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(72) Inventors:
• **TACHIBANA, Hiroshi**
Tokyo 142-8558 (JP)
• **TATSUMI, Takashi**
Tokyo 142-8558 (JP)

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(71) Applicant: **Taiyo Nippon Sanso Corporation**
Tokyo 142-8558 (JP)

(74) Representative: **Hoefer & Partner**
Pilgersheimer Straße 20
81543 München (DE)

(54) **AIR SEPARATION METHOD AND APPARATUS**

(57) Provided is an air separation method and apparatus in which consumed power source can be reduced when oxygen is collected in a three-column process, including:

a first separation step in which feed air is separated into high pressure nitrogen gas and high pressure oxygen-enriched liquid air;

a second separation step in which a high pressure oxygen-enriched liquid air is separated into medium pressure nitrogen gas, and medium pressure oxygen-enriched liquid air;

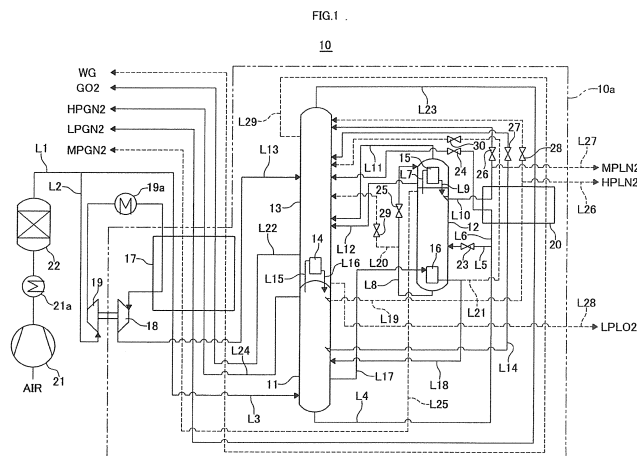
a first indirect heat exchange step in which a medium pressure nitrogen gas is condensed and a low pressure oxygen-enriched liquid air is vaporized by indirect heat exchange;

a third separation step in which a low pressure oxygen-enriched air is separated into low pressure nitrogen gas and low pressure liquid oxygen;

a second indirect heat exchange step in which a high pressure nitrogen gas is condensed and a low pressure liquid oxygen is vaporized by indirect heat exchange;

a third indirect heat exchange step in which a high pressure nitrogen-enriched air in an intermediate stage in the first separation step is condensed and a medium pressure oxygen-enriched liquid air is vaporized by indirect heat exchange; and

a product gas collecting step in which low pressure oxygen gas and/or low pressure liquid oxygen is/are collected as a product oxygen gas after heat recovery.



Description

TECHNICAL FIELD

[0001] The present invention relates to an air separation method and apparatus, and more particularly to an air separation method and apparatus for collecting oxygen gas as a product by cryogenically distilling feed air which is compressed, purified and cooled.

BACKGROUND ART

[0002] As a method of producing a product oxygen gas by cryogenically separating air, a double rectification process is conventionally known as the most popular method. The double rectification process comprises, as main constituting devices: a high pressure column in which compressed, purified and cooled feed air is cryogenically distilled to be separated into high pressure nitrogen gas and high pressure oxygen-enriched liquid air; a low pressure column in which the high pressure oxygen-enriched liquid air is cryogenically distilled after reducing the pressure to be separated into low pressure nitrogen gas and low pressure liquid oxygen; and a main condenser in which the high pressure nitrogen gas at a top portion of the high pressure column is condensed, and at the same time, the low pressure liquid oxygen at a bottom portion of the low pressure column is vaporized. As a process for reducing a consumed power source when a product nitrogen gas in addition to the product oxygen gas is manufactured, a variety of three-column processes are proposed in which a medium pressure column operated at an operating pressure between the operating pressure in the high pressure column and the operating pressure in the low pressure column in a double rectification process is added thereto (see, for example, Patent Documents 1, 2).

PRIOR ART DOCUMENTS

PATENT DOCUMENTS

[0003]

Patent Document 1: JP 8-233457 A
Patent Document 2: JP 2001-263935 A

SUMMARY OF THE INVENTION

PROBLEMS TO BE SOLVED BY THE INVENTION

[0004] In the process described in Patent Document 1, since the oxygen concentration of a fluid provided in the low pressure column becomes high, the rectification condition of the low pressure column can be improved, whereby more high pressure product nitrogen gas can be collected from the column top of the high pressure column compared to the double rectification process. In

the process described in Patent Document 2, although the oxygen concentration of medium pressure oxygen-enriched liquid air withdrawn from a column bottom of the medium pressure column can be made higher than that in a process of Patent Document 1 by lowering the operating pressure of the medium pressure column, since the pressure of the medium pressure nitrogen gas which is collected as a product from a column top of the medium pressure column becomes low, a power source for compressing the medium pressure nitrogen gas is required, and as the result, reduction in the consumed power source has been insufficient.

[0005] Accordingly, an object of the present invention is to provide an air separation method and apparatus in which, by increasing the oxygen concentration of fluid provided in a low pressure column in a three-column process, the amount of nitrogen gas collected which is withdrawn from a column top of a high pressure column or a column top of a medium pressure column can be increased, as well as the consumed power source can be reduced.

MEANS FOR SOLVING THE PROBLEM

[0006] In order to attain the above-mentioned object, in the first constitution of the air separation method of the present invention, an air separation method for cryogenically liquefying and separating feed air to collect product oxygen is characterized by comprising:

a first separation step in which compressed, purified and cooled feed air is cryogenically distilled to be separated into high pressure nitrogen gas and high pressure oxygen-enriched liquid air;

a second separation step in which the high pressure oxygen-enriched liquid air is cryogenically distilled after reducing pressure to be separated into medium pressure nitrogen gas, and medium pressure oxygen-enriched liquid air;

a first indirect heat exchange step in which indirect heat exchange is performed between low pressure oxygen-enriched liquid air which is the medium pressure oxygen-enriched liquid air whose pressure is reduced and the medium pressure nitrogen gas, and the medium pressure nitrogen gas is condensed to obtain a medium pressure liquid nitrogen, and at the same time, the low pressure oxygen-enriched liquid air is vaporized to obtain low pressure oxygen-enriched air;

a third separation step in which the low pressure oxygen-enriched air is cryogenically distilled to be separated into low pressure nitrogen gas and low pressure liquid oxygen;

a second indirect heat exchange step in which indirect heat exchange is performed between the high pressure nitrogen gas and the low pressure liquid oxygen, and the high pressure nitrogen gas is con-

densed to obtain high pressure liquid nitrogen, and at the same time, the low pressure liquid oxygen is vaporized to obtain low pressure oxygen gas; a third indirect heat exchange step in which indirect heat exchange is performed between high pressure nitrogen-enriched air in an intermediate stage in the first separation step and the medium pressure oxygen-enriched liquid air, and the high pressure nitrogen-enriched air is condensed to obtain high pressure nitrogen-enriched liquid air, and at the same time, the medium pressure oxygen-enriched liquid air is vaporized to obtain medium pressure oxygen-enriched air; and a product gas collecting step in which the low pressure oxygen gas and/or the low pressure liquid oxygen is/are collected as a product oxygen gas after heat recovery.

[0007] In the second constitution of the air separation method of the present invention, an air separation method for cryogenically liquefying and separating feed air to collect product oxygen is characterized by comprising:

a first separation step in which compressed, purified and cooled feed air is cryogenically distilled to be separated into high pressure nitrogen gas and high pressure oxygen-enriched liquid air; a second separation step in which the high pressure oxygen-enriched liquid air is cryogenically distilled after reducing pressure to be separated into medium pressure nitrogen gas, and medium pressure oxygen-enriched liquid air; a third separation step in which low pressure oxygen-enriched liquid air which is the medium pressure oxygen-enriched liquid air whose pressure is reduced is cryogenically distilled to be separated into low pressure nitrogen gas and low pressure liquid oxygen; a first indirect heat exchange step in which indirect heat exchange is performed between the medium pressure nitrogen gas, and low pressure oxygen-enriched refluxed liquid air in an intermediate stage in the third separation step, and the medium pressure nitrogen gas is condensed to obtain a medium pressure liquid nitrogen, and at the same time, the low pressure oxygen-enriched refluxed liquid air is vaporized to obtain low pressure oxygen-enriched vaporized air; a second indirect heat exchange step in which indirect heat exchange is performed between the high pressure nitrogen gas and the low pressure liquid oxygen, and the high pressure nitrogen gas is condensed to obtain high pressure liquid nitrogen, and at the same time, the low pressure liquid oxygen is vaporized to obtain low pressure oxygen gas; a third indirect heat exchange step in which indirect heat exchange is performed between high pressure

nitrogen-enriched air in an intermediate stage in the first separation step and the medium pressure oxygen-enriched liquid air, and the high pressure nitrogen-enriched air is condensed to obtain high pressure nitrogen-enriched liquid air, and at the same time, the medium pressure oxygen-enriched liquid air is vaporized to obtain medium pressure oxygen-enriched air; and a product gas collecting step in which the low pressure oxygen gas and/or the low pressure liquid oxygen is/are collected as a product oxygen gas after heat recovery.

[0008] Further, in the air separation method of the present invention, the third indirect heat exchange step can be configured such that in addition to the high pressure nitrogen-enriched liquid air, a high pressure low purity nitrogen gas in which nitrogen is more concentrated is obtained by performing cryogenic distillation in which, while the high pressure nitrogen-enriched air is flowed upward in same passages of a heat integrated distillation device, condensed liquid is flowed downward, and at the same time, in addition to the medium pressure oxygen-enriched air, a medium pressure low purity liquid oxygen in which oxygen is more concentrated is obtained by performing cryogenic distillation in which, while the medium pressure oxygen-enriched liquid air is flowed downward in the other passages of the heat integrated distillation device, vaporized gas is flowed upward. In the third indirect heat exchange step, part of the feed air can be used in place of the high pressure nitrogen-enriched air. In addition, the air separation method of the present invention may include at least one of a high pressure nitrogen gas collecting step in which a high pressure nitrogen gas obtained in the first separation step is collected after heat recovery, a medium pressure nitrogen gas collecting step in which a medium pressure nitrogen gas obtained in the second separation step is collected after heat recovery, a low pressure nitrogen gas collecting step in which a low pressure nitrogen gas obtained in the third separation step is collected after heat recovery, a high pressure liquid nitrogen collecting step in which the high pressure liquid nitrogen condensed in the second indirect heat exchange step is collected, a medium pressure liquid nitrogen collecting step in which the medium pressure liquid nitrogen condensed in the first indirect heat exchange step is collected, and a low pressure liquid oxygen collecting step in which the low pressure liquid oxygen obtained in the third separation step is collected.

[0009] In addition, in the first constitution of the air separation apparatus of the present invention, an air separation apparatus for cryogenically liquefying and separating feed air to collect product oxygen is characterized by comprising:

a high pressure column in which compressed, purified and cooled feed air is cryogenically distilled to be separated into high pressure nitrogen gas and high pressure oxygen-enriched liquid air;

a medium pressure column in which the high pressure oxygen-enriched liquid air is cryogenically distilled after reducing pressure to be separated into medium pressure nitrogen gas, and medium pressure oxygen-enriched liquid air;

a medium pressure column condenser in which indirect heat exchange is performed between low pressure oxygen-enriched liquid air which is the medium pressure oxygen-enriched liquid air whose pressure is reduced and the medium pressure nitrogen gas, and the medium pressure nitrogen gas is condensed to obtain a medium pressure liquid nitrogen, and at the same time, the low pressure oxygen-enriched liquid air is vaporized to obtain low pressure oxygen-enriched air;

a low pressure column in which the low pressure oxygen-enriched air is cryogenically distilled to be separated into low pressure nitrogen gas and low pressure liquid oxygen;

a main condenser reboiler in which indirect heat exchange is performed between the high pressure nitrogen gas and the low pressure liquid oxygen, and the high pressure nitrogen gas is condensed to obtain high pressure liquid nitrogen, and at the same time, the low pressure liquid oxygen is vaporized to obtain low pressure oxygen gas;

a medium pressure column reboiler in which indirect heat exchange is performed between high pressure nitrogen-enriched air taken out from an intermediate portion in the high pressure column and the medium pressure oxygen-enriched liquid air, and the high pressure nitrogen-enriched air is condensed to obtain high pressure nitrogen-enriched liquid air, and at the same time, the medium pressure oxygen-enriched liquid air is vaporized to obtain medium pressure oxygen-enriched air; and

a product oxygen gas collecting path in which the low pressure oxygen gas and/or the low pressure liquid oxygen is/are collected as a product oxygen gas after heat recovery.

[0010] Further, in the second constitution of the air separation apparatus of the present invention, an air separation apparatus for cryogenically liquefying and separating feed air to collect product oxygen is characterized by comprising:

a high pressure column in which compressed, purified and cooled feed air is cryogenically distilled to be separated into high pressure nitrogen gas and high pressure oxygen-enriched liquid air;

a medium pressure column in which the high pressure oxygen-enriched liquid air is cryogenically distilled after reducing pressure to be separated into

medium pressure nitrogen gas, and medium pressure oxygen-enriched liquid air;

a low pressure column in which low pressure oxygen-enriched liquid air which is the medium pressure oxygen-enriched liquid air whose pressure is reduced is cryogenically distilled to be separated into low pressure nitrogen gas and low pressure liquid oxygen;

a medium pressure nitrogen condenser in which indirect heat exchange is performed between the medium pressure nitrogen gas, and low pressure oxygen-enriched refluxed liquid air descending at an intermediate portion of the low pressure column, and the medium pressure nitrogen gas is condensed to obtain medium pressure liquid nitrogen, and at the same time, the low pressure oxygen-enriched refluxed liquid air is vaporized to obtain low pressure oxygen-enriched vaporized air;

a main condenser reboiler in which indirect heat exchange is performed between the high pressure nitrogen gas and the low pressure liquid oxygen, and the high pressure nitrogen gas is condensed to obtain high pressure liquid nitrogen, and at the same time, the low pressure liquid oxygen is vaporized to obtain low pressure oxygen gas;

a medium pressure column reboiler in which indirect heat exchange is performed between high pressure nitrogen-enriched air taken out from an intermediate portion of the high pressure column and the medium pressure oxygen-enriched liquid air, and the high pressure nitrogen-enriched air is condensed to obtain high pressure nitrogen-enriched liquid air, and at the same time, the medium pressure oxygen-enriched liquid air is vaporized to obtain medium pressure oxygen-enriched air; and

a product gas collecting path in which the low pressure oxygen gas and/or the low pressure liquid oxygen is/are collected as a product oxygen gas after heat recovery.

[0011] In the air separation apparatus of the present invention, in place of the medium pressure column reboiler, a heat integrated distillation device in which in addition to the high pressure nitrogen-enriched liquid air, a high pressure low purity nitrogen gas in which nitrogen is more concentrated is obtained by performing cryogenic distillation in which, while part of the high pressure nitrogen-enriched air is flowed upward in same passages of a heat integrated distillation device, condensed liquid is flowed downward, and at the same time, in addition to the medium pressure oxygen-enriched air, a medium pressure low purity liquid oxygen in which oxygen is more concentrated is obtained by performing cryogenic distillation in which, while the medium pressure oxygen-enriched liquid air is flowed downward in the other passage, vaporized gas is flowed upward can be used. In the heat integrated distillation device, part of the feed air can be used in place of the high pressure nitrogen-

enriched air. Further, the air separation apparatus of the present invention may include at least one of
 a high pressure nitrogen gas collecting path in which a high pressure nitrogen gas obtained in the high pressure column is collected after heat recovery,
 a medium pressure nitrogen gas collecting path in which a medium pressure nitrogen gas obtained in the medium pressure column is collected after heat recovery,
 a low pressure nitrogen gas collecting path in which a low pressure nitrogen gas obtained in the low pressure column is collected after heat recovery,
 a high pressure liquid nitrogen collecting path in which the high pressure liquid nitrogen condensed in the main condenser reboiler is collected,
 a medium pressure liquid nitrogen collecting path in which the medium pressure liquid nitrogen condensed in the medium pressure column condenser is collected, and
 a low pressure liquid oxygen collecting path in which the low pressure liquid oxygen obtained in the low pressure column is collected.

ADVANTAGES OF THE INVENTION

[0012] Since, according to the present invention, the oxygen concentration of low pressure oxygen-enriched air or low pressure oxygen-enriched liquid air provided in a low pressure column in which a third separation step is performed, or low pressure oxygen-enriched vaporized air vaporized inside the low pressure column can be made high, rectification conditions of the low pressure column can be improved, and exchanged heat of a main condenser reboiler in which a second indirect heat exchange step is performed can be made small. By this, the withdrawn amount of high pressure nitrogen gas separated in a high pressure column in which a first separation step is performed or medium pressure nitrogen gas separated in a medium pressure column in which a second separation step is performed can be made large. Therefore, when only product oxygen gas is collected, the consumed power source for the whole apparatus can be reduced by expanding high pressure nitrogen gas or medium pressure nitrogen gas to recover a power source. When high pressure nitrogen gas or medium pressure nitrogen gas is collected as a product nitrogen gas, since a large amount of high pressure nitrogen gas can be collected, reduction in equipment cost due to miniaturization of a nitrogen compressor for transmitting nitrogen gas or reduction in power source cost can be attained.

BRIEF DESCRIPTION OF THE DRAWINGS

[0013]

Fig. 1 is a system diagram illustrating a first embodiment of an air separation apparatus to which an air separation method according to the present invention is applied.

Fig. 2 is a system diagram illustrating a second embodiment of an air separation apparatus to which an air separation method according to the present invention is applied.

Fig. 3 is a system diagram illustrating a third embodiment of an air separation apparatus to which an air separation method according to the present invention is applied.

Fig. 4 is a system diagram illustrating a fourth embodiment of an air separation apparatus to which an air separation method according to the present invention is applied.

Mode for Carrying out the Invention

[0014] Fig. 1 illustrates a first embodiment of the present invention. An air separation apparatus 10 illustrated in the present embodiment employs a three-column process, and is provided with, as main devices, a high pressure column 11, a medium pressure column 12, a low pressure column 13, a main condenser reboiler 14, a medium pressure column condenser 15, a medium pressure column reboiler 16, a main heat exchanger 17, an expansion turbine 18 and a blower 19 driven by the expansion turbine 18 and a subcooler 20, wherein the main condenser reboiler 14 is arranged between the high pressure column 11 and the low pressure column 13, the medium pressure column condenser 15 is arranged on an upper portion of the medium pressure column 12, and the medium pressure column reboiler 16 is arranged on a bottom portion of the medium pressure column 12.

[0015] By appropriately setting operating conditions and performing the following steps, in the air separation apparatus, oxygen gas (GO₂), high pressure nitrogen gas (HPGN₂), medium pressure nitrogen gas (MPGN₂), low pressure nitrogen gas (LPGN₂), high pressure liquid nitrogen (HPLN₂), medium pressure liquid nitrogen (MPLN₂) and low pressure liquid oxygen (LPLO₂) can be collected as products.

[0016] The feed air (AIR) is compressed to a preset high pressure in an air compressor 21 and the heat of compression is removed in an air precooler 21a, and then, impurities in the air is removed in an air purifier 22 to be purified. Part of the feed air withdrawn from the air purifier 22 to a path L1 is diverged into a path L2, and the pressure thereof is increased in the blower 19, and then, the air is cooled in a blower aftercooler 19a to enter a cold box 10a, and introduced into the main heat exchanger 17. Large part of feed air directly flows in a path L1 to enter the cold box 10a, and is cooled to a preset temperature in the main heat exchanger 17, then passes through a path L3 to be introduced into the high pressure column 11.

[0017] In the high pressure column 11, a first separation step is performed in which the feed air is cryogenically distilled to be separated into high pressure nitrogen gas in a column top portion and high pressure oxygen-enriched liquid air in column bottom portion. High pres-

sure oxygen-enriched liquid air taken out from a bottom portion of the high pressure column 11 to a path L4 is diverged into a path L5 and a path L6. The pressure of the high pressure oxygen-enriched liquid air in the path L5 is reduced to a preset medium pressure at a pressure reducing valve 23, and then the liquid air is introduced into the medium pressure column 12. The high pressure oxygen-enriched liquid air in the path L6 is cooled in the subcooler 20, and then the pressure thereof is reduced to a preset low pressure at a pressure reducing valve 24, and then, the liquid air is introduced into the low pressure column 13.

[0018] In the medium pressure column 12, a second separation step is performed in which high pressure oxygen-enriched liquid air whose pressure is reduced to an medium pressure and which has been introduced thereinto is cryogenically distilled to be separated into medium pressure nitrogen gas at a column top portion and medium pressure oxygen-enriched liquid air at a column bottom portion. To a medium pressure column condenser 15 provided at an upper portion of the medium pressure column 12, medium pressure nitrogen gas taken out from a top portion of the medium pressure column 12 to a path L7, and low pressure oxygen-enriched liquid air which is part of the medium pressure oxygen-enriched liquid air taken out from a bottom portion of the medium pressure column 12 to a path L8 whose pressure has been reduced at a pressure reducing valve 25 are introduced.

[0019] In the medium pressure column condenser 15, a first indirect heat exchange step is performed in which indirect heat exchange is performed between low pressure oxygen-enriched liquid air, and the medium pressure nitrogen gas, and the medium pressure nitrogen gas is condensed to obtain a medium pressure liquid nitrogen, and at the same time, low pressure oxygen-enriched liquid air is vaporized to obtain low pressure oxygen-enriched air. Medium pressure liquid nitrogen which has been condensed in the medium pressure column condenser 15 passes through a path L9 to be returned to the medium pressure column 12, which turns into descending liquid in the medium pressure column 12. Part of medium pressure liquid nitrogen is taken out to a path L10 and cooled in the subcooler 20, and then the pressure thereof is reduced at a pressure reducing valve 26 and the liquid nitrogen is introduced to the low pressure column 13. Low pressure oxygen-enriched air vaporized at medium pressure column condenser 15 passes through a path L11 and introduced into the low pressure column 13. Part of low pressure oxygen-enriched liquid air which has not been vaporized in the medium pressure column condenser 15 is taken out to a path L12 to be introduced into the low pressure column 13.

[0020] To the low pressure column 13, in addition to a variety of fluids from the paths L6, L10, L11 and L12, feed air whose pressure is increased at the blower 19 and which is cooled at the main heat exchanger 17 and then adiabatically-expanded in the expansion turbine 18 from

a path L13 and a liquid fluid which is part of descending liquid in the high pressure column 11 taken out from an intermediate portion and cooled in the subcooler 20 and whose pressure is reduced at a pressure reducing valve 27 from a path L14 are individually introduced. In the low pressure column 13, a third separation step is performed in which these fluids whose main component is the low pressure oxygen-enriched air are cryogenically distilled to be separated into low pressure nitrogen gas in a column top portion and low pressure liquid oxygen in a column bottom portion.

[0021] In the main condenser reboiler 14, a second indirect heat exchange step is performed in which indirect heat exchange is performed between a low pressure liquid oxygen at a bottom portion of the low pressure column 13 and the high pressure nitrogen gas taken out from a top portion of the high pressure column 11 to a path L15, and the high pressure nitrogen gas is condensed to obtain high pressure liquid nitrogen, and at the same, the low pressure liquid oxygen is vaporized to obtain low pressure oxygen gas. High pressure liquid nitrogen which has been condensed in the main condenser reboiler 14 passes through a path L16 to be returned to a high pressure column 11, which turns into descending liquid in the high pressure column 11.

[0022] On the other hand, in the medium pressure column reboiler 16, a third indirect heat exchange step is performed in which indirect heat exchange is performed between the remainder of the medium pressure oxygen-enriched liquid air separated at a bottom portion of the medium pressure column 12 and part of high pressure nitrogen-enriched air taken out from a lower portion of the high pressure column 11 in an intermediate stage of a first separation step in the high pressure column 11 to a path L17, and the high pressure nitrogen-enriched air is condensed to obtain high pressure nitrogen-enriched liquid air, and at the same time, the medium pressure oxygen-enriched liquid air is vaporized to obtain medium pressure oxygen-enriched air. Medium pressure oxygen-enriched air vaporized in the medium pressure column reboiler 16 turns into ascending gas in the medium pressure column 12. High pressure nitrogen-enriched liquid air which has been condensed in the medium pressure column reboiler 16 passes through a path L18 to be returned to the high pressure column 11, which turns into descending liquid in the high pressure column 11.

[0023] It is also possible that: part of high pressure liquid nitrogen which has been introduced into the main condenser reboiler 14 from a top portion of the high pressure column 11 and condensed is taken out to a path L19 and cooled in the subcooler 20, whose pressure is reduced at a pressure reducing valve 28 and then the liquid nitrogen is introduced into the low pressure column 13; that part of medium pressure oxygen-enriched liquid air flowing in the path L8 is diverged into a path L20, and the pressure of the liquid air is reduced at a pressure reducing valve 29, and then the liquid air is introduced into low pressure column 13; or that part of high pressure

nitrogen-enriched liquid air flowing in a path L18 is diverged into a path L21, the liquid air is cooled in the subcooler 20, and the pressure of the liquid air is reduced at a pressure reducing valve 30, and then, the liquid air is introduced into the low pressure column 13.

[0024] Part of low pressure oxygen gas which has been vaporized in the main condenser reboiler 14 is taken out to a product oxygen gas collecting path L22 and undergoes heat recovery in the main heat exchanger 17, and then collected as a product oxygen gas (GO2). The remainder of low pressure oxygen gas turns into ascending gas in the low pressure column 13. Low pressure nitrogen gas is taken out from a top portion of the low pressure column 13 to a low pressure nitrogen gas collecting path L23 and undergoes heat recovery in the subcooler 20 and the main heat exchanger 17, and then collected as a product low pressure nitrogen gas (LPGN2). Further, part of high pressure nitrogen gas is taken out from a top portion of the high pressure column 11 to a high pressure nitrogen gas collecting path L24 and undergoes heat recovery in the main heat exchanger 17, and then collected as a product high pressure nitrogen gas (HPGN2).

[0025] In addition, as collected, a medium pressure nitrogen gas collecting path L25 in which part of medium pressure nitrogen gas obtained in the medium pressure column 12 undergoes heat recovery and then collected as a product medium pressure nitrogen gas (MPGN2), a high pressure liquid nitrogen collecting path L26 in which part of high pressure liquid nitrogen which has been condensed in the main condenser reboiler 14 is collected as a product high pressure liquid nitrogen (HPLN2), a medium pressure liquid nitrogen collecting path L27 in which part of medium pressure liquid nitrogen which has been condensed in the medium pressure column condenser 15 and collected as a product medium pressure liquid nitrogen (MPLN2), a low pressure liquid oxygen collecting path L28 in which part of low pressure liquid oxygen obtained in the low pressure column 11 is collected as a product low pressure liquid oxygen (LPLO2) can be provided, and a medium pressure nitrogen gas collecting step, a high pressure liquid nitrogen collecting step, a medium pressure liquid nitrogen collecting step, a low pressure liquid oxygen collecting step can be individually performed. Further, from an upper portion of the low pressure column 13, waste gas (WG) can be taken out to a path L29.

[0026] In the air separation apparatus 10 configured in such a manner, in particular, by making the temperature of medium pressure oxygen-enriched liquid air higher than the temperature of the low pressure liquid oxygen at a bottom portion of the low pressure column 13 by taking out from an intermediate portion of a high pressure column 11 and using, as warm fluid for vaporizing medium pressure oxygen-enriched liquid air separated at a bottom portion of medium pressure column 12, high pressure nitrogen-enriched air whose oxygen concentration and temperature are higher than those of high pressure nitrogen gas in the high pressure column 11, preferably

whose oxygen concentration is 8 mole % or higher, more preferably 11 mole % or higher, the oxygen concentration of medium pressure oxygen-enriched liquid air taken out from a bottom portion of the medium pressure column 12 to a path L8, and the oxygen concentration of low pressure oxygen-enriched air which is obtained by vaporizing, after reducing pressure, medium pressure oxygen-enriched liquid air in the medium pressure column condenser 15 can be made high.

[0027] Accordingly, since, by performing the above-mentioned steps, the oxygen concentration of the low pressure oxygen-enriched air provided via the path L11 to the low pressure column 13 can be made high, the rectification condition of the low pressure column 13 can be improved, whereby exchanged heat of the main condenser reboiler 14 can be made small while reducing the consumed power source. By this, the amount of high pressure nitrogen gas which can be withdrawn from a top portion of the high pressure column 11 or medium pressure nitrogen gas which can be withdrawn from a top portion of the medium pressure column 12 can be increased. Consumed power source of the apparatus can be reduced by expanding these high pressure and/or medium pressure nitrogen gas to recover power source, for example, by using power source which has expanded nitrogen gas for the air compressor 21. When these high pressure and/or medium pressure nitrogen gas is/are compressed and transferred as a product, reduction in consumed power source of the nitrogen compressor and miniaturization can be attained.

[0028] Fig. 2 illustrates a second embodiment of the present invention. In the following description, detailed description will be omitted by assigning the same reference sign as that of the component in the air separation apparatus shown in the first embodiment to the same component as in the first embodiment.

[0029] The air separation apparatus in the present embodiment illustrates an example in which, in place of the high pressure nitrogen-enriched air taken out from an intermediate portion of the high pressure column 11 into a path 17, part of feed air cooled in the main heat exchanger 17 is used as warm fluid which is to be introduced into the medium pressure column reboiler 16 in the first embodiment.

[0030] Part of feed air cooled in the main heat exchanger 17 is diverged from the path L3 to a path L31 to be introduced into the medium pressure column reboiler 16. Large part of remainder feed air directly passes through the path L3 to be introduced into the high pressure column 11. In the medium pressure column reboiler 16, the same step as in the third indirect heat exchange step is performed in which indirect heat exchange is performed between feed air from the path L31 and the medium pressure oxygen-enriched liquid air separated at a column bottom portion of the medium pressure column 12, and the medium pressure oxygen-enriched liquid air is vaporized to obtain medium pressure oxygen-enriched air, and at the same time, feed air is condensed to obtained

feed liquid air. Medium pressure oxygen-enriched air vaporized in the medium pressure column reboiler 16 turns to ascending gas in the medium pressure column 12 in the same manner. Feed liquid air which has been condensed in the medium pressure column reboiler 16 passes through a path L32 to be introduced into the high pressure column 11 as descending liquid.

[0031] Also in the present embodiment, a product oxygen gas is collected from a product oxygen gas collecting path L22; a product low pressure nitrogen gas is collected from a low pressure nitrogen gas collecting path L23; and a product high pressure nitrogen gas is collected from a high pressure nitrogen gas collecting path L24. As mentioned above, although the illustration thereof is omitted, a product medium pressure nitrogen gas, a product high pressure liquid nitrogen, a product medium pressure liquid nitrogen and a product low pressure liquid oxygen can be collected or waste gas can also be taken out.

[0032] As illustrated in the present embodiment, by using part of feed air as warm fluid by which indirect heat exchange is performed with medium pressure oxygen-enriched liquid air in the medium pressure column reboiler 16, the composition of the warm fluid in the medium pressure column reboiler 16 can be made constant and the temperature of the warm fluid can be made stable, whereby the load fluctuation of the medium pressure column reboiler 16 can be prevented and the operability of the medium pressure column 12 can be improved. Accordingly, even when the concentration profile of the high pressure column 11 varies by a disturbance, the medium pressure column 12 can be retained in a stable operating condition.

[0033] Fig. 3 illustrates a third embodiment of the present invention and illustrates an example in which a heat integrated distillation device 31 is used in place of the medium pressure column reboiler 16. The heat integrated distillation device 31 comprises first passages 32 in which a gaseous warm fluid flows upward and second passages 33 in which a liquid cold fluid flows as descending flow. In the embodiment, feed air is introduced to first passages 32 and medium pressure oxygen-enriched liquid air is introduced to second passages 32 respectively.

[0034] Namely, part of feed air diverted from the path L3 heading for the high pressure column 11 to a path L31 is introduced from a lower portion of the heat integrated distillation device 31 to first passages 32 as a upward flow, and performing indirect heat exchange with medium pressure oxygen-enriched liquid air flowing in second passages 33 while flowing upward in the first passages 32. In this indirect heat exchange, part of feed air is condensed. By the downward flow thereof in the first passages 32, cryogenic distillation is performed in the first passages 32, and nitrogen in a gas ascending in the first passages 32 is concentrated and oxygen in liquid flowing downward in the first passages 32 is concentrated. Gas (high pressure low purity nitrogen gas) which is obtained by concentration of nitrogen ascending in the first pas-

sages 32 is taken out to a path L34, and passes through a valve 34 to be introduced into the high pressure column 11. The liquid in which oxygen is concentrated which flows downward in the first passages 32 is taken out to a path L35, and is joined with high pressure oxygen-enriched liquid air taken out from a bottom portion of the high pressure column 11 and flowing in the path L4. The valve 34 may be provided on the primary side of the first passages 32 of the heat integrated distillation device 31.

[0035] On the other hand, medium pressure oxygen-enriched liquid air taken out from a bottom portion of the medium pressure column 12 to a path L36 is introduced from an upper portion of the heat integrated distillation device 31 to second passages 33 as a downward flow, and indirect heat exchange is performed with feed air flowing in the first passages 32 while flowing downward in the second passages 33. By this indirect heat exchange, part of medium pressure oxygen-enriched liquid air is vaporized, and by upward flow thereof in the second passages 33, cryogenic distillation is performed in the second passages 33, whereby nitrogen in gas ascending in the second passages 33 is concentrated and oxygen in liquid flowing downward in the second passages 33 is concentrated. Gas obtained by concentration of nitrogen ascending in the second passages 33 is taken out to a path L37 to be introduced into the medium pressure column 12 as ascending gas. Liquid (medium pressure low purity liquid oxygen) obtained by concentration of oxygen flowing downward in the second passages 33 is taken out to a path L38, and the pressure of the liquid is reduced to a low pressure at the pressure reducing valve 35, and the liquid is introduced into the medium pressure column condenser 15. Further, liquid obtained by concentration of oxygen is, as collected, diverged from a path L38 to a path L39, and the pressure of the liquid is reduced to a low pressure at the pressure reducing valve 36, and the liquid can be introduced into the low pressure column 13.

[0036] In such a manner, since gas obtained by concentration of nitrogen in the first passages 32 can be provided into the high pressure column 11 by using the heat integrated distillation device 31 in place of the medium pressure column reboiler 16, the rectification condition of the high pressure column 11 can be improved, as well as, liquid obtained by concentration of oxygen in the second passages 33 can be introduced into the medium pressure column condenser 15 and vaporized gas can be provided into the low pressure column 13, thereby also improving the rectification condition of the low pressure column 13. In place of feed air, high pressure nitrogen-enriched air taken out from an intermediate portion of the high pressure column 11 can be used as warm fluid in the same manner as in the first embodiment. Also in this case, it is possible that: gas obtained by concentration of nitrogen in the first passages 32 and condensed liquid are returned to the high pressure column 11; or that part of condensed liquid is diverged and cooled in the subcooler 20, and then introduced to the low pressure column 13 after reducing pressure. Also in the present

embodiment, a variety of gas products or liquid products can be collected in the same manner as in the first embodiment.

[0037] Fig. 4 illustrates a fourth embodiment of the present invention and illustrates an example in which low pressure oxygen-enriched refluxed liquid air flowing downward at an intermediate portion of the low pressure column 13 as cold fluid for condensing medium pressure nitrogen gas at a top portion of the medium pressure column 12.

[0038] In an intermediate portion of the low pressure column 13 on the lower side from the position where a variety of fluids flow into the low pressure column 13, a medium pressure nitrogen condenser 41 is provided. Medium pressure nitrogen gas taken out from a top portion of the medium pressure column 12 to path L41 is introduced into the medium pressure nitrogen condenser 41. By performing indirect heat exchange between the medium pressure nitrogen gas, and part of low pressure oxygen-enriched refluxed liquid air flowing down inside the low pressure column 13, the medium pressure nitrogen gas is condensed to turn into medium pressure liquid nitrogen, as well as, low pressure oxygen-enriched refluxed liquid air is vaporized to obtain low pressure oxygen-enriched vaporized air. Condensed medium pressure liquid nitrogen passes through a path L42 to be introduced into an upper portion of the medium pressure column 12 as descending liquid. Part of medium pressure liquid nitrogen is diverged into a path L43 and passes the subcooler 20, and the pressure thereof is reduced at the pressure reducing valve 42, and then the liquid nitrogen is introduced into the low pressure column 13. Further, part of medium pressure liquid nitrogen which becomes in a supercooled state in the subcooler 20 may be diverged into a path L44 to be collected as a product medium pressure liquid nitrogen. Vaporized low pressure oxygen-enriched vaporized air becomes ascending gas in the low pressure column 13. On the other hand, the pressure of medium pressure oxygen-enriched liquid air taken out from a bottom portion of the medium pressure column 12 to a path L45 is reduced at a pressure reducing valve 43 and the liquid air is introduced into the low pressure column 13.

[0039] As mentioned above, by using, as a cold fluid for condensing medium pressure nitrogen gas at a top portion of the medium pressure column 12, low pressure oxygen-enriched refluxed liquid air flowing downward at an intermediate portion of the low pressure column 13, the position of medium pressure nitrogen condenser 41 in the low pressure column 13 is adjusted, whereby the composition of low pressure oxygen-enriched refluxed liquid air can be arbitrarily selected and flexibility of operation and design can be enhanced and the air separation efficiency can be improved.

[0040] In each embodiment, when a high pressure product oxygen gas is collected, in place of collecting low pressure oxygen gas vaporized in the main condenser reboiler 14, low pressure liquid oxygen is taken out from

a bottom portion of the low pressure column 13, the pressure of low pressure liquid oxygen is increased to a desired pressure by a liquid oxygen pump to obtain high pressure liquid oxygen, and then the liquid oxygen is vaporized at the main heat exchanger 17, whereby a product high pressure oxygen gas can be collected. By this, there is no need to provide an expensive oxygen compressor, and rise in the equipment cost can be suppressed. Any type of a heat exchanger can be employed which is used for the main condenser reboiler, the medium pressure column condenser, the medium pressure column reboiler, or the like in which indirect heat exchange is performed for a variety of fluid, and a variety of types of heat exchanger can be employed.

REFERENCE NUMERALS

[0041] 10 ... air separation apparatus, 10a ... cold box, 11 ... high pressure column, 12 ... medium pressure column, 13 ... low pressure column, 14 ... main condenser reboiler, 15 ... medium pressure column condenser, 16 ... medium pressure column reboiler, 17 ... main heat exchanger, 18 ... expansion turbine, 19 ... blower, 19a ... blower aftercooler, 20 ... subcooler, 21 ... air compressor, 21a ... air precooler, 22 ... air purifier, 23, 24, 25, 26, 27, 28, 29, 30 ... pressure reducing valve, 31 ... heat integrated distillation device, 32 ... first passages, 33 ... second passages, 34 ... valve, 35, 36, 37 ... pressure reducing valve, 41 ... medium pressure nitrogen condenser, 42, 43 ... pressure reducing valve

Claims

1. An air separation method for cryogenically liquefying and separating feed air to collect product oxygen comprising:

a first separation step in which compressed, purified and cooled feed air is cryogenically distilled to be separated into high pressure nitrogen gas and high pressure oxygen-enriched liquid air;
a second separation step in which the high pressure oxygen-enriched liquid air is cryogenically distilled after reducing pressure to be separated into medium pressure nitrogen gas, and medium pressure oxygen-enriched liquid air;
a first indirect heat exchange step in which indirect heat exchange is performed between low pressure oxygen-enriched liquid air which is the medium pressure oxygen-enriched liquid air whose pressure is reduced and the medium pressure nitrogen gas, and the medium pressure nitrogen gas is condensed to obtain a medium pressure liquid nitrogen, and at the same time, the low pressure oxygen-enriched liquid air is vaporized to obtain low pressure oxygen-enriched air;

- a third separation step in which the low pressure oxygen-enriched air is cryogenically distilled to be separated into low pressure nitrogen gas and low pressure liquid oxygen;
- a second indirect heat exchange step in which indirect heat exchange is performed between the high pressure nitrogen gas and the low pressure liquid oxygen, and the high pressure nitrogen gas is condensed to obtain high pressure liquid nitrogen, and at the same time, the low pressure liquid oxygen is vaporized to obtain low pressure oxygen gas;
- a third indirect heat exchange step in which indirect heat exchange is performed between high pressure nitrogen-enriched air in an intermediate stage in the first separation step and the medium pressure oxygen-enriched liquid air, and the high pressure nitrogen-enriched air is condensed to obtain high pressure nitrogen-enriched liquid air, and at the same time, the medium pressure oxygen-enriched liquid air is vaporized to obtain medium pressure oxygen-enriched air; and
- a product gas collecting step in which the low pressure oxygen gas and/or the low pressure liquid oxygen is/are collected as a product oxygen gas after heat recovery.
2. An air separation method for cryogenically liquefying and separating feed air to collect product oxygen comprising:
- a first separation step in which compressed, purified and cooled feed air is cryogenically distilled to be separated into high pressure nitrogen gas and high pressure oxygen-enriched liquid air;
- a second separation step in which the high pressure oxygen-enriched liquid air is cryogenically distilled after reducing pressure to be separated into medium pressure nitrogen gas, and medium pressure oxygen-enriched liquid air;
- a third separation step in which low pressure oxygen-enriched liquid air which is the medium pressure oxygen-enriched liquid air whose pressure is reduced is cryogenically distilled to be separated into low pressure nitrogen gas and low pressure liquid oxygen;
- a first indirect heat exchange step in which indirect heat exchange is performed between the medium pressure nitrogen gas, and low pressure oxygen-enriched refluxed liquid air in an intermediate stage in the third separation step, and the medium pressure nitrogen gas is condensed to obtain a medium pressure liquid nitrogen, and at the same time, the low pressure oxygen-enriched refluxed liquid air is vaporized to obtain low pressure oxygen-enriched vaporized air;
- a second indirect heat exchange step in which indirect heat exchange is performed between the high pressure nitrogen gas and the low pressure liquid oxygen, and the high pressure nitrogen gas is condensed to obtain high pressure liquid nitrogen, and at the same time, the low pressure liquid oxygen is vaporized to obtain low pressure oxygen gas;
- a third indirect heat exchange step in which indirect heat exchange is performed between high pressure nitrogen-enriched air in an intermediate stage in the first separation step and the medium pressure oxygen-enriched liquid air, and the high pressure nitrogen-enriched air is condensed to obtain high pressure nitrogen-enriched liquid air, and at the same time, the medium pressure oxygen-enriched liquid air is vaporized to obtain medium pressure oxygen-enriched air; and
- a product gas collecting step in which the low pressure oxygen gas and/or the low pressure liquid oxygen is/are collected as a product oxygen gas after heat recovery.
3. The air separation method according to claim 1 or 2, wherein, in the third indirect heat exchange step, in addition to the high pressure nitrogen-enriched liquid air, a high pressure low purity nitrogen gas in which nitrogen is more concentrated is obtained by performing cryogenic distillation in which, while the high pressure nitrogen-enriched air is flowed upward in same passages of a heat integrated distillation device, condensed liquid is flowed downward, and at the same time,
- in addition to the medium pressure oxygen-enriched air, a medium pressure low purity liquid oxygen in which oxygen is more concentrated is obtained by performing cryogenic distillation in which, while the medium pressure oxygen-enriched liquid air is flowed downward in the other passages of the heat integrated distillation device, vaporized gas is flowed upward.
4. The air separation method according to claims 1 to 3, wherein, in the third indirect heat exchange step, part of the feed air can be used in place of the high pressure nitrogen-enriched air.
5. The air separation method according to claims 1 to 4, wherein the air separation method of the present invention comprises at least one of
- a high pressure nitrogen gas collecting step in which a high pressure nitrogen gas obtained in the first separation step is collected after heat recovery,
- a medium pressure nitrogen gas collecting step in which a medium pressure nitrogen gas obtained in the second separation step is collected after heat recovery,

a low pressure nitrogen gas collecting step in which a low pressure nitrogen gas obtained in the third separation step is collected after heat recovery,
 a high pressure liquid nitrogen collecting step in which the high pressure liquid nitrogen condensed in the second indirect heat exchange step is collected,
 a medium pressure liquid nitrogen collecting step in which the medium pressure liquid nitrogen condensed in the first indirect heat exchange step is collected, and
 a low pressure liquid oxygen collecting step in which the low pressure liquid oxygen obtained in the third separation step is collected.

6. An air separation apparatus for cryogenically liquefying and separating feed air to collect product oxygen comprising:

a high pressure column in which compressed, purified and cooled feed air is cryogenically distilled to be separated into high pressure nitrogen gas and high pressure oxygen-enriched liquid air;

a medium pressure column in which the high pressure oxygen-enriched liquid air is cryogenically distilled after reducing pressure to be separated into medium pressure nitrogen gas, and medium pressure oxygen-enriched liquid air;

a medium pressure column condenser in which indirect heat exchange is performed between low pressure oxygen-enriched liquid air which is the medium pressure oxygen-enriched liquid air whose pressure is reduced and the medium pressure nitrogen gas, and the medium pressure nitrogen gas is condensed to obtain a medium pressure liquid nitrogen, and at the same time, the low pressure oxygen-enriched liquid air is vaporized to obtain low pressure oxygen-enriched air;

a low pressure column in which the low pressure oxygen-enriched air is cryogenically distilled to be separated into low pressure nitrogen gas and low pressure liquid oxygen;

a main condenser reboiler in which indirect heat exchange is performed between the high pressure nitrogen gas and the low pressure liquid oxygen, and the high pressure nitrogen gas is condensed to obtain high pressure liquid nitrogen, and at the same time, the low pressure liquid oxygen is vaporized to obtain low pressure oxygen gas;

a medium pressure column reboiler in which indirect heat exchange is performed between high pressure nitrogen-enriched air taken out from an intermediate portion in the high pressure column and the medium pressure oxygen-enriched liquid air, and the high pressure nitrogen-en-

riched air is condensed to obtain high pressure nitrogen-enriched liquid air, and at the same time, the medium pressure oxygen-enriched liquid air is vaporized to obtain medium pressure oxygen-enriched air; and

a product oxygen gas collecting path in which the low pressure oxygen gas and/or the low pressure liquid oxygen is/are collected as a product oxygen gas after heat recovery.

7. An air separation apparatus for cryogenically liquefying and separating feed air to collect product oxygen comprising:

a high pressure column in which compressed, purified and cooled feed air is cryogenically distilled to be separated into high pressure nitrogen gas and high pressure oxygen-enriched liquid air;

a medium pressure column in which the high pressure oxygen-enriched liquid air is cryogenically distilled after reducing pressure to be separated into medium pressure nitrogen gas, and medium pressure oxygen-enriched liquid air;

a low pressure column in which low pressure oxygen-enriched liquid air which is the medium pressure oxygen-enriched liquid air whose pressure is reduced is cryogenically distilled to be separated into low pressure nitrogen gas and low pressure liquid oxygen;

a medium pressure nitrogen condenser in which indirect heat exchange is performed between the medium pressure nitrogen gas, and low pressure oxygen-enriched refluxed liquid air descending at an intermediate portion of the low pressure column, and the medium pressure nitrogen gas is condensed to obtain medium pressure liquid nitrogen, and at the same time, the low pressure oxygen-enriched refluxed liquid air is vaporized to obtain low pressure oxygen-enriched vaporized air;

a main condenser reboiler in which indirect heat exchange is performed between the high pressure nitrogen gas and the low pressure liquid oxygen, and the high pressure nitrogen gas is condensed to obtain high pressure liquid nitrogen, and at the same time, the low pressure liquid oxygen is vaporized to obtain low pressure oxygen gas;

a medium pressure column reboiler in which indirect heat exchange is performed between high pressure nitrogen-enriched air taken out from an intermediate portion of the high pressure column and the medium pressure oxygen-enriched liquid air, and the high pressure nitrogen-enriched air is condensed to obtain high pressure nitrogen-enriched liquid air, and at the same time, the medium pressure oxygen-enriched liq-

uid air is vaporized to obtain medium pressure oxygen-enriched air; and
 a product gas collecting path in which the low pressure oxygen gas and/or the low pressure liquid oxygen is/are collected as a product oxygen gas after heat recovery. 5

8. The air separation apparatus according to claim 6 or 7, wherein a heat integrated distillation device in which 10
 in addition to the high pressure nitrogen-enriched liquid air, a high pressure low purity nitrogen gas in which nitrogen is more concentrated is obtained by performing cryogenic distillation in which, while part of the high pressure nitrogen-enriched air is flowed upward in same passages of a heat integrated distillation device, condensed liquid is flowed downward, and at the same time, 15
 in addition to the medium pressure oxygen-enriched air, a medium pressure low purity liquid oxygen in which oxygen is more concentrated is obtained by performing cryogenic distillation in which, while the medium pressure oxygen-enriched liquid air is flowed downward in the other passage, vaporized gas is flowed upward is used. 20 25
9. The air separation apparatus according to claims 6 to 8, wherein part of the feed air is used in place of the high pressure nitrogen-enriched air. 30
10. The air separation apparatus according to claims 6 to 9, comprising at least one of
 a high pressure nitrogen gas collecting path in which a high pressure nitrogen gas obtained in the high pressure column is collected after heat recovery, 35
 a medium pressure nitrogen gas collecting path in which a medium pressure nitrogen gas obtained in the medium pressure column is collected after heat recovery,
 a low pressure nitrogen gas collecting path in which a low pressure nitrogen gas obtained in the low pressure column is collected after heat recovery, 40
 a high pressure liquid nitrogen collecting path in which the high pressure liquid nitrogen condensed in the main condenser reboiler is collected, 45
 a medium pressure liquid nitrogen collecting path in which the medium pressure liquid nitrogen condensed in the medium pressure column condenser is collected, and
 a low pressure liquid oxygen collecting path in which the low pressure liquid oxygen obtained in the low pressure column is collected. 50

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FIG.1

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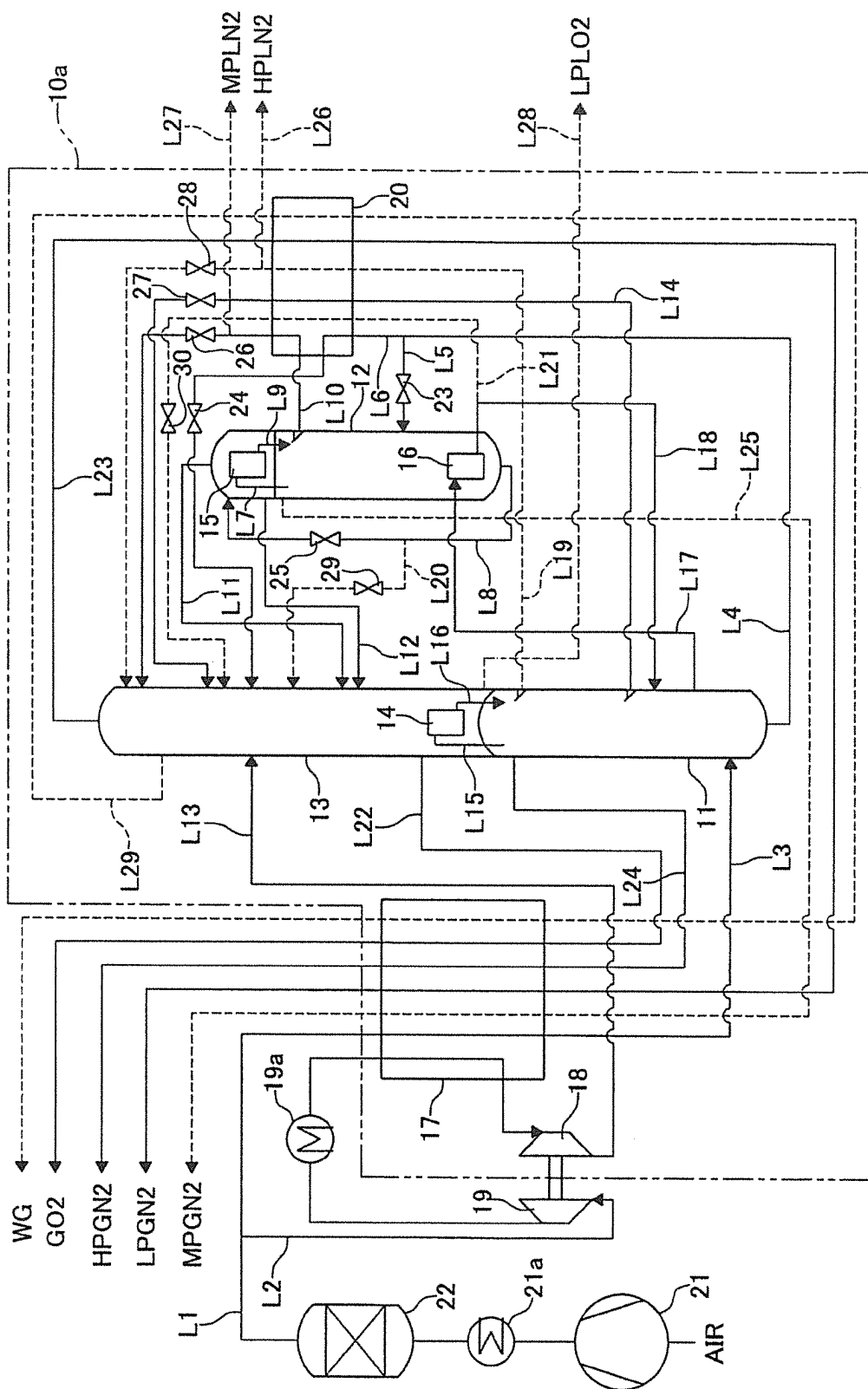


FIG.2

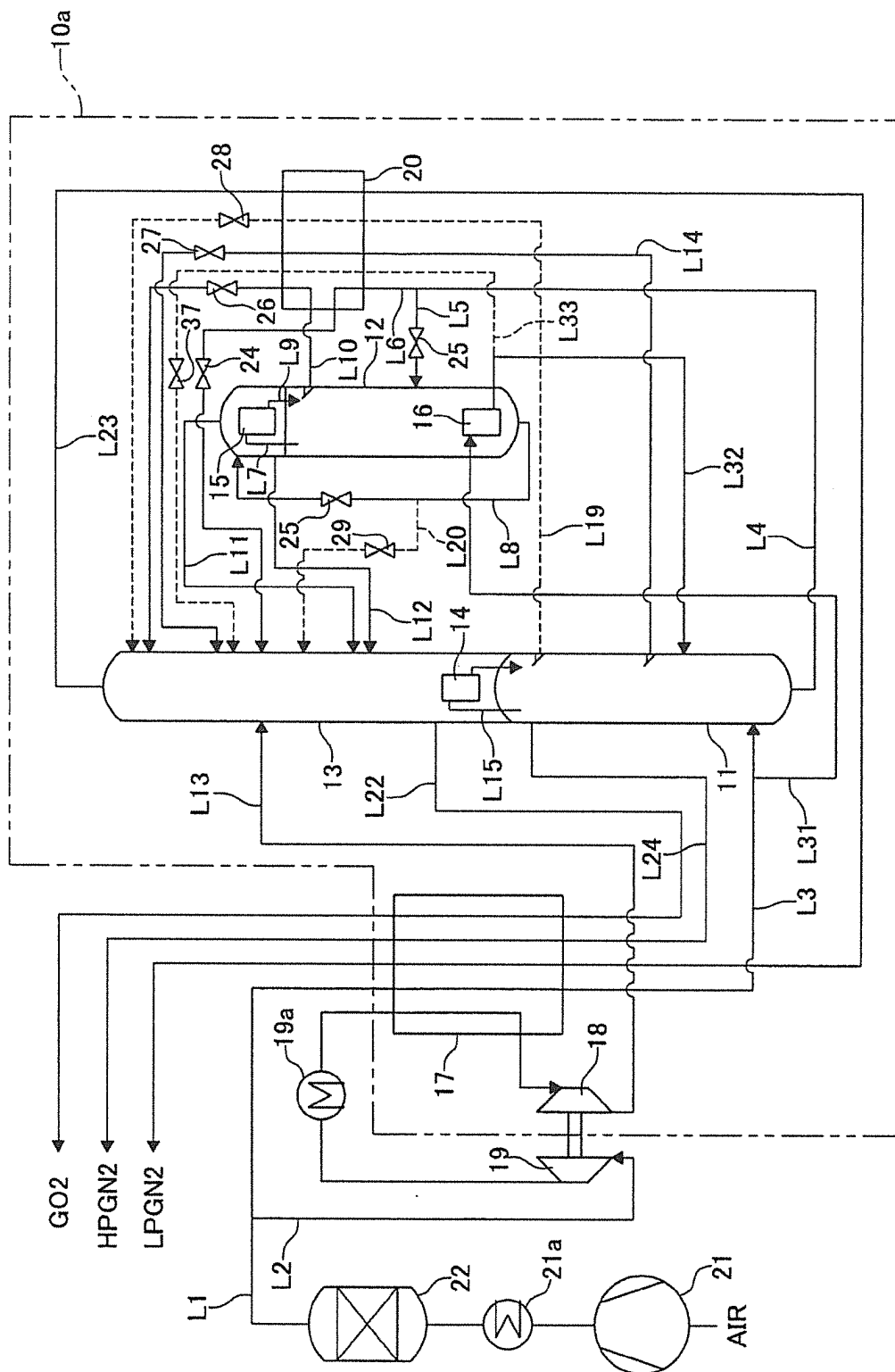


FIG. 3

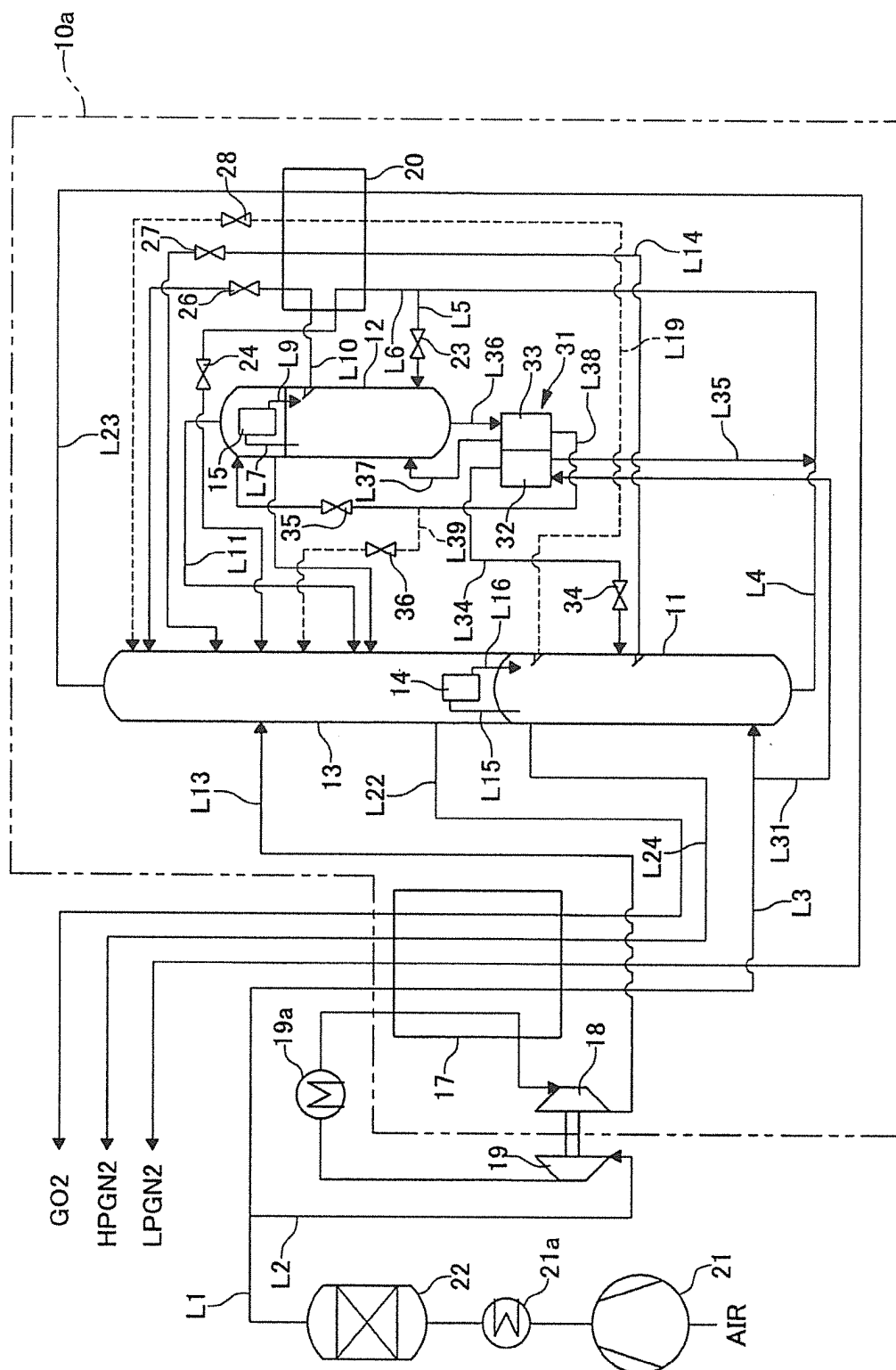
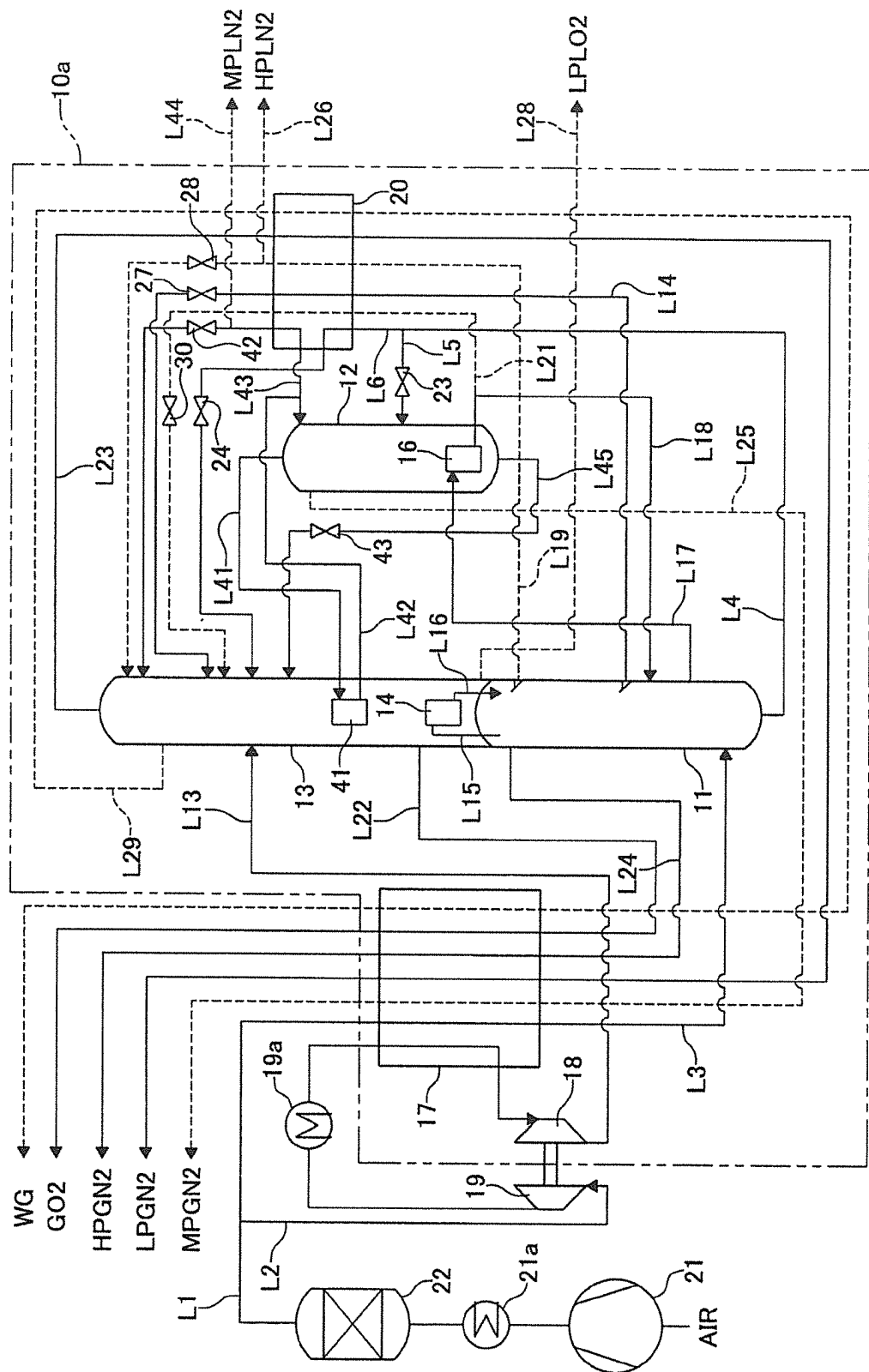


FIG. 4



INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP2012/065071

A. CLASSIFICATION OF SUBJECT MATTER

F25J3/04 (2006.01) i

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

F25J3/04

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Jitsuyo Shinan Koho	1922-1996	Jitsuyo Shinan Toroku Koho	1996-2012
Kokai Jitsuyo Shinan Koho	1971-2012	Toroku Jitsuyo Shinan Koho	1994-2012

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

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Y A	JP 8-233457 A (Air Products and Chemicals Inc.), 13 September 1996 (13.09.1996), paragraph [0028]; fig. 4, 5 & JP 10-47852 A & US 5513497 A & US 5678426 A & US 5692395 A & EP 722758 A2 & FI 960266 A & AU 675194 B & CA 2167265 A	1, 6 3, 8



Further documents are listed in the continuation of Box C.



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Date of the actual completion of the international search

20 August, 2012 (20.08.12)

Date of mailing of the international search report

28 August, 2012 (28.08.12)

Name and mailing address of the ISA/
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INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP2012/065071

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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