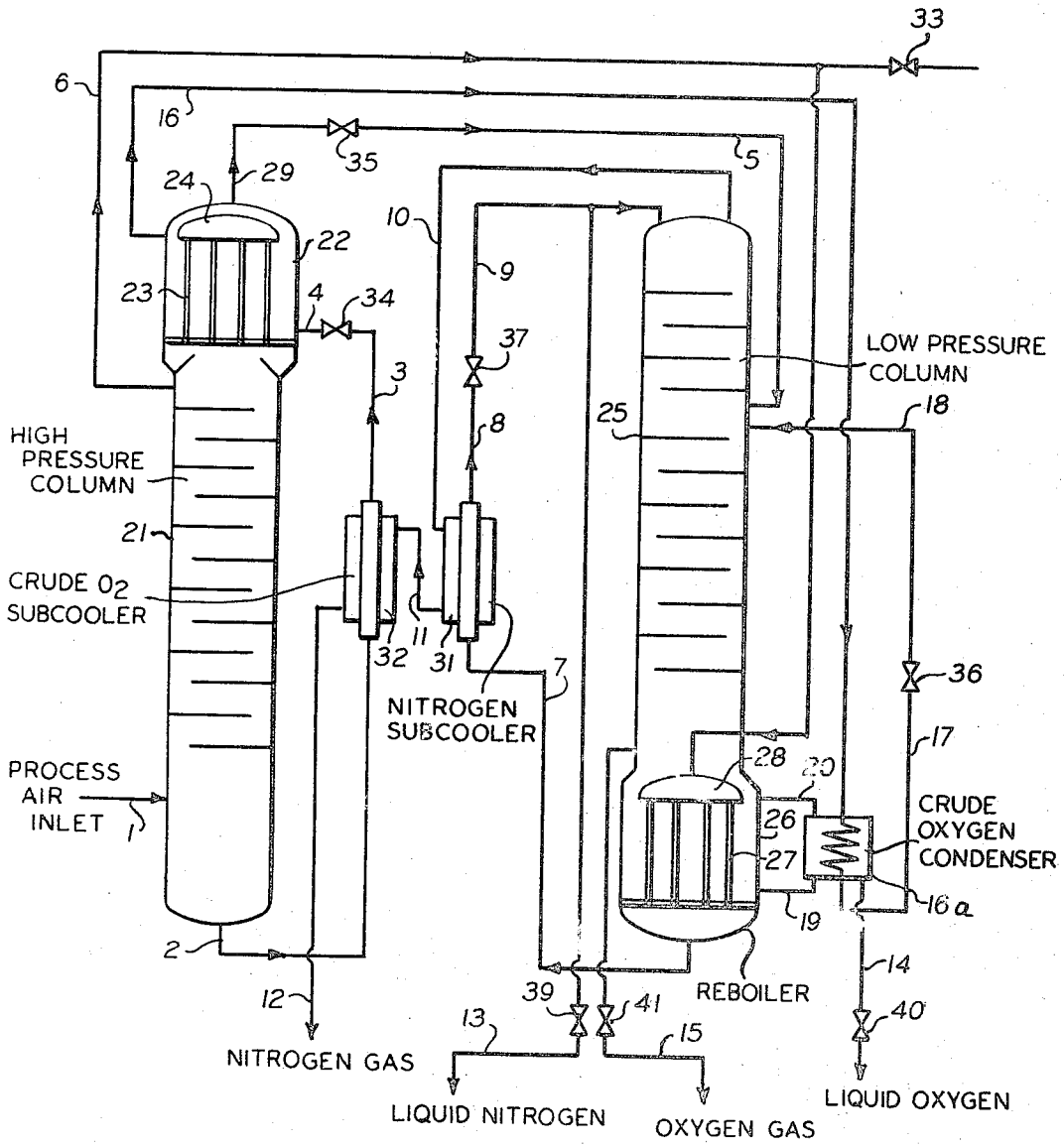
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SEPARATION OF GAS MIXTURES

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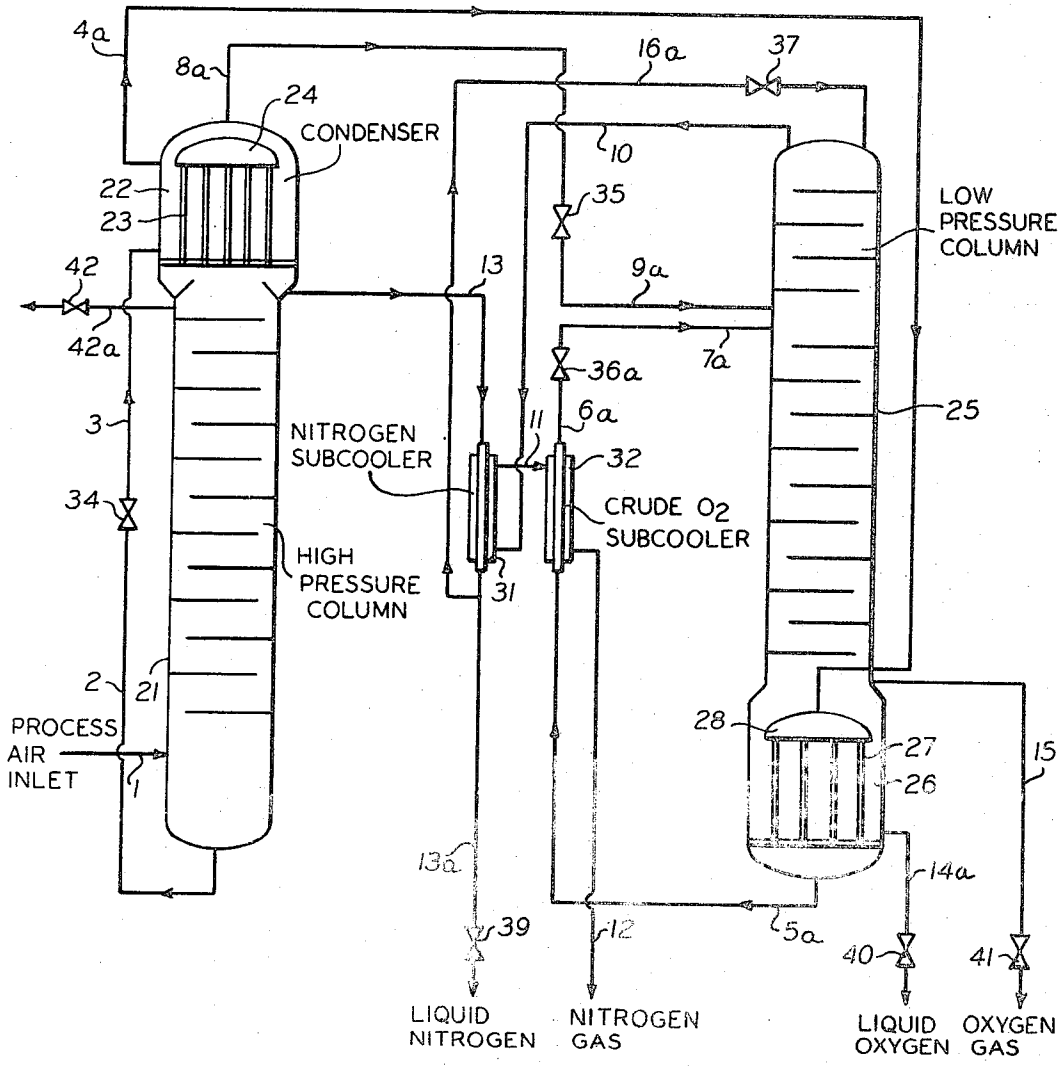
FIG. 1.



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FIG. 2.



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3,348,385

SEPARATION OF GAS MIXTURES

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Filed Dec. 23, 1964, Ser. No. 420,673
10 Claims. (Cl. 62—29)

ABSTRACT OF THE DISCLOSURE

A process for separation of a gas mixture, such as a liquid air separation plant, wherein the high pressure column and the low pressure column are separate, i.e. the low pressure column is not mounted atop the high pressure column, and, for liquid air separation, a crude oxygen cut from the high pressure column is used to condense nitrogen at the top of the high pressure column, causing some vaporization of the oxygen cut, and the crude cut is then cooled for condensation and is then introduced into the low pressure column.

This invention relates to separation of gas mixtures. While the invention was occasioned by the need for improvement in separation of air into oxygen and nitrogen, the procedure and the apparatus provided by the invention have general application for separation of gas mixtures which include components differing in properties as do the components of air, so that the separation can be effected by liquefaction and fractionation. Thus, whereas the invention will be described in detail with reference to separation of air into oxygen and nitrogen, it will be understood that the procedure and apparatus of the invention find application in the separation of other gas mixtures.

It is known to separate air into oxygen and nitrogen by a two-step fractionating procedure, the first step being a high pressure fractionation and the second step being a low pressure fractionation. Thus, air cooled to a low temperature is introduced into a high pressure fractionation column which can be operated at, for example, 75–150 p.s.i.a. to provide a crude oxygen cut as bottoms of the high pressure fractionation and a vapor rich in nitrogen as overhead of the high pressure fractionation. The crude oxygen cut is then transferred to the low pressure column and is there fractionated to provide oxygen of high purity, for example, 99% oxygen. The overhead of the low pressure column, like the overhead of the high pressure column, is nitrogen. It is common to provide the equipment so that the condenser for the high pressure column functions as the reboiler for the low pressure column and to utilize as the cooling fluid for condensation of overhead of the high pressure column, the liquid oxygen product (bottoms) of the low pressure column. Thus, nitrogen for reflux for the high pressure column is provided. Further, by the condensation of the overhead of the high pressure column, liquid nitrogen can be obtained for utilization as reflux in the low pressure column. In general, the principal product of such operation is liquid oyygen. Oxygen gas, nitrogen gas, or liquid nitrogen can also be taken as product, depending on the particular operating conditions employed. In apparatus of the type referred to here, the low pressure column is disposed above the high pressure column and the condenser-reboiler is disposed between the two columns. This apparatus has the disadvantage of requiring very considerable height due to the arrangement of the low pressure column atop the high pressure column.

It has been proposed to reduce the height requirements for an air separation plant by providing a so-

called split column, i.e. a plant wherein the high pressure column and the low pressure column are located side by side, rather than one atop the other.

A split column operation is the subject of a co-pending application by one of us, namely Jeram S. Kamlani, Ser. No. 275,009, filed Apr. 23, 1963, now Patent No. 3,270,514. The said co-pending application is directed to providing a split column operation characterized in that heat transfer area requirements are relatively low, and in that the process is carried out in a manner not requiring the use of mechanical, heat, or vapor pumps to transfer material between the columns, and further characterized in that savings can be realized in respect to equipment requirements since a substantial portion of one of the columns, namely the low pressure column, can be made of a diameter smaller than heretofore commonly used.

The instant invention is directed to provide an improvement in the process of the aforementioned patent, more particularly, it is a principal object of the invention to further accommodate the process to operation without the use of mechanical, heat, or vapor pumps. Another principal object of the invention is to provide improvement in over-all oxygen recovery. Other objects of the invention will be apparent from the ensuing description.

According to the process of said co-pending application, a gas mixture including a low boiling component and a high boiling component is separated by liquefaction and fractionation. The mixture is first fractionated in a high pressure column to provide an overhead vapor rich in the low boiling component and as bottoms a crude cut of the high boiling component in liquid state. A portion of the high pressure column overhead vapor is condensed and the resulting condensate is used as reflux for the high pressure column. Crude cut, i.e. bottoms of the high pressure column, is transferred to a low pressure column and fractionated therein to provide a low pressure column overhead vapor rich in the low boiling component and a low pressure column bottoms rich in the high boiling component. In these operations, the process of the co-pending application is akin to known procedures. It departs from the prior art, however, in that crude cut, i.e. bottoms of the high pressure column, is reduced in pressure and is then utilized for condensing the reflux for the high pressure column by indirect heat exchange of crude cut at the reduced pressure and the overhead vapor of the high pressure column. Further, an overhead vapor product rich in the low boiling component can be withdrawn from the high pressure column and bottoms of the low pressure column can be reboiled by indirect heat exchange of the overhead vapor product of the high pressure column and the bottoms of the low pressure column.

The transfer of crude cut from the bottom of the high pressure column to the top of the high pressure column, for utilization thereof in the condenser, is effected by the pressure of the crude cut at the bottom of the high pressure column, so that it is not necessary to resort to mechanical pumping or other means for effecting the desired lifting of the crude cut. Further, the crude cut utilized for the indirect heat exchange in the condenser of the high pressure column can be transferred to the low pressure column for fractionation, and this transfer is effected by the pressure of crude cut in the condenser. Thus, the transfer of crude cut to the condenser and from the condenser to the low pressure column is effected by pressure of the crude cut and without the utilization of means such as pumps.

According to the improvement of the instant invention, the crude cut which is transferred from the condenser of the high pressure column to the low pressure column is

cooled prior to introduction into the low pressure column.

In the indirect heat exchange between the crude cut and the overhead vapor of the high pressure column, at least a portion of the crude cut is vaporized. It has been found that it is desirable to cool the crude cut before introduction thereof into the low pressure column to condense at least part of the material so vaporized. The cooling of the crude cut transferred to the low pressure column can be by indirect heat exchange with a low pressure column fraction rich in high boiling component, and preferably is by such heat exchange with bottoms of the low pressure column. A distinctive feature of the operation is that all of the air or other gas treated in the system can be introduced into the high pressure column.

An alternative procedure for cooling crude cut according to the invention, during transfer thereof to the low pressure column, is to utilize the crude cut from the overhead condenser of the high pressure column as the heating fluid in the reboiler of the low pressure column. The resulting cooled crude cut can then be introduced into an intermediate point of the low pressure column. Preferably all of the crude cut vapor is condensed in the low pressure column reboiler, and the crude cut from the reboiler is further cooled in a subcooler before introduction into the low pressure column. The crude cut is reduced to the pressure of the low pressure column following passage through the reboiler and, preferably, following passage through the subcooler. In this embodiment overhead of the high pressure column is condensed to the extent necessary to provide not only reflux for the high pressure column, but further to the extent necessary to provide reflux for the low pressure column. All of the high pressure column overhead can be condensed. The condensing is effected with crude cut from the bottoms of the high pressure column, as is described above. In this embodiment all air or other gas treated can be introduced into the high pressure column and, further, all of the air or other gas can be condensed in the high pressure column.

The invention is further described in reference to the accompanying drawings, wherein:

FIG. 1 is a flow diagram for an embodiment wherein crude cut is cooled during transfer with bottoms of the low pressure column and overhead vapor of the high pressure column is used as the heating fluid in the low pressure column reboiler; and

FIG. 2 is an embodiment wherein the crude cut is cooled during transfer by use thereof as the heating fluid in the reboiler, the reflux for the low pressure column being produced in the condenser of the high pressure column.

The apparatus of the invention, as shown in FIG. 1, can include a first or high pressure fractionating column **21** outfitted with an overhead condenser **22**. The overhead condenser can be of the dome type common to air separation apparatus and includes the tubes **23** and dome **24**. The tubes are in communication with the fractionating column so that overhead vapor from the fractionating column can pass into the tubes and the dome for indirect heat exchange with material on the outside of the tubes and the dome. Further, the high pressure column **21** is provided with process air inlet line **1** and with conduit **2** which communicates the bottom of the fractionating column with the condenser for transfer of crude cut or bottoms of the fractionating column **21** to the condenser for indirect heat exchange between crude cut and overhead vapor product inside the tubes and dome of the condenser **22**. The bottoms passed through line **2** to the condenser **22** must be a cut rich in high boiling component. It need not necessarily be the cut at the lowermost point in the high pressure column **21**. All of the gas treated in the system can be introduced into the high pressure column **21** via line **1**.

The apparatus further includes a second or low pressure column **25** which is provided with a reboiler **26**. The arrangement employing two separate columns is called a "split column," or separated column operation. The reboiler **26** of the low pressure column can be constructed similar to the condenser of the high pressure column and includes the tubes **27** and the dome **28**. Conduit means in the form of line **6** communicate the top of the high pressure column **21** with the tube side of the reboiler **26** for transfer of vapor from the top of the high pressure column to the reboiler wherein the vapor is utilized as heating medium for the reboiler. Further, there can be provided conduit means in the form of line **5** for transferring crude cut from the shell side of the condenser **22** to the low pressure column **25**. Vapor generated in the condenser **22** can pass through the line **29** and into line **5**, and the vapor can be introduced into the low pressure column **25** at an intermediate point thereof. Preferably, there is no flow or little flow in line **5**.

Thus, according to the invention, crude cut from condenser **22** passes through line **16** to cooler **16***a* wherein it is passed in indirect heat exchange relation with bottoms of the low pressure column. Bottoms passes through line **19** to the exchanger **16***a* and from the exchanger back to the column through line **20**. Oxygen product can be withdrawn through line **14** as liquid oxygen from the exchanger **16***a*. Upon cooling of the crude cut, vapor, preferably all of the vapor, is condensed and the crude liquid then passes through line **17** to pressure reducing valve **36** and then through line **18** to an inlet, at an intermediate point, to the low pressure column **25**.

In the reboiler **26** the overhead vapor utilized for heating condenses and the condensate passes through line **7** to the subcooler **31**, then through line **8** and line **9** to the top of low pressure column **25** where the condensate serves as reflux. The overhead vapor from the low pressure column passes through line **10** to the subcooler **31** and in the subcooler is passed in indirect heat exchange relationship with respect to the condensate flowing to the top of the low pressure column for use as reflux. From the subcooler **31**, the overhead of the low pressure column **25** flows through line **11** to subcooler **32**, wherein the vapor overhead from the low pressure column is passed in indirect heat exchange relationship with the crude cut passed from the bottom of the high pressure column **21** to the condenser of the high pressure column. The vapor leaves the subcooler **32** through line **12**. The high boiling component, substantially pure, in the form of gas, can be withdrawn through line **15**. The low boiling component, in liquid state, can be withdrawn through line **13**.

The apparatus of the invention is provided with suitable control valves, and can be operated in a manner to provide liquid or gas products in selected proportions.

In utilization for separation of air into oxygen and nitrogen, the process air introduced through line **1** can be at a low temperature and can be about 6–25% liquid. The high pressure column can be operated at a pressure in the range of 75–150 p.s.i.a. The crude oxygen cut passed through line **2** is reduced in pressure by valve **34** to the pressure utilized on the shell side of the condenser **22** and then passes as a liquid into the condenser **22**. The pressure of the crude cut at this point is a pressure intermediate the pressure of the high pressure column and the pressure of the low pressure column. The pressure of the low pressure column can be 20–40 p.s.i.a. The drawoff line **29** is provided with a valve **35** and as noted above, preferably, this valve is closed. Nitrogen gas passed via line **6** to the reboiler **26** is condensed in the reboiler and passed through lines **7**, **8**, and **9** to the top of the low pressure column. The valve **37** is utilized to control the flow of nitrogen to the top of the low pressure column. The nitrogen gas line **6** connecting the top of the high pressure column **21** with the reboiler **26** of the low pressure column **25**, is provided with a draw-off line **33** having valve **38**; the liquid nitrogen line **13** is provided with valve **39**; the liquid oxygen line **14** is provided with valve **40**; these various

valves can be manipulated to provide suitable operation of the process.

The invention as represented by FIG. 1 is further described in the following example.

Example I

A plant according to the flow sheet set forth in the drawing was assembled using available equipment. The high pressure column had 20 GEECO Navy-type perforated trays with a helical coil condenser at the top. The low pressure column had twenty-three GEECO Navy-type perforated trays. The reboiler associated with the low pressure column was a horizontal shell-and-tube type exchanger located adjacent to but separated from the column itself. The subcoolers were wound "Helicoil" exchangers. The plant was operated to provide a liquid oxygen product of purity 99.6% and a nitrogen vapor product. Neither gaseous oxygen nor liquid nitrogen was taken as product. Conditions for the various streams, measured or calculated were as is set forth in the following table. All of the air was introduced into the high pressure column.

Line	Flow, lbs./hr.	Press., p.s.i.g.	Temp., ° F.	Comp., percent O_2
1	400	74	(−282.5)	21.0
2	(238)	74	(−280.0)	31.0
3	(238)			31.0
4	(238)	54	(−286.5)	31.0
5	0			
6	(162)	74	(−283.0)	7.0
7	(162)	74	(−283.0)	7.0
8	(162)	74	(−283.0)	7.0
9	(162)	6	(−314.0)	7.0
10	333.5	6	(−314)	7.5
11	333.5			7.5
12	333.5			7.5
13	0			
14	66.5	6	(−291)	99.60
15	0			
16	238	54	(−286.5)	31.0
17	238	54	(−286.5)	31.0
18	238	6	(−310)	31.0

Figures in parentheses were calculated.

Equipment having more trays can be utilized to provide higher purity oxygen. Thus, by utilizing more trays, oxygen of purity of 99.9% can be produced.

In the example, the product is liquid oxygen. The system can, however, be operated to produce high purity oxygen gas, nitrogen gas and/or nitrogen liquid.

In the embodiment shown in FIG. 2 the crude cut is cooled during transfer by use thereof as the heating medium in the reboiler of the low pressure column. Reflux for the low pressure column is obtained from the condenser atop the high pressure column. As between FIG. 1 and FIG. 2, like reference characters refer to corresponding parts. The process as shown in FIG. 2 differs from that shown in FIG. 1 as is indicated in the following with reference to FIG. 2.

All of the gas treated is introduced into the high pressure column and is condensed therein, and all the nitrogen gas at the top of the high pressure column **21** is condensed in condenser **22**. Liquid crude oxygen from the bottom of the high pressure column is used at a reduced pressure to condense the nitrogen. Part of the liquid nitrogen from the condenser is returned to the high pressure column and the balance is passed through line **13** to nitrogen subcooler **31** and then passes through line **16***a* to the top of the low pressure column **25**. If desired, nitrogen gas can be withdrawn from the top of the high pressure column through line **42***a*. Normally, however, valve **42** in line **42***a* will be closed. The crude oxygen gas generated in the condenser **22** passes through line **4***a* to the reboiler **26** located at the bottom of the low pressure column and is condensed therein by heat exchange with the liquid oxygen bottoms of the low pressure column. Liquid crude oxygen passes from the reboiler through line **5***a* to oxygen subcooler **32** and then through line **6***a* and pressure reducing valve **36***a* and on through line **7***a* to an inlet at about the middle of the low pressure column. Lines **8***a* and **9***a* which interconnect the crude oxygen side of the condenser **22** with about the middle of the low pressure column **25** are outfitted with a valve **35**. This line is a by-pass line and the valve **35** is normally closed. Liquid nitrogen can be taken as product through line **13***a* outfitted with valve **39**; nitrogen gas is taken off as product through line **12**; liquid oxygen is taken off as product through line **14***a* which connects with the reboiler of the low pressure column; and oxygen gas can be taken off through line **15** which connects with the low pressure column near the bottom thereof. If desired, the crude oxygen passed from the bottom of the high pressure column to the condenser **22** thereof, through line **2**, can be cooled in a subcooler. A portion of the overhead nitrogen gas from top of low pressure column can be used as cooling medium in such a cooler.

The embodiment shown in FIG. 2 is further described in the following example.

Example 2

In this example, equipment as is described in Example 1 is used. The plant can be operated to provide a liquid oxygen product of purity 99.5% and a nitrogen vapor product, without taking any gaseous oxygen or liquid nitrogen as product. All of the air is introduced into and condensed in the high pressure column. Conditions for the various streams are as is set forth in the following table:

Line	Flow, lbs./hr.	Pressure, p.s.i.g.	Temp., ° F.	Composition, percent O_2
1	400	74	−282.5	21.0
2	238	74	−280.0	31.0
3	238	54	−286.5	31.0
4	238	54	−286.5	31.0
5	238	54	−286.5	31.0
6	238	54		31.0
7	238	6	−310.0	31.0
8	0			
9	0			
10	333.5	6	−314.5	7.5
11	333.5	6		7.5
12	333.5	6		7.5
13	162	74	−288	7.0
13a	0			7.0
14	162	74		7.0
14a	66.5	6	−291	99.5
15	0	6	−291	99.5
16	162	74		7.0
17	162	6		7.0

As can be observed from the examples, the process of the invention provides a high yield of high purity oxygen. By reason of utilization of the procedure of the invention, the split column operation can be utilized while, at the same time, performance is comparable to that for the conventional double column, wherein the low pressure column is mounted over the high pressure column with a condenser-reboilder interposed between the upper and lower column sections.

While the invention has been described in reference to particular embodiments thereof, various modifications will be apparent from the description, and it is desired to secure by these Letters Patent all such variations.

What is claimed is:

1. In a process for separation of a gas mixture including a low boiling component and a high boiling component by liquefaction and fractionation in a high pressure column and a low pressure column separate from the high pressure column, comprising

(a) fractionating the mixture in the high pressure column to provide an overhead vapor rich in the low boiling component and as bottoms a crude cut of the high boiling component in liquid state;

(b) condensing a portion of the high pressure column overhead vapor and using the resulting condensate as reflux for the high pressure column;

(c) reducing the pressure of crude cut and condensing the reflux for the high pressure column by in-

direct heat exchange of crude cut at the reduced pressure and the overhead vapor of the high pressure column;

(d) transferring crude cut utilized in said indirect heat exchange to the low pressure column and fractionating the crude cut therein to provide a low pressure column overhead vapor rich in the low boiling component and a low pressure column bottoms rich in the high boiling component;

the improvement which comprises:

(e) vaporizing at least a portion of the crude cut during said indirect heat exchange and cooling said at least partially vaporized crude cut during said transfer to the low pressure column to condense crude cut.

2. Process according to claim **1**, wherein

(a) crude cut for said indirect heat exchange is transferred from the bottom of the high pressure column to the top of the high pressure column for said heat exchange and said transfer is effected by reduction in pressure of crude cut, and said transfer of crude cut, utilized for said indirect heat exchange, to the low pressure column is effected by pressure of crude cut.

3. Process according to claim **2**, wherein said cooling during transfer of the crude cut to the low pressure column is effected by indirect heat exchange with a low pressure column fraction rich in high boiling component.

4. Process according to claim **3**, wherein said fraction is low pressure column bottoms.

5. Process according to claim **1**, wherein an overhead vapor product rich in low boiling component is withdrawn from the high pressure column, and bottoms of the low pressure column are reboiled by indirect heat exchange of said overhead vapor product of the high pressure column and the bottoms of the low pressure column.

6. Process according to claim **1** wherein all of the gas is introduced into the high pressure column and is condensed in the high pressure column.

7. In a process for separation of a gas mixture including a low boiling component and a high boiling component by liquefaction and fractionation in a high pressure column and a low pressure column separate from the high pressure column, comprising

(a) fractionating the mixture in the high pressure column to provide an overhead vapor rich in the low boiling component and as bottoms a crude cut of the high boiling component in liquid state;

(b) condensing a portion of the high pressure column overhead vapor and using the resulting condensate as reflux for the high pressure column;

(c) reducing the pressure of crude cut and condensing the reflux for the high pressure column by indirect

heat exchange of crude cut at the reduced pressure and the overhead vapor of the high pressure column;

(d) transferring crude cut utilized in said indirect heat exchange to the low pressure column and fractionating the crude cut therein to provide a low pressure column overhead vapor rich in the low boiling component and a low pressure column bottoms rich in the high boiling component;

the improvement which comprises:

(e) condensing a further portion of the high pressure column overhead vapor and using the resulting further condensate as reflux in the low pressure column;

(f) cooling crude cut during said transfer by indirect heat exchange of crude cut and bottoms of the low pressure column.

8. Process according to claim **7**, wherein

(a) crude cut for said indirect heat exchange is transferred from the bottom of the high pressure column to the top of the high pressure column for said heat exchange and said transfer is effected by reduction in pressure of crude cut, and said transfer of crude cut, utilized for said indirect heat exchange, to the low pressure column is effected by pressure of crude cut;

(b) at least a portion of the crude cut is vaporized during said indirect heat exchange with high pressure column overhead vapor and

(c) vapor of crude cut produced during said indirect heat exchange is condensed during said cooling during transfer of the crude cut to the low pressure column.

9. Process according to claim **8**, wherein the crude cut cooled by indirect heat exchange with the bottoms of the low pressure column is introduced into the low pressure column at an intermediate height thereof.

10. Process according to claim **7**, wherein all of the gas treated is introduced into the high pressure column.

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UNITED STATES PATENT OFFICE

CERTIFICATE OF CORRECTION

Patent No. 3,348,385 October 24, 1967

Jeram S. Kamlani et al.

It is hereby certified that error appears in the above numbered patent requiring correction and that the said Letters Patent should read as corrected below.

Column 6, in the table, fourth column, line 13 thereof, for "-288" read -- -283 --.

Signed and sealed this 26th day of November 1968.

(SEAL)
Attest:

Edward M. Fletcher, Jr.
Attesting Officer

EDWARD J. BRENNER
Commissioner of Patents