



US 20100242537A1

(19) **United States**(12) **Patent Application Publication**
LOCHNER(10) **Pub. No.: US 2010/0242537 A1**(43) **Pub. Date: Sep. 30, 2010**(54) **PROCESS AND APPARATUS FOR**
CRYOGENIC AIR SEPARATION(52) **U.S. Cl. 62/644; 62/649**(57) **ABSTRACT**(75) **Inventor: Stefan LOCHNER, Grafing (DE)**

Correspondence Address:

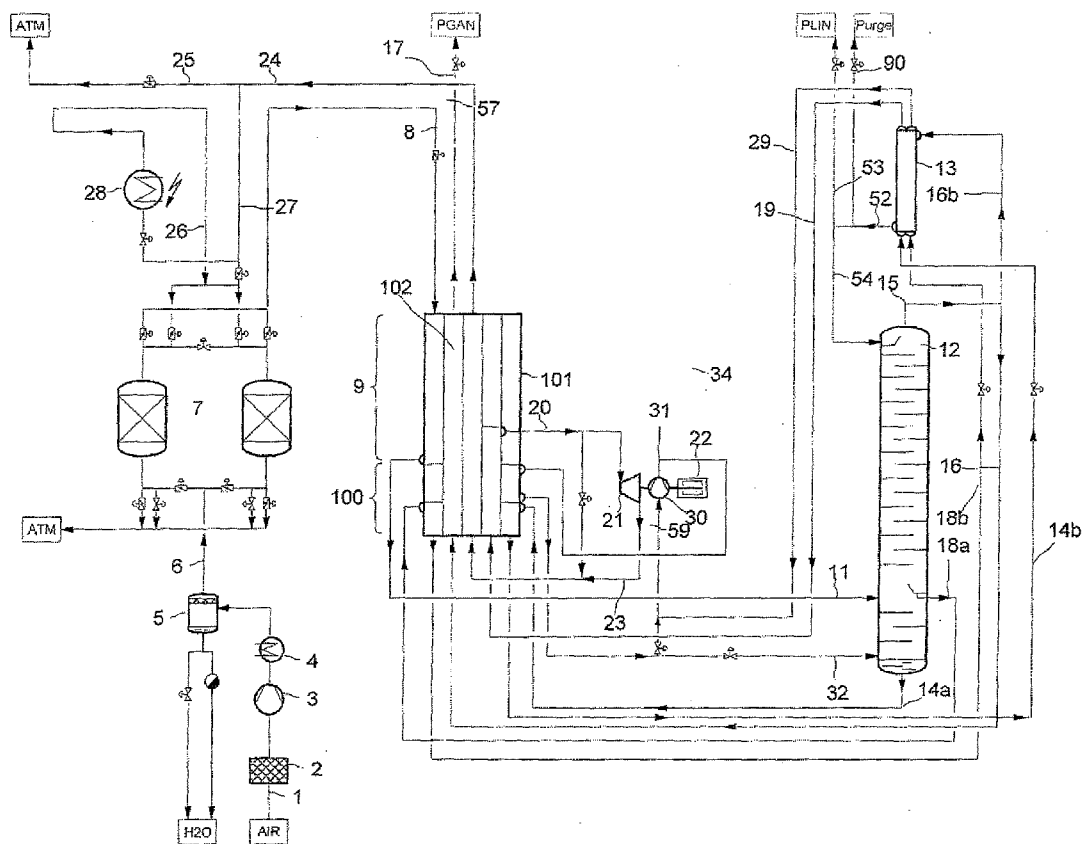
MILLEN, WHITE, ZELANO & BRANIGAN, P.C.
2200 CLARENDON BLVD., SUITE 1400
ARLINGTON, VA 22201 (US)(73) **Assignee: LINDE AG, Muenchen (DE)**(21) **Appl. No.: 12/730,429**(22) **Filed: Mar. 24, 2010**(30) **Foreign Application Priority Data**

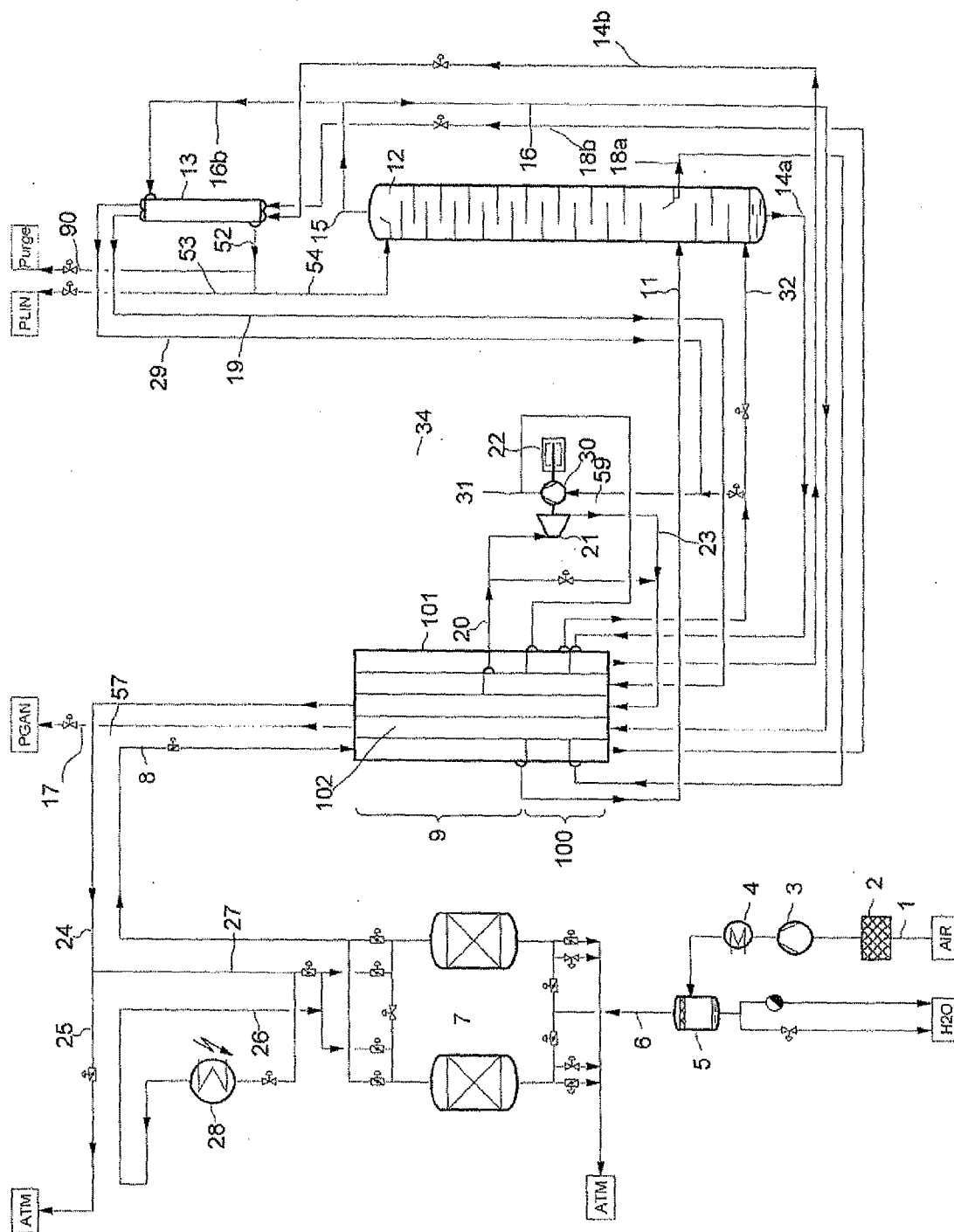
Mar. 24, 2009 (DE) 102009014557.5

Jun. 23, 2009 (EP) 09008224.9

Publication Classification(51) **Int. Cl.**
F25J 3/04 (2006.01)

The process and the apparatus in accordance with the invention relate to cryogenic separation of air in a distillation column system that has at least one single column (12). A compressed feed air stream (6, 8) is cooled in a main heat exchanger (9) in counter-current flow to a first return stream (16, 23) from the distillation column system. Cooled feed air stream (11) is fed into the distillation column system. A nitrogen-rich fraction (15) is produced in the upper region of the single column (12). At least part (16b) of the nitrogen-rich fraction (15) is condensed in a top condenser (13), which is constructed as a condenser-evaporator. At least part (54) of the liquid nitrogen-rich fraction (52) produced in the top condenser (13) is fed into the single column (12) as reflux. An oxygen-containing recycle fraction (18a) is drawn off from the single column (12) in liquid form. The liquid recycle fraction (18a) is cooled in a counter-current subcooler (100). The cooled recycle fraction (18b) is evaporated in the top condenser (13). The evaporated recycle fraction (29) is re-compressed in a re-compressor (30). The re-compressed recycle fraction (31, 32) is fed to the lower region of the single column (12). The main heat exchanger (9) and the counter-current subcooler (100) are formed as an integrated heat exchanger (102). The first return stream (16, 23) is fed into a group of passages (102) within the integrated heat exchanger which extend from the cold end thereof to the warm end thereof, and, in the process, the first return stream is brought into indirect heat exchange with both the liquid recycle fraction (18a) and the feed air stream (8).





PROCESS AND APPARATUS FOR CRYOGENIC AIR SEPARATION

SUMMARY OF THE INVENTION

[0001] The invention relates to a process for cryogenic air separation which has at least one single column, wherein:

[0002] a compressed feed air stream is cooled down in a main heat exchanger in counter-current flow to a first return stream (i.e., returned from the distillation column system to the main heat exchanger),

[0003] the cooled feed air stream is fed into the single column,

[0004] a nitrogen-rich fraction is produced in the upper region of the single column,

[0005] at least part of the nitrogen-rich fraction is condensed in a top condenser, which is constructed as a condenser-evaporator,

[0006] at least part of the liquid nitrogen-rich fraction produced in the top condenser is fed into the single column as reflux,

[0007] an oxygen-containing liquid recycle fraction is drawn off from the single column in liquid form,

[0008] the liquid recycle fraction is cooled down in a counter-current subcooler,

[0009] the cooled liquid recycle fraction is evaporated in the top condenser,

[0010] the evaporated recycle fraction is re-compressed in a re-compressor, and

[0011] the re-compressed recycle fraction is fed to the lower region of the single column.

[0012] Similar processes with residual gas recycling are known from DE 2261234, U.S. Pat. No. 4,966,002, U.S. Pat. No. 5,363,657, U.S. Pat. No. 5,528,906, U.S. Pat. No. 5,934,106, U.S. Pat. No. 5,611,218, U.S. Pat. No. 5,582,034, US 2004244417, DE 19909744 A1, DE 19919933 A1, DE 19954593 A1, US 2007204652 A1, DE 102006027650 A1 and EP 1995537 A2. In this case, and also in U.S. Pat. No. 4,966,002 and U.S. Pat. No. 5,582,034, a counter-current subcooler is used, in which the liquid oxygen-containing recycle fraction is "subcooled", i.e. cooled below its boiling point.

[0013] Here, "single column" is understood to mean a distillation column which is operated in a uniform pressure range—which means here that the pressure difference between top and bottom of the column is based exclusively on the pressure loss of the vapour rising in the column—and in which both the feed air is fed in as main feed fraction and also the nitrogen product is produced in the form of part of the nitrogen-rich fraction accumulating in the upper region of the column. Double-column or triple-column processes for nitrogen/oxygen separation are therefore not covered. However, a pure oxygen column, which is connected to the single column and is operated as a pure stripping column, is not ruled out.

[0014] Processes and apparatuses for the cryogenic separation of air are described in general terms in Hausen/Linde, Tieftemperaturtechnik, 2nd Edition 1985, Chapter 4 (pages 281 to 337).

[0015] An aspect of the invention is to provide a process of the type mentioned above, as well as a corresponding apparatus, which are economically particularly beneficial.

[0016] Thus, in accordance with the invention, there is provided a process of the type mentioned above wherein (references numerals refer to those of the FIGURE):

[0017] the main heat exchanger (9) and the counter-current subcooler (100) are formed as an integrated heat exchanger (101),

[0018] the integrated heat exchanger (101) having a first group of passages for the first return stream (16, 23), which extends from the cold end of the integrated heat exchanger (101) to the warm end of the integrated heat exchanger (101),

[0019] the first return stream (16, 23) is introduced into this group of passages (102) at the cold end and flowing through the integrated heat exchanger (101) as far as the warm end thereof,

[0020] the first return stream (16, 23) being brought into indirect heat exchange with both the liquid recycle fraction (18a) and with the feed air stream (8) in the integrated heat exchanger (101), and

[0021] the cooled feed air stream (11) being withdrawn from the integrated heat exchanger (101) in completely gaseous form and being fed into the single column (12) in completely gaseous form.

[0022] Surprisingly, the use of an integrated heat exchanger, which combines the functions of a main heat exchanger and a counter-current subcooler, permits any preliquefaction of the air to be avoided. As a result, all of the air fed to the column is able to rise and participate in the rectification. Thus, the separation effect becomes higher and, overall, the process according to the invention is therefore particularly beneficial. The precise layout of the integrated heat exchanger depends on the boundary conditions of the individual case and must be defined for each plant by using the usual calculation tools of the process engineer.

[0023] Besides this, the integration according to the invention simplifies the design considerably with respect to the pipework. Since the counter-current subcooler is given a substantially larger cross section as a result of the integration in the main heat exchanger, the liquid streams that flow in counter-current relation to the gas streams are offered an optimum heating surface area. It is merely necessary for a heat exchanger to be supported and piped in the coldbox. The absolute number of headers of the two heat exchangers decreases. The gas streams (residual gas to the turbine, product nitrogen, residual gas from the turbine) from the top of the coldbox do not have to be led via two fixed points (counter-current subcooler and main heat exchanger). Expansion loops can be dispensed with; the integrated solution permits a pipe run with minimized pipe stresses.

[0024] The integration of main heat exchanger and counter-current subcooler is certainly known from air separation processes having two or more columns for nitrogen/oxygen separation. However, this measure has not previously been applied to processes of the type mentioned above, since the manufacturing outlay for a particularly long integrated heat exchanger did not appear to be justified in single column processes. The surprising effect of the avoidance of pre-liquefaction of the air was previously unknown.

[0025] In principle, any heat exchanger type can be used as an integrated heat exchanger in the process according to the invention, for example a helically coiled heat exchanger or else a straight pipe exchanger. However, the use of a plate-type heat exchanger, in particular a brazed aluminium plate heat exchanger, is particularly beneficial. In this case, the integrated heat exchanger is formed by a single plate-type heat exchanger block.

[0026] It is particularly cost-effective if the single column constitutes the only distillation column of the distillation column system.

[0027] In order to generate refrigerating capacity, a further oxygen-containing fraction can be expanded, producing work. Thus, for example, the process can further comprise:

[0028] withdrawing a further oxygen-containing fraction (14a) in liquid form from the single column (12),

[0029] this further oxygen-containing liquid fraction (14a) is cooled down in the integrated heat exchanger (101),

[0030] the cooled further oxygen-containing fraction is evaporated in the top condenser (13),

[0031] the evaporated further oxygen-containing fraction (19) is warmed in the integrated heat exchanger (101) in counter-current flow to air, and

[0032] the warmed further oxygen-containing fraction is expanded in an expansion machine (21) to produce work, and

[0033] the temperature of the liquid further oxygen-containing fraction (14a), as it is introduced into the integrated heat exchanger (101), is higher than the temperature of the cooled feed air stream (11) as it is withdrawn from the integrated heat exchanger (101).

[0034] The integrated heat exchanger is also used for the subcooling of the further oxygen-containing fraction, in that the liquid further oxygen-containing fraction is cooled down in the counter-current subcooler before its evaporation. The integration according to the invention makes it possible to introduce the further oxygen-containing liquid fraction into the heat exchanger above the temperature of the air removal. The temperature difference is, for example, 0.2 to 5 K. This contributes to the avoidance of the pre-liquefaction.

[0035] In addition, before being subjected to work-producing expansion, the evaporated further oxygen-containing fraction is warmed up by counter-current heat exchange with air in the integrated heat exchanger.

[0036] The further oxygen-containing fraction can, for example, have the same composition as the recycle fraction. In this case, the two fractions can be led in common lines and passages until after the top condenser.

[0037] Alternatively, the oxygen-containing recycle fraction is removed from the single column at an intermediate point which is located at least one theoretical or practical plate above the point at which the further oxygen-containing fraction is removed. In this case, separate lines and separate passages must be provided for the two fractions in the top condenser and possibly in the counter-current subcooler.

[0038] Advantageously, the expansion machine is coupled mechanically to the re-compressor. As a result, the mechanical energy obtained during the work-producing expansion is used for re-compression. This is preferably the only energy source for the drive of the re-compressor.

[0039] It is beneficial if the re-compressor is constructed as a cold compressor. Here, a "cold compressor" is understood to mean an apparatus in which the gas to be compressed is fed in at a temperature which lies considerably below the ambient temperature, in general below 250 K, preferably below 200 K.

[0040] It is also beneficial if, in the process according to the invention, the re-compressed recycle fraction is cooled in the integrated heat exchanger before being introduced into the lower region of the single column, the re-compressed recycle fraction being drawn off from the integrated heat exchanger in completely gaseous form and led into the single column in

completely gaseous form. The recycle fraction is therefore also free of pre-liquefaction and participates in the rectification in the single column completely as rising vapor. Therefore, the pre-liquefaction is avoided completely in both feed streams to the single column, namely in the feed air and in the recycle fraction.

[0041] According to an apparatus aspect, the invention provides an apparatus for cryogenic air separation in a distillation column system, comprising:

[0042] at least one single column (12),

[0043] a main heat exchanger (9) for cooling a compressed feed air stream (6, 8) in counter-current flow to a first return stream (16, 23) from the distillation column system,

[0044] means (such as conduits or piping) for introducing the cooled feed air stream (11) into the single column (12),

[0045] means (such as conduits or piping) for removing a nitrogen-rich fraction (15) from the upper region of the single column (12),

[0046] a top condenser for condensing at least part of the nitrogen-rich fraction, the top condenser being constructed as a condenser-evaporator,

[0047] means (such as conduits or piping) for introducing the liquid nitrogen-rich fraction (52) produced in the top condenser (13) into the single column (12) as reflux,

[0048] means (such as conduits or piping) for withdrawing an oxygen-containing recycle fraction (18a) from the single column (12) in the liquid state,

[0049] a counter-current subcooler (100) for cooling down the liquid recycle fraction (18a),

[0050] means (such as conduits or piping) for introducing the cooled recycle fraction (18b) into the top condenser (13),

[0051] a re-compressor (30) for compressing the evaporated recycle fraction (29) from the top condenser (13), the re-compressor (30) being constructed, for example, as a cold compressor, and

[0052] means (such as conduits or piping) for introducing the re-compressed recycle fraction (31, 32) into the lower region of the single column (12),

wherein

[0053] the main heat exchanger (9) and the counter-current subcooler (100) are formed as an integrated heat exchanger (101),

[0054] the integrated heat exchanger (101) having a first group of passages (102) for the first return stream (16, 23), which extends from the cold end to the warm end of the integrated heat exchanger,

[0055] the cold end of the integrated heat exchanger (101) being connected to means (such as conduits or piping) for introducing the first return stream (16, 23) into the first group of passages,

[0056] the warm end of the integrated heat exchanger (101) being connected to means (such as conduits or piping) for withdrawing the first return stream (16, 23) from the first group of passages,

[0057] the integrated heat exchanger (101) being constructed in such a way that, during operation, the first return stream (16, 23) is brought into indirect heat exchange with both the liquid recycle fraction (18a) and the feed air stream (8), and

[0058] the passages in the integrated heat exchanger (101) are arranged in such a way that, during operation,

the cooled feed air stream (11) is withdrawn from the integrated heat exchanger (101) in completely gaseous form and is fed into the single column (12) in completely gaseous form.

BRIEF DESCRIPTION OF THE DRAWINGS

[0059] The invention and further details, such as features and attendant advantages, of the invention are explained in more detail below on the basis of an exemplary embodiment which is diagrammatically depicted in the drawing, and wherein:

[0060] FIG. 1 shows an embodiment of the device according to the invention.

DETAILED DESCRIPTION OF THE DRAWINGS

[0061] Atmospheric air 1 is taken in by an air compressor 3, via a filter 2, and compressed to an absolute pressure of 6 to 20 bar, preferably about 9 bar. After flowing through a re-cooler 4 and a water separator 5, the compressed air 6 is cleaned in a cleaning device 7. The cleaning device 7 has a pair of containers which are filled with adsorption material, preferably a molecular sieve. The cleaned air 8 is cooled down to somewhat above the dew point in a main heat exchanger 9 and finally led into a single column 12 as a completely gaseous feed air stream 11.

[0062] 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 with an oxygen-containing recycle fraction 18a, 18b and a further oxygen-containing fraction 14a, 14b. The further oxygen-containing fraction 14a is drawn off from the bottom of the single column 12, the recycle fraction 18a from an intermediate point some practical or theoretical plates further above the bottom of the single column 12. Before they are fed 14b, 18b into the top condenser 13, both fractions 14a, 18a are cooled down in a counter-current subcooler 100. The main heat exchanger 9 and counter-current subcooler 100, according to the invention, are formed by an integrated heat exchanger 101, which is implemented here as a single plate-type heat exchanger block. The height difference between the exit of the stream 14a from the single column 12 (more precisely the liquid level at the bottom of the column) and the entry into the integrated heat exchanger 101 should in principle be chosen such that the proportion of gas as a result of the expansion lies below 5% by volume. If, in a departure from this, the proportion of gas is higher than 5% by volume, a perforated plate is fitted in the header over the entire region above the point of entry of the two-phase mixture into the passages. The pressure loss across the perforated plate is chosen such that the gas bubbles are distributed over all the passages. The two-phase mixture is then fed into the integrated heat exchanger (101), first transversely with respect to the other streams (possibly with one or more deflections), in which the gas proportion is condensed completely, that is to say the adjacent passages are correspondingly colder in every operating case. After the initial transverse flow, the fluid streams flows counter-current to the other streams.

[0063] The main product from the single column 12, gaseous nitrogen 15, 16 is drawn off at the top and, as first return stream, is led through a group of passages 102 which extend from the cold end to the warm end of the integrated heat exchanger. In the process, the recycle stream (16) in the region of the counter-current subcooler 100 comes into indirect heat exchange with the two oxygen-containing fractions

14a, 18a and then, in the region of the main heat exchanger 9, into indirect heat exchange with the feed air stream 8. Via a line 17, it is finally drawn off at approximately ambient temperature as a gaseous pressurized product (PGAN).

[0064] The remainder 16b of the gaseous nitrogen 15 is condensed completely or substantially completely in the top condenser 13. Part 53 of the condensate 52 from the top condenser 13 can be removed as liquid nitrogen product (PLIN); the remainder 54 is introduced into the top of the single column as reflux. Non-condensed constituents can be drawn off via a purge line 90.

[0065] The recycle fraction 18b is evaporated in the top condenser 13 under a pressure of 2 to 9 bar, preferably about 4 bar, and flows in gaseous form via line 29 to a cold compressor 30, in which it is re-compressed approximately to a pressure which is sufficient to feed it back into the single column. The re-compressed recycle fraction 31 is cooled down to column temperature again in the counter-current subcooler 100 and fed to the single column 12 at or near the bottom in completely gaseous form via line 32.

[0066] The further oxygen-containing fraction 14b is evaporated in the top condenser 13 under a pressure of 2 to 9 bar, preferably about 4 bar, and flows in gaseous form via line 19 to the cold end of the integrated heat exchanger 101. There, in the region of the counter-current subcooler 100, it comes into indirect heat exchange with the two liquid oxygen-containing fractions 14a, 18a and then, in the region of the main heat exchanger 9, into indirect heat exchange with the feed air stream 8. It is removed from the main heat exchanger 9 again (line 20) at an intermediate temperature and is expanded to about 300 mbar above atmospheric pressure, producing work, in an expansion machine 21 which, in the example, is constructed as a turbo-expander. The expansion machine is coupled mechanically to the cold compressor 30 and a braking device 22 which, in the exemplary embodiment, is formed by an oil-filled brake. The expanded further fraction 23 is warmed up to about ambient temperature in the integrated heat exchanger 101. The warm further fraction 24 is blown off into the atmosphere (line 25) and/or used in the cleaning device 7 as regeneration gas 26, 27, possibly following heating in the heating device 28.

[0067] As mentioned above, the further oxygen-containing fraction (14a, 14b) can, for example, have the same composition as the oxygen recycle fraction (18a, 18b). For example, these two fractions can be removed from column 12 as a single stream, for example the stream (14a, 14b), and introduced as a single stream into top condenser 13. Thereafter, the streams are divided into two streams (19) and (29).

[0068] In the exemplary embodiment, the top condenser 13 is constructed as a forced-flow evaporator. Alternatively, a bath evaporator or falling film evaporator can be used.

[0069] The entire disclosure[s] of all applications, patents and publications, cited herein and of corresponding German Application No. 10 2009 9014557.5, filed Mar. 24, 2009 are incorporated by reference herein.

[0070] The preceding examples can be repeated with similar success by substituting the generically or specifically described reactants and/or operating conditions of this invention for those used in the preceding examples.

[0071] 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 cryogenic air separation in a distillation column system comprising at least one single column (12), said process comprising:

cooling a compressed feed air stream (6, 8) in a main heat exchanger (9) in counter-current to a first return stream (16, 23) from the distillation column system, introducing the cooled feed air stream (11) into said single column (12), removing a nitrogen-rich fraction (15) from the upper region of said single column (12), condensing at least part (16b) of the nitrogen-rich fraction (15) in a top condenser-evaporator (13), introducing at least part (54) of the condensed liquid nitrogen-rich fraction (52) from the top condenser-evaporator (13) into said single column (12) as reflux, withdrawing an oxygen-containing recycle fraction (18a) from said single column (12) in liquid form, cooling the oxygen-containing recycle liquid fraction (18a) in a counter-current subcooler (100), evaporating the cooled oxygen-containing recycle fraction (18b) in the top condenser-evaporator (13), re-compressing the evaporated oxygen-containing recycle fraction (29) in a re-compressor (30), and introducing the re-compressed recycle fraction (31, 32) into the lower region of said single column (12),

wherein

the main heat exchanger (9) and the counter-current subcooler (100) are formed as an integrated heat exchanger (101),

the integrated heat exchanger (101) having a first group of passages for said first return stream (16, 23), which extend from the cold end of said integrated heat exchanger (101) to the warm end of said integrated heat exchanger (101),

said first return stream (16, 23) being introduced into said first group of passages (102) at the cold end of said integrated heat exchanger (101), and flowing through said integrated heat exchanger (101) to the warm end of said integrated heat exchanger (101) and,

during passage through said integrated heat exchanger (101), said first return stream is brought into indirect heat exchange with both said liquid recycle fraction (18a) and said the feed air stream (8), and

the cooled feed air stream (11) is withdrawn from said integrated heat exchanger (101) in completely gaseous form and is fed into said single column (12) in completely gaseous form.

2. A process according to claim 1, wherein said integrated heat exchanger is a single plate-type heat exchanger block.

3. A process according to claim 1, wherein said single column is the only distillation column of said distillation column system.

4. A process according to claim 1, further comprising withdrawing a further oxygen-containing fraction (14a) from said single column (12) in liquid form, cooling the further oxygen-containing liquid fraction (14a) in said integrated heat exchanger (101), evaporating the cooled further oxygen-containing liquid fraction in the top condenser-evaporator (13), warming the evaporated further oxygen-containing fraction (19) in said integrated heat exchanger (101) in counter-current flow to air, and

expanding the warmed evaporated further oxygen-containing fraction in an expansion machine (21) to produce work,

wherein the temperature of the further oxygen-containing liquid fraction (14a) as introduced into said integrated heat exchanger (101) is higher than the temperature of the cooled feed air stream (11) withdrawn off from said integrated heat exchanger (101).

5. A process according to claim 4, wherein, before the work-producing expansion, the warmed evaporated further oxygen-containing fraction is warmed up in counter-current flow to air in said integrated heat exchanger.

6. A process according to claim 4, wherein said oxygen-containing recycle fraction is removed from said single column at an intermediate point which is located at least one theoretical or practical plate above the point at which said further oxygen-containing fraction is removed from said single column (12).

7. A process according to claim 4, wherein said expansion machine (21) is coupled mechanically to said re-compressor (30).

8. A process according to claim 1, wherein said re-compressor (30) is constructed as a cold compressor.

9. A process according to claim 1, wherein said re-compressed recycle fraction (31) is cooled in said integrated heat exchanger (101) before being introduced into the lower region of said single column (12), and said re-compressed recycle fraction (32) is withdrawn from said integrated heat exchanger (101) in completely gaseous form and fed into said single column (12) in completely gaseous form.

10. An apparatus for cryogenic air separation in a distillation column system, comprising:

at least one single column (12),

a main heat exchanger (9) for cooling a compressed feed air stream (6, 8) in counter-current flow to a first return stream (16, 23) from the distillation column system,

means for introducing a cooled feed air stream (11) into said single column (12),

means for removing a nitrogen-rich fraction (15) from the upper region of said single column (12),

a top condenser-evaporator for condensing at least part of the nitrogen-rich fraction,

means for introducing condensed nitrogen-rich fraction (52) from said top condenser-evaporator (13) into said single column (12) as reflux,

means for withdrawing an oxygen-containing liquid recycle fraction (18a) from said single column (12),

a counter-current subcooler (100) for cooling down liquid recycle fraction (18a),

means for introducing cooled recycle fraction (18b) into said top condenser-evaporator (13),

a re-compressor (30) for compressing evaporated recycle fraction (29) from said top condenser-evaporator (13), and

means for introducing re-compressed recycle fraction (31, 32) into the lower region of said single column (12),

wherein

said main heat exchanger (9) and said counter-current subcooler (100) are formed as an integrated heat exchanger (101),

said integrated heat exchanger (101) having a first group of passages (102) for the first return stream (16, 23), which extends from the cold end of said integrated heat exchanger to the warm end of said integrated heat exchanger,

the cold end of said integrated heat exchanger (101) being connected to means for introducing the first return stream (16, 23) into said first group of passages,

the warm end of said integrated heat exchanger (101) being connected to means for withdrawing the first return stream (16, 23) from said first group of passages,

the integrated heat exchanger (101) being constructed so that, during operation, the first return stream (16, 23) is brought into indirect heat exchange with both liquid recycle fraction (18a) and feed air stream (8), and

the passages in the integrated heat exchanger (101) are arranged so that, during operation, cooled feed air stream (11) is withdrawn from said integrated heat exchanger (101) in completely gaseous form and is fed into said single column (12) in completely gaseous form.

* * * * *