be produced quite inexpensively without a separate source of cold; however, the process can also be employed in those cases where oxygen or nitrogen is not to be withdrawn in the liquid form, but rather in the gaseous phase.

The production of liquid nitrogen can be utilized in an especially advantageous manner for a nitrogen scrubbing stage of hydrogen production from coke oven gas, refinery waste gas, or other industrial gases, especially for the production of ammonia synthesis gas.

BRIEF DESCRIPTION OF THE DRAWING

The attached drawing is a schematic diagram of a preferred embodiment of this invention wherein a double column is employed and liquid nitrogen and liquid oxygen can be withdrawn as products.

DETAILED DESCRIPTION OF THE DRAWING

Air of **6** atmospheres absolute enters a main heat exchanger **2** via conduit **1**. In the embodiment illustrated in the drawing, the main heat exchanger **2** is a reversing exchanger having four cross sections, i.e. four flow paths. However, this heat exchanger can just as well be substituted by a corresponding number of reversible regenerators with tubular coils installed therein.

In the main heat exchanger **2**, the air is cooled to close to its liquefaction temperature, thereby congealing out CO_2 and H_2O. The remaining cold air passes through conduit **3** into the bottom of the high pressure column of a double-column apparatus **4**. From the bottom of the high pressure column, air enriched to about 40 percent in oxygen is withdrawn via conduit **5**, expanded in the throttle valve **6** to 1.5 to 2 atmospheres absolute, and then fed to the middle section of the low-pressure column. Through conduit **7**, liquid nitrogen is withdrawn from the high-pressure column, which nitrogen is expanded in the throttle valve **8** and fed, as reflux, to the head of the low-pressure column. Through conduits **9** and **10**, it is possible to withdraw liquid products from the rectification column (conduit **9**: liquid oxygen; conduit **10**: liquid nitrogen). Gaseous nitrogen and gaseous oxygen are withdrawn through conduits **11** and **12**, respectively, and are passed through in main heat exchanger **2**, to cool the raw air.

Gaseous nitrogen withdrawn from the upper part of the high-pressure column through conduit **13** is divided into two partial streams at **14**. The partial stream **15** is heated in the main heat exchanger **2**, in a separate cross section, to approximately room temperature and serves to achieve a heat balance in the main heat exchanger for the products withdrawn in the liquid phase at **9** and **10**, respectively. (The amount in each stream is dependent on how much liquid product is withdrawn from the process. If none, 100 percent of the nitrogen would be line **17**.) This partial stream **15** is cooled, in heat exchanger **16**, countercurrently with liquid, evaporating and/or vaporized methane of about 1 atmosphere absolute. Thereafter, the nitrogen is combined with the other portion of the nitrogen, flowing through conduit **17**, and liquefied in heat exchanger **18** countercurrently to liquid methane, the latter being vaporized during this step under a pressure of 0.12 atmospheres absolute. The entire resultant liquefied nitrogen is then recycled as reflux liquid through conduit **19** to the upper part of the high-pressure column. The methane evaporated in the heat exchanger **18** is compressed in the vacuum pump **20** and then combined, at **21**, with the methane which is under a higher pressure. Instead of methane or natural gas there could be used also Oxygen, Argon and Krypton for the purposes of the invention. These gases, however, have no real practical importance, because it is rather expensive to provide sufficient amounts especially of liquid rare gases.

In the embodiment illustrated in the figure, the entire amount of air to be separated is compressed and fed to the high-pressure column. However, the process of this invention could also be made even more inexpensive by providing that only about 80 percent, rather than the entire quantity, of the air to be separated is introduced into the high-pressure column, whereas the remainder, about 20 percent, could be fed immediately into the low-pressure column at the pressure therein.

The preceding example can be repeated with similar success by substituting the generically or specifically described reactants and/or operating conditions of this invention for those used in the preceding example.

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

What is claimed is:

1. In a process for the low-temperature separation of air in an air fractionating column, wherein refrigeration is supplied by the evaporation of methane used as an external refrigerant, and wherein the transfer of the cold from the refrigerant to the air is conducted by way of an intermediate auxiliary gas, the improvement comprising the steps of:

withdrawing nitrogen gas from said air fractionating column, dividing the withdrawn nitrogen into two partial streams, warming one of said partial streams of nitrogen to ambient temperature in indirect heat exchange with unfractionated air, precooling resultant warmed partial stream in heat exchange with said refrigerant, the latter being at a pressure of about 1 atmosphere absolute, passing both said precooled partial streams of nitrogen and the other of said two partial streams of nitrogen as said auxiliary gas, in indirect heat exchange relationship with said methane refrigerant, the latter evaporating at a pressure of 0.12 – 0.3 atmospheres absolute, thereby condensing said nitrogen, returning resultant liquid nitrogen to the air fractionating column, and employing said liquid nitrogen as reflux liquid, with the provision that the steps of withdrawing said nitrogen from the air fractionating column, condensing said nitrogen, returning said liquid nitrogen to the column and of employing said liquid nitrogen as reflux are conducted at approximately the same pressure within the range of 4 – 8 atmospheres absolute, with the further provision that said methane be maintained out of heat exchange contact with said air through a common heat exchange wall.

2. A process as defined by claim 1, wherein said air fractionating column is a double column and the nitrogen gas is withdrawn from the high-pressure column and is returned as liquid to the same.

* * * * *