US 20090107177A1

(19) **United States**

(12) **Patent Application Publication**

(10) **Pub. No.: US 2009/0107177 A1**

(43) **Pub. Date: Apr. 30, 2009**

Lochner

(54) **PROCESS AND DEVICE FOR LOW TEMPERATURE AIR FRACTIONATION**

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(21) Appl. No.: **12/256,660**

(22) Filed: **Oct. 23, 2008**

(30) **Foreign Application Priority Data**

Oct. 25, 2007 (DE) 10 2007 0511 84.3
Oct. 25, 2007 (DE) 102007051183.5

Publication Classification

(51) **Int. Cl.**
F25J 3/00 (2006.01)

(52) **U.S. Cl.** **62/648**

(57) **ABSTRACT**

The process and the device serve for low temperature air fractionation in a distillation column system for obtaining nitrogen which has a single column (**12**). Feed air (**8**) is cooled in a main heat exchanger (**9**) and introduced (**11**, **43**) into the single column (**12**). The single column (**12**) has a top condenser (**13**) in which vapour from the upper region of the single column is at least in part condensed. A nitrogen product stream (**15**, **16**, **17**) is withdrawn from the upper region of the single column (**12**). A first residual fraction (**14**, **19**) is withdrawn in the liquid state from the single column (**12**), at least in part vaporized in the top condenser (**13**) and subsequently taken off from the top condenser as vaporized first residual fraction (**19**). A first part (**20**) of the vaporized first residual fraction (**19**) is expanded in a work-producing manner in an expansion machine (**21**). A second residual fraction (**18**, **29**) is withdrawn from the lower or intermediate region of the single column (**12**), recompressed (**30**) and subsequently passed (**32**) back at least to a first part of the single column (**12**). A second part of the vaporized first residual fraction (**19**) is not passed into the expansion machine (**21**) but is taken off as gaseous impure oxygen product (**60**) at about the inlet pressure of the expansion machine (**21**).

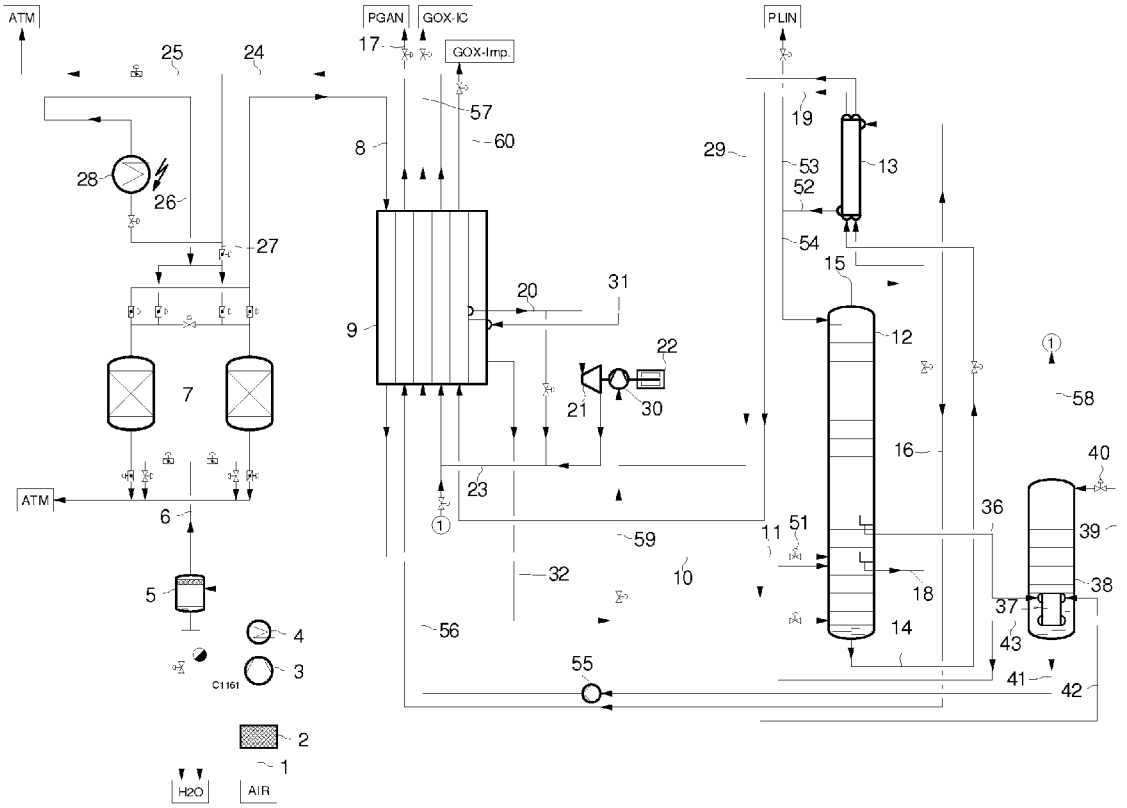


Fig. 1

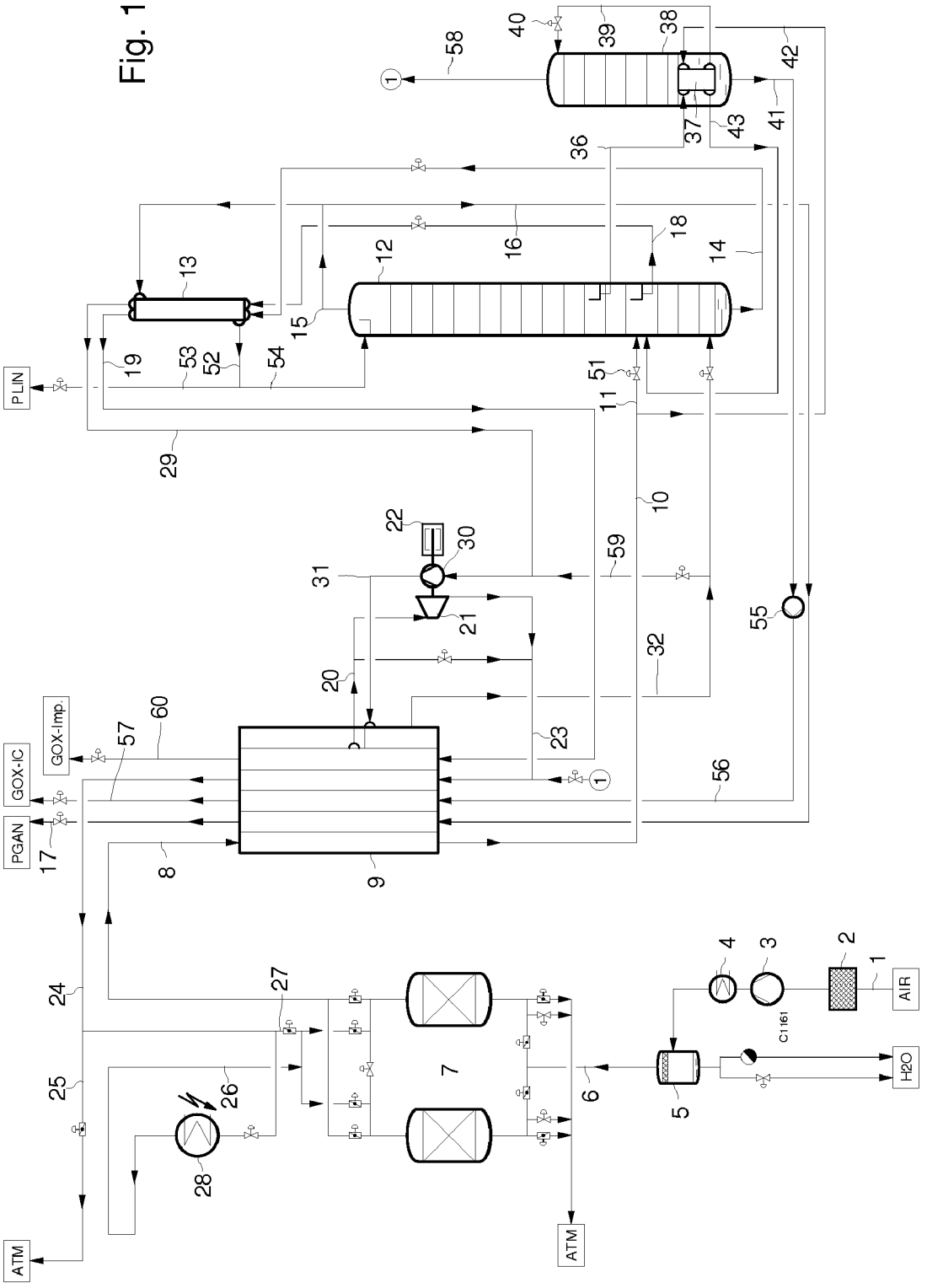
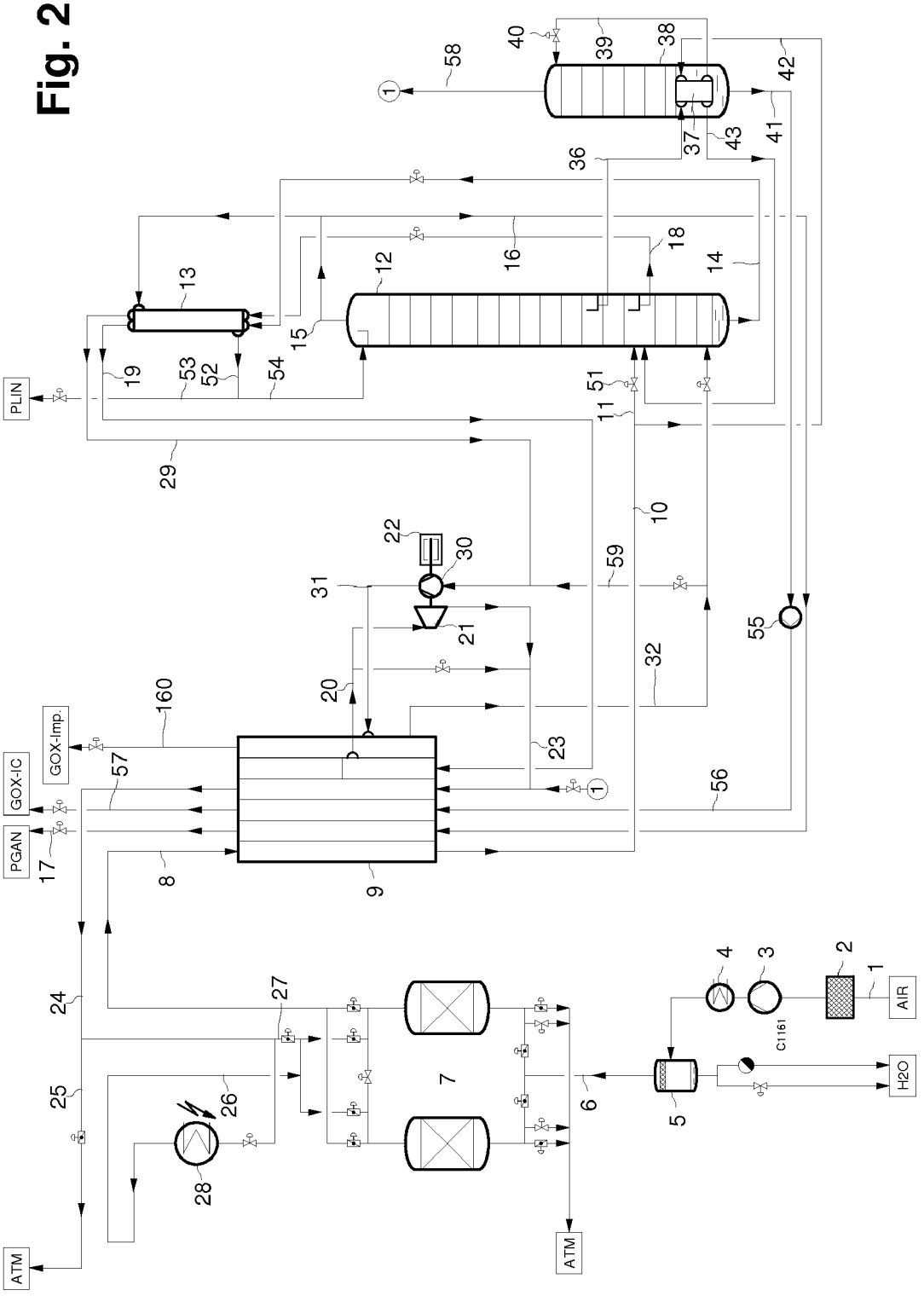


Fig. 2



PROCESS AND DEVICE FOR LOW TEMPERATURE AIR FRACTIONATION

CROSS-REFERENCE TO RELATED APPLICATION

[0001] This application is related to concurrently filed application "Low Temperature Air Fractionation With External Fluid" by Stefan Lochner, Attorney Docket No. LINDE-0678, claiming priority of DE 102007051183.5 filed Oct. 25, 2007, incorporated by reference herein.

[0002] The present invention relates to a process for low temperature air fractionation having a distillation column system for obtaining nitrogen.

[0003] Processes and devices for low temperature fractionation of air are known, for example, from Hausen/Linde, Tieftemperaturtechnik [Cryogenic engineering], 2nd edition 1985, chapter 4 (pages 281 to 337).

[0004] Examples of the type of air fractionation processes are described in EP 412793 B2, EP 773417 B1, EP 780648 B1, EP 807792 B1, EP 932004 A2 and US 2007204652 A1. It is known in such processes to withdraw a first oxygen-enriched residual fraction from the single column, vaporize it and expand at least a part of the vaporized residual fraction in an expansion machine in a work-producing manner in order to generate the refrigeration required to make up the exchange losses and, if appropriate, for product liquefaction. The first residual fraction is given off at the exit pressure of the expansion machine, that is customarily at approximately atmospheric pressure, and can then generally be used as only regeneration gas for an adsorption system for purifying the feed air. In SPECTRA processes, a second residual fraction can in this case be taken off from the single column together with the first residual fraction and at least in part vaporized; alternatively, the two residual fractions can be withdrawn at different points of the single column and vaporized separately from one another, for example in different passages of the top condenser of the single column.

[0005] Objects of this invention are to provide a process of the type mentioned above and a corresponding apparatus which may be operated with particularly favorable economics.

[0006] Upon further study of the specification to appended claims, other objects and advantages will become apparent. The figures in parentheses refer to the drawings to facilitate an understanding of the invention.

[0007] To achieve the process object there is provided a process for the low temperature air fractionation in a distillation column system for obtaining nitrogen which has a single column (**12**) in which

[0008] feed air (**8**) is cooled in a main heat exchanger (**9**) and introduced (**11**, **43**) into the single column (**12**),

[0009] the single column (**12**) has a top condenser (**13**) in which vapour from the upper region of the single column is at least in part condensed,

[0010] a nitrogen product stream (**15**, **16**, **17**) is withdrawn from the upper region of the single column (**12**),

[0011] a first residual fraction (**14**, **19**) is withdrawn in the liquid state from the single column (**12**), at least in part vaporized in the top condenser (**13**) and subsequently taken off from the top condenser as vaporized first residual fraction (**19**),

[0012] at least a first part (**20**) of the vaporized first residual fraction (**19**) is expanded in a work-producing manner in an expansion machine (**21**) and

[0013] a second residual fraction (**18**, **29**) is withdrawn from the lower or intermediate region of the single column (**12**), recompressed (**30**) and subsequently passed (**32**) back at least to a first part of the single column (**12**), and

[0014] an improvement wherein

[0015] a second part of the vaporized first residual fraction (**19**) is not passed into the expansion machine (**21**) but is taken off as gaseous impure oxygen product (**60**) at about the inlet pressure of the expansion machine (**21**) and/or

[0016] a second part of the second residual fraction is taken off downstream of the recompression (**30**) as gaseous impure oxygen product (**160**).

[0017] Generally, in the prior unimproved processes, all of the oxygen-enriched residual gas is used, either for generation of refrigeration by work-producing expansion or for recycling into the column. If, in addition, an oxygen product is required, hitherto either more complex processes having a double column have been used, or the required oxygen is delivered in the liquid state and vaporized externally. In the context of the invention, it has been found that it is nevertheless expedient, according to the first variant of the process according to the invention, to use only a part of the first residual fraction for production of refrigeration and to take off the rest, or a part of the rest, of the first fraction directly as gaseous impure oxygen product, more precisely at about the inlet pressure of the expansion machine, that is to say at a significantly superatmospheric pressure of 3 to 6 bar, preferably 3.5 to 5.5 bar. Although the required amount of feed air is somewhat increased thereby, despite the corresponding increased requirement of compression energy, the process according to the invention is more expedient than the known methods for additionally delivering impure oxygen as product. "At about the inlet pressure of the expansion machine" here means that the pressure of the gaseous impure oxygen product on being taken off from the process, because of the corresponding transmission losses, need not be exactly the same as the inlet pressure of the expansion machine; however, positive measures for changing pressure by more than 0.5 bar, for example expansion in a further expansion machine are excluded.

[0018] In a second variant of the process according to the invention, the recompressor for the second residual fraction is simultaneously used as product compressor for the gaseous impure oxygen product. This impure oxygen product can thereby be delivered at a comparatively high pressure. "Take off as gaseous impure oxygen product" here means that the second part of the second residual fraction is taken off from the process as end product in the same composition as prevails downstream of the compression without it being subjected to a further separation step.

[0019] The gaseous impure oxygen product can be used for any application which requires a corresponding pressure without a compressor being necessary, for example as oxidizing agent in a chemical reaction, for instance a combustion. Typically, there, a pressure of 1.8 bar and above is required. In the case of use at a still higher pressure, in the case of the invention, savings are made of a part of the expenditure on compression, compared with the unpressurized residual gas downstream of the expansion machine.

[0020] The impure oxygen product can be used in the invention for example for delivering O2-enriched combustion air. Compared with the use of air, this offers a number of

advantages. The increased oxygen content leads to an increase in the burner temperature. Since less nitrogen needs to be co-warmed and co-compressed, the energy expenditure is reduced. The burner performance is increased by more reactive mixture being able to flow through the burner.

[0021] Preferably, the first and second parts of the first residual fraction are introduced into the main heat exchanger, wherein the first part is withdrawn from the main heat exchanger at an intermediate temperature and passed to the expansion machine and the second part is warmed to about ambient temperature and is taken off as gaseous impure oxygen product.

[0022] The "main heat exchanger" is preferably formed by a single heat exchange block. In the case of relatively large plants, it can be expedient to implement the main heat exchanger by a plurality of trains which are connected in parallel with respect to the temperature course, which trains are formed by components which are separate from one another. In principle, it is possible that the main heat exchanger, or each of these trains, is formed by two or more blocks which are connected in series.

[0023] The first residual fraction can be branched into its two parts immediately downstream of the top condenser, which two parts are then introduced separately from one another into the main heat exchanger. However, preferably, both parts are warmed at least in part in the same passage of the main heat exchanger by the first and second parts of the first residual fraction being introduced together into the main heat exchanger.

[0024] Upstream of the recompression, the second residual fraction can be warmed to about ambient temperature. The recompression is performed in this case in the warming. The recompressed second residual fraction is then again cooled to about the operating temperature of the column before it is introduced into the single column. The warming and recooling of the second residual fraction can be performed in the main heat exchanger or in one or more other heat exchangers.

[0025] In many cases, however, it is more expedient if the second residual fraction is recompressed by means of a cold compressor. Then, the second residual fraction need not be warmed, or need be warmed only relatively slightly, upstream of the compression. "Cold compressor" here is taken to mean a compressor which is operated at an inlet pressure of below 200 K, preferably below 150 K, in particular between 90 and 120 K.

[0026] Both in cold and in warm compression, the mechanical energy generated in the work-producing expansion can be used at least in part for recompressing the second residual fraction. The mechanical energy is transmitted to the recompressor directly mechanically, for example via a shared shaft of expansion machine and recompressor. In particular when the recompressor is constructed as a cold compressor, preferably only a part of the mechanical energy generated by the expansion machine is transmitted to the recompressor; the remainder passes to a warm braking appliance, e.g. a braking fan, a generator or a dissipating brake.

[0027] In the context of the invention, in addition, pure oxygen can be obtained by withdrawing an oxygen-containing stream of the single column at an intermediate point and passing it to a pure oxygen column and withdrawing a pure oxygen product stream in the liquid state from the lower region of the pure oxygen column, vaporizing and warming the pure oxygen product stream—if appropriate after pressure elevation in the liquid state—in the main heat exchanger against feed air and finally obtaining it as gaseous product.

[0028] This procedure is also termed "internal compression" and is employed as an alternative to the gaseous product compression (external compression) when the gaseous product is obtained under pressure. A corresponding process—without obtaining a gaseous impure oxygen product according to the invention—is described in detail in earlier German patent application 102007024168 incorporated by reference herein, and the applications corresponding thereto. Alternatively, or in addition, the pure oxygen can also be given off as liquid product.

[0029] The expression "vaporization" here includes pseudo vaporization at supercritical pressure. The pressure at which the pure oxygen product stream is introduced into the main heat exchanger can therefore also be above the critical pressure, just as can the pressure of the heat carrier which is (pseudo) condensed against the pure oxygen product stream.

[0030] If the oxygen is required on site at an elevated pressure which is above the operating pressure of the pure oxygen column, it is expedient if the pure oxygen product stream is brought to an elevated pressure in the liquid state. By this means, in the context of the invention, a warm oxygen compressor can be omitted, or at least can be constructed to be relatively small. If withdrawal with highly varying rates is required, the oxygen can be transported at a pressure higher than release pressure into a gas pressure vessel which acts as a buffer.

[0031] It is expedient if the first residual fraction is taken off at the bottom of the single column.

[0032] The first and second residual fractions can in principle be taken off together with the second from the single column, for example at the bottom (see EP 412793 B2 incorporated by reference herein). However, in many cases it is more expedient if the second residual fraction has a higher nitrogen content than the first residual fraction. Then, the second residual fraction is taken off from an intermediate point of the single column which is arranged above the bottom, in particular above the point at which the first residual fraction is withdrawn. The two residual fractions are then vaporized separately in the top condenser and fed to the recompression and/or the work-producing expansion and the product withdrawal.

[0033] In addition, the invention relates to apparatus for low temperature air fractionation:

[0034] having a distillation column system (**12**) for obtaining nitrogen which has a single column (**12**),

[0035] having a main heat exchanger (**9**) for cooling feed air (**8**)

[0036] having means for introducing cooled feed air into the single column (**12**),

[0037] having a nitrogen product line (**15**, **16**, **17**) which is connected to the single column (**12**),

[0038] having a top condenser (**13**) for condensing vapour from the upper region of the single column,

[0039] having means for withdrawing a first residual fraction (**14**, **19**) in the liquid state from the single column (**12**) and for introducing the first residual fraction into the top condenser,

[0040] having means for taking off the at least in part vaporized first residual fraction (**19**) from the top condenser (**13**),

[0041] having a turbine line (**20**) for introducing at least a first part of the vaporized first residual fraction (**19**) into an expansion machine (**21**),

[0042] having means for withdrawing a second residual fraction (**18**, **29**) from the lower or intermediate region of the single column (**12**),

[0043] having means for recompressing (**30**) the second residual fraction and

[0044] having means for recycling at least a first part of the recompressed second residual fraction to the single column (**12**), and

[0045] an improvement comprising an impure oxygen product line (**60**, **160**)

[0046] for taking off a second part of the vaporized first residual fraction (**19**) as gaseous impure oxygen product at about the inlet pressure of the expansion machine (**21**) and/or

[0047] for taking off a second part of the second residual fraction as gaseous impure oxygen product downstream of the means for recompression (**30**).

BRIEF DESCRIPTION OF DRAWINGS

[0048] The invention and also further details of the invention will be described in more detail hereinafter with reference to two exemplary embodiments shown diagrammatically. In the drawings wherein:

[0049] FIG. **1** shows an exemplary embodiment of a first variant of the invention having takeoff of the gaseous impure oxygen product upstream of the expansion machine and

[0050] FIG. **2** shows an exemplary embodiment of a second variant of the invention with takeoff of gaseous impure oxygen products downstream of the recompressor.

DETAILED DESCRIPTION OF DRAWINGS

[0051] With reference to FIG. **1**, atmospheric air **1**, via a filter **2**, is taken in by an air compressor and there compressed to an absolute pressure of 6 to 20 bar, preferably about 9 bar. After flowing through an aftercooler **4** and a water separator **5**, the compressed air **6** is purified in a purification device **7** which has a pair of containers filled with adsorption material, preferably molecular sieve. The purified air **8** is cooled to about dew point in a main heat exchanger **9** and in part liquefied. A first part **11** of the cooled air **10** is introduced into a single column **12** via a throttle valve **51**. The infeed proceeds preferably some practical or theoretical plates above the bottom.

[0052] The operating pressure of the single column **12** (at the top) is 6 to 20 bar, preferably about 9 bar. Its top condenser is cooled by a second residual fraction **18** and a first residual fraction **14**. The first residual fraction **14** is taken off from the bottom of the single column **12**, the second residual fraction **18** from an intermediate point some practical or theoretical plates above the air infeed or at the same height as this. As the main product of single column **12**, gaseous nitrogen **15**, **16** is taken off at the top, warmed in the main heat exchanger **9** to approximately ambient temperature and finally taken off via line **17** as pressurized gaseous product (PGAN). A part **53** of the condensate **52** from the top condenser **13** can be obtained as product liquid nitrogen (PLIN); the remainder **54** is applied as reflux to the top of the single column.

[0053] The second residual fraction **18** is vaporized in the top condenser **13** at a pressure of 2 to 9 bar, preferably about 4 bar, and flows in the gaseous state via line **29** to a cold compressor **30** in which it is recompressed to approximately the operating pressure of the single column. The recompressed residual fraction **31** is cooled back to column temperature in the main heat exchanger **9** and finally fed back via line **32** to the single column **12** at the bottom.

[0054] The first residual fraction **14** is vaporized in the top condenser **13** at a pressure of 2 to 9 bar, preferably about 4 bar, and flows in the gaseous state via line **19** to the cold end of the main heat exchanger **9**. A first part **20** of the first residual fraction is withdrawn again (line **20**) at an intermediate temperature. A second part remains in the main heat exchanger **9**, is warmed there again to approximately ambient temperature and leaves the installation via line **60** as gaseous impure oxygen product (GOX-Imp.). The first part **20** of the first residual fraction is expanded to about 300 mbar over atmospheric pressure so as to produce work in an expansion machine **21** which is constructed in the example as a turbo expander. The expansion machine is mechanically coupled to the cold compressor **30** and a braking appliance **22** which, in the exemplary embodiment, is formed by an oil brake. The expanded first residual fraction **23** is warmed in the main heat exchanger **9** to approximately ambient temperature. The warm first residual fraction **24** is blown off into atmosphere (line **25**) and/or used as regeneration gas **26**, **27** in the purification device **7**, if appropriate after heating in the heating appliance **28**.

[0055] An oxygen-containing stream **36** which is essentially free of low volatility impurities is taken off from an intermediate point of the single column **12** in the liquid state, which intermediate point is arranged 5 to 25 theoretical or practical plates above the air infeed. The oxygen-containing stream **36** is, if appropriate, subcooled in a bottoms evaporator **37** of a pure oxygen column **38** and applied to the top of the pure oxygen column **38** via line **39** and throttle valve **40**. The operating pressure of the pure oxygen column **38** (at the top) is 1.3 to 4 bar, preferably about 2.5 bar.

[0056] The bottoms evaporator **37** of the pure oxygen column **38** is in addition cooled by means of a second part **42** of the cooled feed air **10**. The feed air stream **42** is in this case at least in part, for example completely, condensed and flows via line **43** to the single column **12** where it is introduced approximately at the height of the infeed of the remaining feed air **11**.

[0057] From the bottom of the pure oxygen column **38**, a pure oxygen product stream **41** is withdrawn in the liquid state, brought by means of a pump **55** to an elevated pressure of 2 to 100 bar, preferably about 12 bar, passed via line **56** to the cold end of the main heat exchanger **9**, vaporized there at the elevated pressure and warmed to about ambient temperature and finally obtained via line **57** as gaseous product (GOX-IC).

[0058] The overhead gas **58** of the pure oxygen column **38** is admixed to the expanded first residual fraction **23**. Via a bypass line **59**, if appropriate a part of the feed air is passed for pump protection of the cold compressor **30** to the inlet thereof (anti-surge control).

[0059] If required, a liquid oxygen can be withdrawn as liquid product from the installation upstream and/or downstream of the pump **55** (not shown in the drawing). In addition, an external liquid, for example liquid argon, liquid nitrogen or liquid oxygen from a liquid tank can be vaporized in the main heat exchanger **9** in indirect heat exchange with the feed air (not shown in the drawing).

[0060] The process according to the invention and the corresponding device can be used particularly expediently in the semiconductor industry or in pyrogenic silicic acid production which require not only nitrogen but also impure oxygen and if appropriate pure oxygen as products.

[0061] FIG. **2** is differentiated from FIG. **1** only in that here the gaseous impure oxygen product of FIG. **1** is not branched off from the first residual fraction upstream of the expansion machine, but is branched off from the second residual fraction **31** downstream of the recompressor **30**. Only a first part of the recompressed second residual gas fraction **31** is cooled in the main heat exchanger and conducted via line **32** back into the column; the remainder flows in the main heat exchanger to the

warm end and is there taken off as gaseous impure oxygen product **160**. The first residual fraction **19**/**20** is entirely fed to the expansion machine **21**.

[0062] Also a combination of the two variants of FIGS. **1** and **2** is possible in principle, that is to say taking off two gaseous impure oxygen products at different pressures.

[0063] Without further elaboration, it is believed that one skilled in the art can, using the preceding description, utilize the present invention to its fullest extent. The preceding preferred specific embodiments are, therefore, to be construed as merely illustrative, and not limitative of the remainder of the disclosure in any way whatsoever.

[0064] The entire disclosures of all applications, patents and publications, cited herein and of corresponding German application No. 102007051184.3, filed Oct. 25, 2007 are incorporated by reference herein.

[0065] 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.

1. A process for the low temperature air fractionation in a distillation column system for obtaining nitrogen which has a single column (**12**) in which

- feed air (**8**) is cooled in a main heat exchanger (**9**) and introduced (**11**, **43**) into the single column (**12**),
- the single column (**12**) has a top condenser (**13**) in which vapor from the upper region of the single column is at least in part condensed,
- a nitrogen product stream (**15**, **16**, **17**) is withdrawn from the upper region of the single column (**12**),
- a first residual fraction (**14**, **19**) is withdrawn in the liquid state from the single column (**12**), at least in part vaporized in the top condenser (**13**) and subsequently taken off from the top condenser as vaporized first residual fraction (**19**),
- at least a first part (**20**) of the vaporized first residual fraction (**19**) is expanded in a work-producing manner in an expansion machine (**21**) and
- a second residual fraction (**18**, **29**) is withdrawn from the lower or intermediate region of the single column (**12**), recompressed (**30**) and subsequently passed (**32**) back at least to a first part of the single column (**12**),

characterized in that

- a second part of the vaporized first residual fraction (**19**) is not passed into the expansion machine (**21**) but is taken off as gaseous impure oxygen product (**60**) at about the inlet pressure of the expansion machine (**21**) and/or
- a second part of the second residual fraction is taken off downstream of the recompression (**30**) as gaseous impure oxygen product (**160**).

2. A process according to claim **1**, characterized in that the first and second parts of the first residual fraction are introduced into the main heat exchanger (**9**), wherein the first part (**20**) is withdrawn from the main heat exchanger (**9**) at an intermediate temperature and passed to the expansion machine (**21**) and the second part is warmed to about ambient temperature and is taken off as gaseous impure oxygen product (**60**).

3. A process according to claim **2**, characterized in that the first and second parts of the first residual fraction are introduced (**19**) together into the main heat exchanger (**9**).

4. A process according to claim **1**, characterized in that the second residual fraction (**18**, **29**) is recompressed by means of a cold compressor (**30**).

5. A process according to claim **1**, characterized in that the mechanical energy generated in the work-producing expansion (**21**) is used at least in part for recompressing (**30**) the second residual fraction.

6. A process according to claim **1**, characterized in that

- an oxygen-containing stream (**36**) is withdrawn from the single column (**12**) at an intermediate point and passed (**39**) to a pure oxygen column (**38**) and
- a pure oxygen product stream (**41**) is withdrawn in the liquid state from the lower region of the pure oxygen column (**38**),
- the pure oxygen product stream (**41**, **56**), optionally after pressure elevation (**55**) in the liquid state, is vaporized and warmed against feed air (**8**) in the main heat exchanger (**9**)
- and finally is obtained as gaseous product (**57**).

7. A process according to claim **1**, characterized in that the first residual fraction (**14**) is taken off the bottom of the single column (**12**).

8. A process according to claim **1**, characterized in that the second residual fraction (**18**) is taken off from an intermediate point of the single column (**12**), which intermediate point is arranged above the bottom, in particular above the point at which the first residual fraction (**14**) is withdrawn.

9. A process according to claim **1**, characterized in that the exit pressure of the expansion machine (**21**) is less than 0.5 bar above atmospheric pressure.

10. A process according to claim **1**, characterized in that the second residual fraction (**18**, **29**) is recompressed (**30**) to a pressure which is less than 0.5 bar above the operating pressure of the single column (**12**).

11. Apparatus for low temperature air fractionation:

- having a distillation column system (**12**) for obtaining nitrogen which has a single column (**12**),
- having a main heat exchanger (**9**) for cooling feed air (**8**)
- having means for introducing cooled feed air into the single column (**12**),
- having a nitrogen product line (**15**, **16**, **17**) which is connected to the single column (**12**),
- having a top condenser (**13**) for condensing vapor from the upper region of the single column,
- having means for withdrawing a first residual fraction (**14**, **19**) in the liquid state from the single column (**12**) and for introducing the first residual fraction into the top condenser,
- having means for taking off the at least in part vaporized first residual fraction (**19**) from the top condenser (**13**),
- having a turbine line (**20**) for introducing at least a first part of the vaporized first residual fraction (**19**) into an expansion machine (**21**),
- having means for withdrawing a second residual fraction (**18**, **29**) from the lower or intermediate region of the single column (**12**),
- having means for recompressing (**30**) the second residual fraction and
- having means for recycling at least a first part of the recompressed second residual fraction to the single column (**12**),

characterized by an impure oxygen product line (**60**, **160**)

- for taking off a second part of the vaporized first residual fraction (**19**) as gaseous impure oxygen product at about the inlet pressure of the expansion machine (**21**) and/or
- for taking off a second part of the second residual fraction as gaseous impure oxygen product downstream of the means for recompression (**30**).

* * * * *