

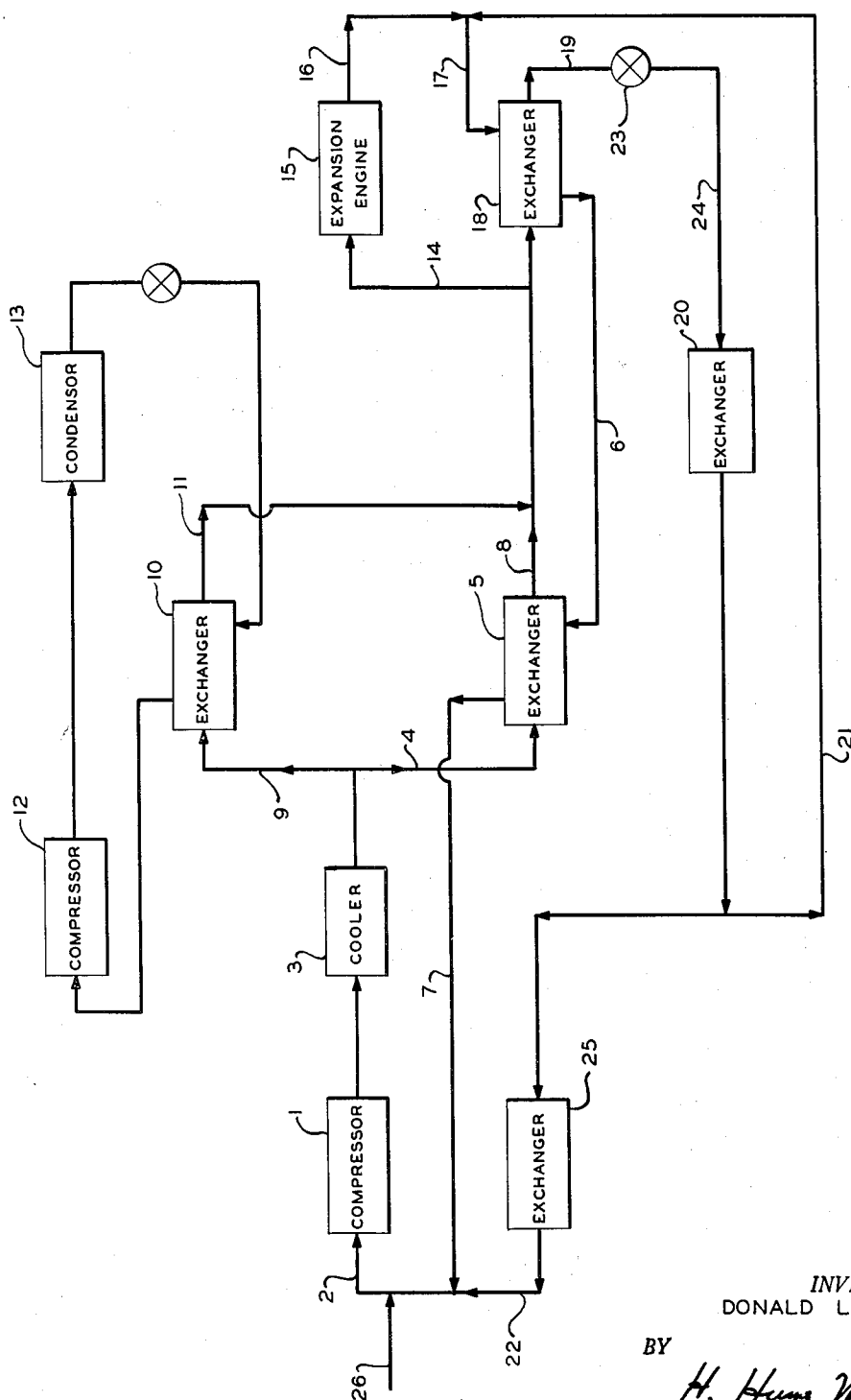
Dec. 20, 1960

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2,964,913

SEPARATION OF AIR

Filed Dec. 31, 1956



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2,964,913

## SEPARATION OF AIR

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Filed Dec. 31, 1956, Ser. No. 631,714

6 Claims. (Cl. 62—9)

This invention relates to the separation of air by low temperature techniques. More specifically, this invention provides an improved refrigeration system for liquefying and separating air at low temperatures.

In a process where air is separated into component constituents at least one of which is liquid by low temperature techniques, the efficiency of the refrigeration system is of great importance in determining the economic success of the process.

It is an object of the invention to provide an improved refrigeration system for low temperature separation of air into component constituents at least one of which is liquid.

Other objects of this invention will be apparent hereinafter.

The improved refrigeration system of this invention comprises two separate refrigeration cycles in mutual heat exchange. A closed nitrogen cycle is used to attain the very low temperatures required for liquefaction and separation of air into component constituents. In this cycle, the nitrogen is compressed and cooled, and a portion of the high pressure nitrogen is expanded in an expansion engine to very low temperature. The remainder of the high pressure nitrogen is cooled by exchange with the nitrogen from the expansion engine and with nitrogen which had been used to refrigerate process streams. The cooled, high pressure nitrogen is then expanded through an expansion valve and used to refrigerate process streams in the gas separation.

A second refrigeration cycle is provided in this invention in heat exchange with a portion of the high pressure nitrogen in the above closed nitrogen cycle. This second refrigeration cycle is used to cool a portion of the high pressure nitrogen during a certain part of the nitrogen cycle as more fully described hereinafter. Inclusion of this second refrigeration cycle, in accordance with the invention, greatly improves the efficiency of the gas separation process; liquid product yield increases of 30% or more can be obtained with power input increases of only about 4%, for example.

Practice of the instant invention is best described in conjunction with the drawing which accompanies this specification and claims; the drawing illustrates in diagrammatic form an embodiment of the invention.

Referring to the accompanying drawing, the refrigeration system of the invention includes compressor 1 and aftercooler 3 wherein the cycle nitrogen is compressed and the heat of compression removed; heat exchangers 5 and 18 wherein high pressure cycle nitrogen is cooled to very low temperature by exchange with low pressure cycle nitrogen; exchanger 10 wherein high pressure cycle nitrogen is refrigerated by a second separate refrigeration cycle including compressor 12 and condenser 13; expansion engine 15 wherein high pressure cycle nitrogen is expanded, while doing external work, to low temperature and pressure; pressure reducing valve 23 through which cold high pressure cycle nitrogen is expanded thereby to liquefy this nitrogen; heat exchanger 20 wherein the

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liquid cycle nitrogen vaporizes thereby liquefying a process gas stream and providing the refrigeration necessary to produce liquid air separation product; and heat exchanger 25 wherein low pressure cycle nitrogen cools air which is to be separated.

For most efficient operation of the refrigeration system, two factors must be considered. Since the refrigeration necessary to separate air into at least one liquid product is provided by vaporizing liquid cycle nitrogen in exchanger 20, a maximum percentage of the cycle nitrogen from exchanger 18 must be liquefied in order to minimize the amount of high pressure nitrogen circulated in the cycle. In addition, a maximum percentage of refrigeration must be recovered from the cycle nitrogen in each cycle prior to its recompression in compressor 1 for the next cycle. These objectives are substantially achieved in practice of the instant invention.

Cycle nitrogen, which is used to refrigerate air which is to be separated into at least one liquid component, is compressed in compressor 1 to high pressure. The heat of compression is removed and the high pressure nitrogen is cooled almost to ambient temperature in aftercooler 3. One or more compression stages with or without cooling after each stage can be employed in compressing the cycle nitrogen and removing the heat of compression.

The high pressure nitrogen at almost ambient temperature is divided into two portions after leaving cooler 3. One portion is passed through line 4 to exchanger 5 wherein the high pressure nitrogen is cooled by counter-current indirect heat transfer with low pressure cycle nitrogen which is returning to compressor 1 through lines 7 and 2 for use in another refrigeration cycle. The relative portions of the streams passing through exchanger 5 are adjusted according to the heat content of the streams to achieve a minimum temperature difference at the warm end in order that a maximum amount of the refrigeration be recovered from the low pressure cycle nitrogen stream returning to the compressor while at the same time the high pressure nitrogen is cooled to a minimum temperature necessary for efficient subsequent expansion engine operation.

In the refrigeration system of the invention, not all of the high pressure cycle nitrogen can be passed through exchanger 5 in heat exchange with low pressure cycle nitrogen while maintaining a minimum temperature difference at the warm end of the exchanger and while lowering the high pressure nitrogen temperature at the cold end of exchanger 5 to a suitably low temperature for subsequent highly efficient operation of the expansion engine. This is due to the difference in heat capacities of the gas streams and to the fact that there is a greater amount of high pressure cycle nitrogen from cooler 3 than low pressure cycle nitrogen to exchanger 5 since part of the low pressure cycle nitrogen is used to cool air in exchanger 25 rather than to cool compressed cycle nitrogen.

The portion of the high pressure cycle nitrogen from cooler 3 which cannot be cooled to efficient expansion engine inlet temperature in exchanger 5 is passed through line 9 to exchanger 10. In exchanger 10, the high pressure cycle nitrogen is refrigerated by a separate refrigeration system to about the minimum temperature necessary for efficient expansion engine operation, i.e. to about the same temperature as the outlet high pressure nitrogen from exchanger 5. The separate refrigeration system used to refrigerate the nitrogen in exchanger 10 can be any efficient commercial refrigeration system. A monochlorodifluoromethane (Freon 22) refrigeration system is preferred although substantially equivalent systems such as other Freon systems as well as ammonia systems and the like can be provided. The refrigerant used to cool

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the high pressure nitrogen in exchanger 10 is compressed in compressor 12 and condensed in condenser 13.

The high pressure cycle nitrogen streams from exchangers 5 and 10 are passed through lines 8 and 11, respectively, and are recombined. The temperature of this combined stream is such that this nitrogen can be expanded in an efficient expansion engine from the high pressure of the system to about saturation temperature at a desired lower pressure.

A portion of the recombined high pressure cycle nitrogen is passed to exchanger 18 wherein by indirect heat transfer with low pressure cycle nitrogen from the expansion engine and from the process exchanger 20, this high pressure nitrogen is cooled to a very low temperature. The flow rates through exchanger 18 are adjusted to give a minimum temperature difference at the cold end, in order that the high pressure cycle nitrogen be cooled to the lowest possible temperature. The remaining portion of the high pressure cycle nitrogen is passed through line 14 to expansion engine 15. In this engine, the nitrogen is expanded with the performance of external work to about its saturation temperature at a lower pressure. The expanded nitrogen is passed through lines 16 and 17 to exchanger 18.

The cold high pressure cycle nitrogen is removed from exchanger 18 and passed through line 19 to expansion valve 23. The nitrogen is expanded through valve 23 with a consequent temperature reduction and formation of liquid. This expanded cycle nitrogen is passed through line 24 to exchanger 20. In exchanger 20, the liquid nitrogen is vaporized by indirect heat transfer with a stream, e.g. a nitrogen reflux stream, from the gas separation process whereby refrigeration necessary to liquefy process streams and produce liquid air separation product is provided.

The gaseous cycle nitrogen from exchanger 20 is divided into two portions. One portion is passed to exchanger 25 wherein the nitrogen is used in conjunction with waste gaseous streams (not shown) from the air separation to cool incoming air to substantially its liquefaction temperature. The cycle nitrogen is heated almost to ambient temperature in exchanger 25 before returning through lines 22 and 2 to compressor 1. In this way, substantially all of the refrigeration is recovered from this portion of the cycle nitrogen.

The remainder of the gaseous cycle nitrogen from exchanger 20 is passed through lines 21 and 17 to exchanger 18 wherein as above described it is used in conjunction with exhaust nitrogen from the expansion engine to cool the high pressure cycle nitrogen almost to the temperature of the low pressure nitrogen.

The low pressure cycle nitrogen from exchanger 18 is passed through line 6 to exchanger 5 wherein it is used to cool high pressure cycle nitrogen as hereinbefore described. The low pressure nitrogen leaves exchanger 5 almost at ambient temperature and is returned through lines 7 and 2 to compressor 1. In this way substantially all of the refrigeration is recovered from this cycle nitrogen prior to recompression.

Make-up nitrogen is added to the cycle nitrogen as needed through line 26.

Operation of the refrigeration system of this invention as described above results in the economic separation of air into at least one liquid component. Inclusion of the supplementary refrigeration cycle in conjunction with the nitrogen cycle results in balanced operation with a minimum of power input necessary for a given liquid production.

The following illustrates a practice of the invention:

A refrigeration system of the invention is employed to provide refrigeration for an air separating and liquid oxygen producing process such as described in U.S. Patent No. 2,762,208 to Wolcott Dennis. Cycle nitrogen is compressed to about 170 atm. in compressor 1 and cooled almost to ambient temperature in cooler 3.

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The cooled high pressure nitrogen is then divided into two streams, one passing to exchanger 5 and the other to exchanger 10. In exchanger 5, the high pressure nitrogen is cooled to about 225° K. by heat exchange with low pressure cycle nitrogen at about 4.35 atm., entering exchanger 5 through line 6. The relative quantities of high pressure cycle nitrogen and low pressure cycle nitrogen in exchanger 5 are adjusted to provide minimum (about 5° K.) temperature difference at the warm end and to insure cooling the high pressure nitrogen to about 225° K. at the cold end. By this operation high recovery of refrigeration from the low pressure cycle nitrogen prior to recompression is insured, while at the same time the high pressure cycle nitrogen is cooled to an efficient expansion engine inlet temperature as explained more fully hereinafter. A weight ratio of about 1.8 parts of the low pressure cycle nitrogen to about 1.37 parts of the high pressure cycle nitrogen in exchanger 5 is suitable to obtain the above results at conditions in the present system.

The high pressure cycle nitrogen which cannot be cooled to suitable expansion engine inlet temperature in exchanger 5 is passed to exchanger 10 wherein it is cooled to about the same temperature as the high pressure nitrogen from exchanger 5, i.e. about 225° K. by indirect heat transfer with a separate refrigerant. In the instant system, a separate Freon 22 refrigeration cycle is employed to cool the high pressure cycle nitrogen. About 40% of the high pressure cycle nitrogen from cooler 3 is cooled in exchanger 10.

The cooled high pressure nitrogen streams from exchangers 10 and 5 are recombined by means of lines 11 and 8. A portion of this cycle nitrogen is passed to exchanger 18 wherein it is cooled to very low temperature. It is this portion of the cycle nitrogen which is used to liquefy and separate components of air. The remainder of the cooled high pressure cycle nitrogen at about 225° K. is passed through line 14 to expansion engine 15 wherein the nitrogen is expanded with the performance of work to substantially saturation temperature, 95° K. at the low pressure of the nitrogen cycle, about 4.35 atm. About 1.32 parts by weight of the nitrogen are expanded in expansion engine 15 per part of nitrogen which is cooled in exchanger 18.

The high pressure cycle nitrogen is cooled to very low temperature in exchanger 18 by indirect heat exchange with low pressure nitrogen at substantially saturation temperature of 95° K. from the expansion engine and from process exchanger 18. The exchanger 18 is a commercial heat exchanger capable of close temperature approaches (about 5° K.) at the ends thereof. The quantities of the nitrogen streams are adjusted in accordance with their heat contents such that a minimum of low pressure nitrogen sufficient to cool the high pressure cycle nitrogen almost to the temperature of the low pressure stream is employed. About 1.8 parts of low pressure nitrogen by weight are employed per part of high pressure nitrogen to cool the high pressure stream to about 100° K.

The cold high pressure nitrogen stream is then expanded to lower pressure with resultant liquefaction of a large percentage of the nitrogen. The expanded nitrogen is then passed to process exchanger 20 wherein it is vaporized by indirect heat exchange with a process stream from the air separation process. In the instant practice of the invention, the refrigeration cycle nitrogen is vaporized while liquefying reflux nitrogen (not shown) which is used in the fractionation of air and production of liquid oxygen as shown in U.S. Patent No. 2,762,208 to Wolcott Dennis.

The vaporized cycle nitrogen from exchanger 20 is divided into two portions, one going to exchanger 25 and the other to exchanger 18. Sufficient of the vaporized nitrogen, about 50% thereof, is used to cool the air which is to be separated to about liquefaction temperature in

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exchanger 25 prior to recompression in compressor 1. Gas from the air separation, e.g., waste nitrogen, is also used to cool the air in exchanger 25. This cooling is not shown in the drawing.

The remainder of the gaseous cycle nitrogen from exchanger 20 is passed by lines 21 and 17 to exchanger 18 where with the exhaust nitrogen from the expansion engine it is used to cool high pressure nitrogen. The low pressure nitrogen leaves exchanger 18 at about 230° K., and is passed to exchanger 5 through line 6. In exchanger 5 the low pressure nitrogen is heated almost to ambient temperature prior to recompression by heat exchange with a portion of the high pressure nitrogen from cooler 3.

Practice of the invention as described above results in minimum nitrogen cycle size with substantially complete recovery of refrigeration per cycle. When compared to a similar nitrogen cycle refrigeration system without a second refrigeration cycle in heat exchange therewith, substantial liquid product increases are obtained through practice of this invention without correspondingly increased operating costs.

I claim:

1. The method of providing refrigeration for the separation of air into components at least one of which is liquid which comprises: compressing a closed cycle nitrogen refrigerant to elevated pressure, cooling a portion of the compressed cycle nitrogen by heat exchange with lower pressure cycle nitrogen, cooling the remainder of the compressed cycle nitrogen by heat exchange with a separate refrigerant, expanding a portion of the cooled, compressed cycle nitrogen to low temperature, further cooling the remainder of the cooled compressed cycle nitrogen by heat exchange with the expanded cycle nitrogen, liquefying at least a portion of the cooled, compressed cycle nitrogen, and vaporizing the liquefied cycle nitrogen thereby providing refrigeration for the separation of air into at least one liquid component, the expanded cycle nitrogen and the vaporized liquefied cycle nitrogen being recompressed after the recovery of refrigeration therefrom.

2. The method according to claim 1 wherein said separate refrigerant is monochlorodifluoromethane.

3. The method of providing refrigeration for the separation of air into at least one liquid component which comprises: compressing a closed cycle nitrogen refrigerant to elevated pressure, cooling a portion of the compressed cycle nitrogen by heat exchange with lower pressure cycle nitrogen, cooling the remainder of the compressed cycle nitrogen by heat exchange with a separate refrigerant, expanding a portion of the cooled compressed cycle nitrogen to substantially saturation temperature at a lower pressure with the performance of work, further cooling the remainder of the cooled compressed cycle nitrogen by heat exchange with the expanded cycle nitro-

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gen, expanding the so-cooled cycle nitrogen to liquefy at least a portion thereof, vaporizing said liquefied cycle nitrogen thereby providing refrigeration for the separation of air into at least one liquid component, substantially completely recovering the refrigeration in said vaporized cycle nitrogen by heat transfer with compressed cycle nitrogen and with air which is to be separated, the refrigeration in the work expanded cycle nitrogen being recovered by heat exchange with compressed cycle nitrogen, and recompressing said vaporized cycle nitrogen and work expanded cycle nitrogen.

4. The method according to claim 3 wherein said separate refrigerant is monochlorodifluoromethane.

5. The method of providing refrigeration for the separation of air into components at least one of which is liquid which comprises: compressing a closed cycle nitrogen refrigerant to high pressure, cooling the compressed cycle nitrogen to about ambient temperature, cooling a portion of said compressed cycle nitrogen by indirect heat exchange with low pressure cycle nitrogen thereby to recover substantially all of the refrigeration from said low pressure cycle nitrogen, cooling the remainder of said compressed cycle nitrogen by indirect heat exchange with a separate refrigerant, combining said cooled compressed cycle nitrogen, expanding a portion of the cooled compressed cycle nitrogen to substantially saturation temperature at lower pressure with the performance of work, further cooling the remainder of the cooled compressed cycle nitrogen by indirect heat exchange with low pressure cycle nitrogen including the cycle nitrogen from said expansion, expanding said further cooled cycle nitrogen thereby to liquefy at least a portion thereof, vaporizing said liquefied cycle nitrogen thereby to provide refrigeration for the separation of air into at least one liquid component, recovering substantially all of the refrigeration from a portion of the vaporized cycle nitrogen by heat exchange with compressed cycle nitrogen, and recovering all of the refrigeration from the remainder of the vaporized cycle nitrogen by heat exchange with air to be separated, the refrigeration in the work expanded cycle nitrogen being recovered by heat exchange with compressed cycle nitrogen, and recompressing the work expanded cycle nitrogen and the vaporized cycle nitrogen.

6. The method according to claim 3 wherein said separate refrigerant is monochlorodifluoromethane.

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