METHOD AND APPARATUS FOR FRACTIONATION OF AIR

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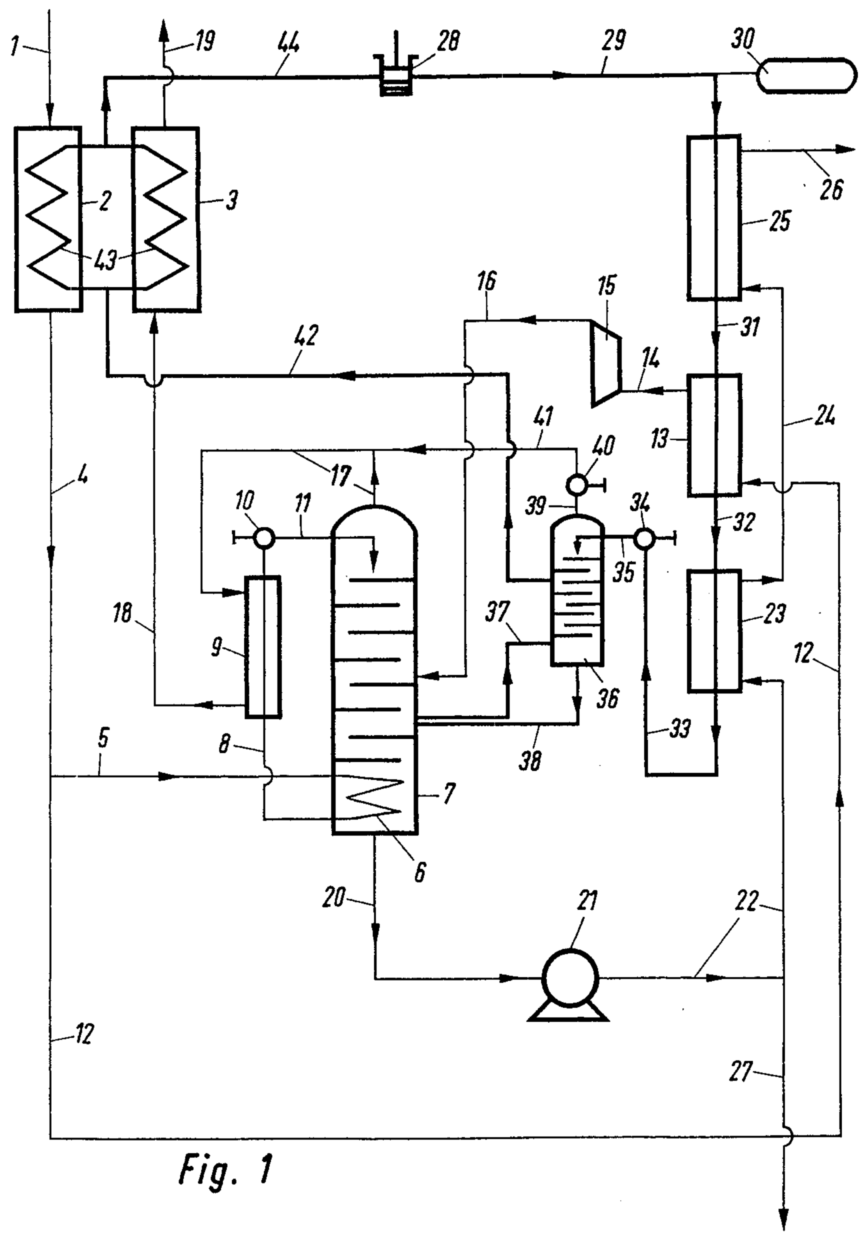

Fig. 1

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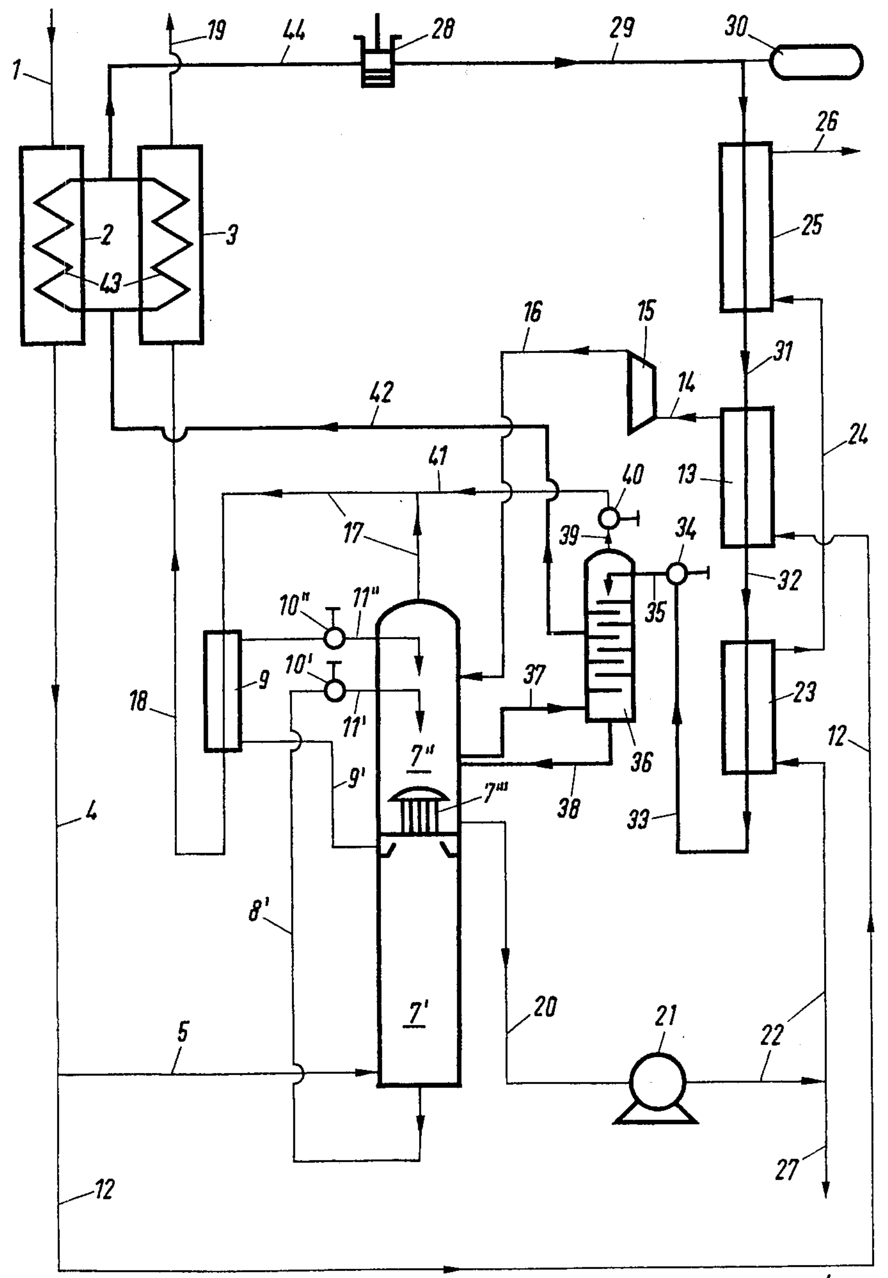

Fig. 2

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METHOD AND APPARATUS FOR FRACTIONATION OF AIR

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This invention relates to a method and an apparatus for fractionation of air by liquefaction and rectification, in which the separated oxygen product is passed in liquid condition through pressure increasing means and is then passed through a heat exchanger to be vaporized by heat from a circulating gas and brought to the temperature of the surroundings.

It is old to use nitrogen in open circulation where it is liquefied by heat exchange with oxygen that is being vaporized. It is then necessary, however, to use very high nitrogen pressures in order to cause the nitrogen to liquefy while the oxygen is under a pressure of about 25 atmospheres.

It is also known that lower gas pressures can be used if argon, having a boiling point closer to that of oxygen, is used as the circulating gas. The use of argon, however, makes it necessary to use closed circuit circulation and to continually replenish the argon by that which has been separated from the fractionated air.

The principal objects of this invention are to eliminate the above-mentioned disadvantages by keeping the circulating gas under only a moderate pressure, and also at the same time using open circulation.

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

In order to accomplish the objects of this invention, a circulating gas is used which consists of a mixture of 8 to 30% oxygen, preferably 20 to 25% oxygen, not more than 10% nitrogen, and the remainder being argon.

A preferred feature of this invention is that the moderately compressed gaseous mixture which is liquefied by heat exchange with vaporizing oxygen is delivered with expansion in open circulation directly into the head of an auxiliary column. The bottom portion of this auxiliary column is supplied with argon-containing gases from a low pressure air fractionating column while oxygen-enriched liquid from the foot of the auxiliary column is returned to the air-fractionating column. Small amounts of nitrogen-enriched gases are released from the head of the auxiliary column, and from the middle portion of the latter, the vaporized and partially rectified circulating gas is drawn off, to be warmed by heat exchange with the air to be fractionated, and then to be compressed.

In order to prevent the circulating gas, which is continually being replenished by argon-, oxygen- and nitrogen-containing gases from the low pressure column, from becoming excessively enriched with nitrogen or other undesired components, it is advantageous for a small amount of nitrogen-enriched gas from the head of the auxiliary column to be combined with the nitrogen taken from the head of the low pressure air-fractionating column. This combined nitrogen stream, after passage through a countercurrent cooler in indirect heat exchange with the reflux liquid for the low pressure column, serves as regenerator-purging gas or can be used for other purposes.

With this method of operation it is sufficient if the circulating gas mixture is compressed to about 1.2 to 1.4 times the pressure of the compressed liquid oxygen before the gas mixture enters the heat exchangers for vaporizing and warming the oxygen.

The amount of the circulating gases is preferably only 20 to 50%, more preferably 30 to 40% greater than the amount of oxygen to be vaporized.

For producing the necessary refrigerant values and establishing a heat balance in the various exchange aggregates, it is desirable for a portion of the air that is to be fractionated and which has been cooled and purified in a regenerator system, to be passed through a heat exchanger to cool the circulating gas mixture and then to be expanded while doing external work, into the middle portion of the low pressure air fractionating column.

In order to establish the required temperature conditions in the various fluid streams, it is a preferred technique to have the compressed circulating gas mixture cooled and at least partly liquefied before it passes through the expansion value and into the auxiliary column, by being brought into heat exchange first with at least partly vaporized compressed oxygen that is to be heated, then with the above-mentioned portion of the cooled and cleansed air, and finally for vaporizing the liquid oxygen that has been directly taken from the liquid oxygen pressure-pump.

Reference is now directed to the attached drawings wherein for purposes of illustration, and compliance with 35 U.S.C. 112, FIGURES 1 and 2 are schematic flowsheets of preferred specific embodiments of this invention. It is to be understood, however, that these embodiments are not to be considered limitative in any way whatsoever of the appended claims which particularly point out and distinctly claim the subject matter which is regarded as the invention.

In FIGURE 1 air under about 4.5 atm. pressure (about 3000 Nm.^{3} per hour) is delivered by conduit **1** to one of the two regenerators **2** and **3** (e.g. **2**) in which it is extremely cooled with separation therefrom of water and carbon dioxide. The extremely cooled and cleansed air is discharged through conduit **4**, and a portion of it (e.g. 1800 Nm.^{3} per hour) passes through conduit **5**, heater **6** in the low pressure fractionating or separating column **7**, conduit **8**, sub-cooler **9**, expansion valve **10** and conduit **11** for delivery as reflux liquid to the fractionating column **7**.

Another portion (e.g. 1200 Nm.^{3} per hour) of the extremely cooled and purified air is passed through conduit **12**, through heat exchanger **13** to be warmed again by the circulating gas which is cooled thereby, then through conduit **14**, expansion turbine **15**, and conduit **16** to be delivered in the gaseous condition to the middle portion of the fractionating column **7**.

From the head of the fractionating column **7**, nitrogen is withdrawn and passed through conduit **17**, countercurrent heat exchanger **9** for cooling the reflux liquid for the fractionating column, then through conduit **18** and through one of the two regenerators **2, 3** (e.g. **3**) for being warmed to the temperature of the surroundings and from where it will be discharged through conduit **19** together with the impurities (e.g. 2630 Nm.^{3} per hour) that were condensed and collected in the regenerator **3** but which are now being revaporized.

From the sump of fractionating column **7** liquid oxygen is drawn off through pipe **20** by means of a pump **21** while being brought to the desired pressure (e.g. 25 atm. gage) and is delivered through pipe **22** to vaporizing heat exchanger **23** and then through pipe **24** to heat exchanger **25** in which it is warmed to the surrounding temperature by the circulating gas. Gaseous oxygen under pressure (e.g. 370 Nm.^{3} per hour) is then delivered by conduit **26**. If desirable, a certain amount of liquid oxygen can also be obtained from conduit **27**.

A circulating gas mixture (e.g. 500 Nm.^{3} per hour) containing about 25% oxygen, about 5% nitrogen and about 70% argon is compressed by the compressor **28** to about 32 atm. gage, and delivered by conduit **29**, in communication with a gas storage cylinder **30** of about 2 m.^{3} capacity to the heat exchanger **25**, then through conduit **31** to the heat exchanger **13**, then through conduit **32** to the heat exchanger **23**, to be at least partially liquified by means of liquid oxygen under pressure while the latter is being vaporized, then through conduit **33** and through expansion valve **34**, to be delivered by conduit **35** directly into the head of the auxiliary column **36** in an at least partly liquefied condition. Into the bottom portion of this auxiliary column a gaseaus mixture of about 94% oxygen, about 2% nitrogen, and about 4% argon (e.g. 500 Nm.^{3} per hour) which was drawn off from the fractionating column **7** below its middle point where air enters from conduit **16**, is introduced through conduit **37**. From the foot or sump of the auxiliary column **36** an oxygen-enriched liquid (e.g. 490 Nm.^{3} per hour) of about 96% oxygen, about 1% nitrogen, and about 3% argon is delivered through conduit **38** into the fractionating column **7** at the same level.

From the head of the auxiliary column **36** a small amount (e.g. 10 Nm.^{3} per hour) of nitrogen-enriched gas of about 10% O_2, 50% N_2 and 40% A is drawn off by conduit **39** so that the circulating gas will not become enriched with nitrogen and other undesired substances. This drawn-off gas after passing through regulating valve **40** and conduit **41** is mixed with the nitrogen in conduit **17** from separating column **7**. From the middle portion of the auxiliary column **36**, vaporized and partially rectified circulating gas is taken off by conduit **42** and after passage through coils **43** in regenerators **2** and **3** to be warmed up by the air that is to be fractionated, is delivered by conduit **44** to compressor **28** which completes the closed circuit.

In this manner the accumulation of undesired components in the circulating gas mixture is effectively prevented, so that the compression of the circulating gas can be accomplished with a minimum amount of energy. Furthermore, in this method by the use of open circulation, it is also possible to replenish the circulating gas with components that have been lost therefrom.

FIGURE 2 differs from FIGURE 1 only in that a double fractionating column **7′**, **7″** is used instead of the single column rectifier. Corresponding parts are designated by the same reference characters as in FIGURE 1.

The branch conduit **5** now leads into the high pressure column **7′** of the double rectifier. Conduit **8′** leads from the sump of the high pressure column **7′** to deliver impure oxygen to the expansion valve **10′** and from there through conduit **11′** into the middle portion of the low pressure column **7″** into the sump of which the head condenser **7‴** of the high pressure column **7′** has been built. Liquefied nitrogen then passes from the upper portion of the high pressure column **7′** through pipe **9′** to the sub-cooling countercurrent heat exchanger **9** and from there through expansion valve **10″** and conduit **11″** into the head of the low pressure column **7″** as reflux fluid. The conduit **16** for expanded air then enters the low pressure column **7″** at an intermediate level between the conduits **11′** and **11″**. The auxiliary column **36** is connected by pipes **37** and **38** to column **7″** at a level somewhat below the level of the conduit **11′**, while pipe **20** delivers liquid oxygen from the sump of the low pressure column **7″**. In all other respects, the system in FIGURE 2 is the same as in FIGURE 1.

The advantages of this invention over a closed circulation with pure argon are:

(1) The temperature for the condensation of the gas mixture according to the invention—caused by its content of oxygen—lies closer to the boiling point of the pure oxygen; therefore a lower compression-pressure for said gas mixture is necessary than for pure argon, in order to vaporize the oxygen.

(2) The fabrication of pure argon in order to sustain a closed circulation is more expensive than the preservation of said gas mixture.

(3) At a closed circulation with indirect heat exchange in an auxiliary column a greater difference of temperature is further to overcome than at the proposed open circulation with direct heat- and mass-exchange in the auxiliary column.

From the foregoing description, one skilled in the art 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. Consequently, such changes and modifications are properly, equitably, and intended to be, within the full range of equivalence of the following claims.

What is claimed is:

1. In a process for producing oxygen by low temperature fractionation, which comprises the after-fractionation steps of withdrawing liquid oxygen from a fractionation column, compressing said liquid oxygen, and vaporizing the resultant compressed liquid oxygen in indirect heat transfer relationship with a compressed circulating gas, the improvement which comprises employing as said gas, a gaseous mixture of 8–30 mol percent oxygen, not more than 10 mol percent nitrogen, and the remainder consisting essentially of argon, said gaseous mixture being at least partially condensed during the vaporization of oxygen.

2. The process of claim **1**, wherein the oxygen content of gaseous mixture is 20–25 mol percent.

3. The process of claim **1**, further comprising the steps of expanding the gaseous mixture into the head of an auxiliary column; passing argon containing gases from said fractionation column to the bottom portion of said auxiliary column, passing oxygen-enriched liquid from the sump of said auxiliary column to said fractionation column, releasing a minor quantity of nitrogen-enriched gas from the head of said auxiliary column; withdrawing from the middle portion of said auxiliary column vaporized and partially rectified circulating gas; passing said partially rectified circulating gas in indirect heat exchange relationship with air to be fractionated in said fractionation column, whereby said partially rectified circulated gas is heated; and compressing the resultant heated partially rectified circulating gas whereby it can be again passed in indirect heat exchange relationship with the oxygen in order to vaporize the same.

4. The process of claim **3**, further comprising the step of combining the minor amount of nitrogen-enriched gas released from the head of the auxiliary column with nitrogen that is withdrawn from the head of the fractionation column.

5. The process of claim **1**, wherein the circulating gas mixture is compressed to 1.2–1.4 times the pressure of the oxygen after the oxygen is compressed, said compression of the circulating gas mixture being accomplished before said mixture is employed to vaporize the oxygen.

6. The process of claim **1**, wherein the amount of circulating gas exceeds the amount of oxygen to be vaporized by 20–50%.

7. The process of claim **1**, wherein the amount of circulating gas exceeds the amount of oxygen to be vaporized by 30–40%.

8. The process of claim **1**, further comprising the steps of passing cool and clean air in indirect heat exchange relationship with the circulating gaseous mixture; expanding the resultant air through a turbine while doing external work; and passing the expanded air to the middle portion of the fractionation column.

9. The process of claim **3**, further comprising the steps of passing cool and clean air in indirect heat exchange relationship with the circulating gaseous mixture; expanding the resultant air through a turbine while doing external work; and passing the expanded air to the middle portion of the fractionation column.

10. The process of claim **9**, further comprising the steps of cooling the compressed circulating gas prior to expanding said gas to the head of the auxiliary column, first by cooling it in indirect heat exchange relationship with preheated compressed oxygen, whereby the oxygen is heated, then with cooled and clean air to be fractionated in the fractionation column, and lastly with the liquid oxygen which has been withdrawn from the fractionation column and compressed to its final pressure, whereby said liquid oxygen is vaporized.

11. Apparatus for producing oxygen by low temperature fractionation, which apparatus comprises:

a compressor having a low pressure side and a high pressure side, for compressing a circulating gas;

indirect heat exchange means including a cold side inlet and outlet and a warm side inlet and outlet;

a conduit connected between the high pressure side of the compressor and the inlet of the warm side of the indirect heat exchange means;

an auxiliary fractionation column for enriching the circulating gas;

expansion means and associated conduit means connecting the outlet of the warm side of the indirect heat exchange means to the head of the auxiliary fractionation column;

first outlet conduit means connected to the head of the auxiliary fractionation column, said outlet conduit means being connected to a regulating valve to permit the controlled release of minor amount of nitrogen-enriched gases;

second outlet conduit means connected to the middle portion of the auxiliary fractionation column;

third outlet conduit means connected to the sump of said auxiliary fractionation column;

inlet conduit means connected to the bottom portion of the auxiliary fractionation column;

a fractionation column for the separation of air, said fractionation column having an outlet and an inlet conduit connected to the middle portion thereof;

conduit means connecting said outlet in said middle portion of said fractionation column with the inlet connected to the auxiliary fractionation column;

conduit means connecting the third outlet from the auxiliary fractionation column to the inlet in said fractionation column;

regenerator means for cooling and cleaning raw gas;

heating coils for circulating gas being imbedded in the regenerators;

conduit means connecting the second outlet from said auxiliary fractionation column to said heating coils imbedded in said regenerator means;

and conduit means connecting said heating coils with said compressor.

12. The apparatus of claim 11, further comprising conduit means connecting the regulating valve at the outlet from the head of the auxiliary fractionation column to the head of the fractionation column, whereby nitrogen-enriched gas from the auxiliary fractionation column can be mixed with nitrogen from the fractionation column; second indirect heat exchange means for conducting heat exchange between cooled and cleaned and at least partially condensed air and said combined nitrogen fractions whereby the cooled and cleaned air is subcooled for use as a reflux liquid for the fractionation column; conduit means connecting the head of the fractionation column with said second indirect heat exchange means; and conduit means connecting the second indirect heat exchange means with the regenerators.

13. The apparatus of claim 12 further comprising third indirect heat exchange means; turbine means; and conduit means connecting the regenerators with the third heat exchange means; conduit means connecting the third heat exchange means with said turbine means; and conduit means connecting said turbine means with the middle portion of the fractionation column, whereby the cooled and cleaned air from the regenerators is warmed by circulating gas expanded in said turbine means and passed to the fractionation column.

14. The apparatus of claim 11, wherein the indirect heat exchange means comprises two heat exchangers; and further comprising third heat exchange means interposed in series between the two heat exchangers with relation to the passage of the circulating gas, said third heat exchange means being connected on the cold side by conduit means connecting the regenerators with the third heat exchange means; and a turbine; and further conduit means connecting the turbine with the middle portion of the fractionation column whereby the cooled and cleaned air is heated in indirect heat transfer relationship with the circulating gas, is further expanded while doing external work, and is then passed to the fractionation column.

15. The apparatus of claim 11, wherein the fractionation column comprises one low pressure column having heating means disposed in the sump thereof, said heating means comprising conduit means being connected on the inlet side to the regenerator means and on the outlet side to the head of the fractionation column by way of an expansion device and an oxygen outlet at the foot of said one low pressure column.

16. The apparatus of claim 11, wherein the fractiontion column comprises a double column having a high pressure lower column and a low pressure upper column, and wherein the condenser for the high pressure column is disposed in the bottom of the low pressure column, whereby it can function as a vaporizer and a condenser; outlet conduit means at the sump of the high pressure column; and wherein the conduit means connecting the auxiliary fractionation column with the fractionation column are connected with the low pressure column; and conduit and expansion valve means connecting the foot of the high pressure column with the upper portion of the low pressure column.

17. The apparatus of claim 11, further comprising gas storage cylinder means connected to the conduit means leading from the compressor to the indirect heat exchange means.

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