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(54) METHOD AND APPARATUS FOR SEPARATING DESUBLIMATABLE COMPONENTS FROM GAS MIXTURES

(71) We, KERNFORSCHUNGSZENTRUM KARLSRUHE GESELLSCHAFT MIT BESCHRAENKTER HAFTUNG, formerly GESELLSCHAFT FÜR KERNFORSCHUNG M.B.H., of 5 Weberstrasse, D-7500 Karlsruhe 1, Federal Republic of Germany; a German body corporate do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:-

The present invention relates to a method, and apparatus for practicing the method, for separating desublimatable components from gas mixtures containing an inert gas on the surfaces of a heat exchanger apparatus.

The desublimation of such components is of significance, for example, in the case in which UF_6 (uranium hexafluoride) contained in inert gas streams of high intensity must be separated, in some cases, until a final concentration of less than 1 ppm has been attained.

Due to the vapor pressure curve of UF_6, a final temperature of about $-120°C$ is required for the desublimation of UF_6 at operating pressures of about 1 atmosphere absolute. In the past, countercurrent systems have been used in order to keep the costs involved in obtaining the required low temperatures for the high-intensity inert gas stream to be processed within economical limits. Such a low temperature separator system operates in a discontinuous manner because when the separator becomes charged, it must be emptied by cutting off the process stream. During the emptying operation, the separator system is heated and thus is available for further separating operation only after it has again been cooled to the required final temperature. It is therefore important for economical separator operation that the pressure drop at the separator does not only have a low absolute value, but that the relative change in the pressure reduction during switching from a charged to an empty separator be as small as possible and that nevertheless the separator efficiency be as high as possible.

Methods and apparatus are known to cool gases, such as disclosed in German Auslegeschriften No. 10 03 240 and No. 10 37 489, but with the known methods and apparatus it is neither possible to obtain uniform layer formation of desublimatable components on separator surfaces nor a constant low pressure reduction at a constant final temperature.

It is therefore a primary object of the present invention to provide a separating method which makes it possible to operate with a very slight and simultaneously constant (in time) pressure reduction and still charge a significant fraction of the separator volume with the desublimating component of the gas mixture while at the same time, for economic reasons, not bringing the lowest temperature impressed on the gas stream to be processed to a value lower than what is absolutely required, in view of the vapor pressure curve of the desublimating component, to obtain the desired degree of separation.

A further object of the present invention is to provide an apparatus for performing the separating method.

According to the present invention there is provided a method for separating a desublimatable component from a gas mixture containing an inert gas onto the surface of an indirect heat exchanger apparatus having a heat exchanger separator containing a separator passage, said separator having a cold end and a warmer end, comprising: producing a desired starting temperature profile in the heat exchanger separator, flowing the gas mixture through the separator from the warmer end toward the cold end to build up a desired layer thickness profile in a direction opposite the direction of flow of the gas mixture by providing a nonstationary temperature distribution profile in the heat exchanger separator to shift the position of the dew point at a controlled speed from a given point near or at the cold end of the heat exchanger separator at the beginning of the separation process to the warmer end of the separator and thereby build up the desired layer thickness profile and charge the separator with the desublimate, controlling change in the nonstationary temperature profile and thereby controlling the displacement speed of the dew point along the surface of the separator by supplying additional cooling energy to the separator, the additional cooling energy being regulated in time and regulated to produce a constant final temperature in the separator to achieve a desired purity for the inert gas or to achieve a desired degree of separation of the desublimate, terminating the flow of the gas mixture into the separator after full charging of the separator, then evacuating any remaining inert gas in the separator, and thereafter recovering the desublimate by heating up the separator and exhausting the desublimate.

In the process of the present invention, by removing the desublimatable component from the gas mixture, a "pure" gas is formed which is a cooled inert gas that emerges from the separator passage and which then passes through a pure gas passage where it is reheated to its starting temperature. Between passage from the separator passage to the pure gas passage, the gas passes through a final cooler region in which supersaturation produced in the separator passage can be reduced.

In the present invention, the process during the separation process enters the heat exchanger at a temperature T_1 and a pressure P_1. At the end of the separator passage, the gas, from which the major portion of the desublimatable component has been removed, has a temperature T_2 and a pressure P_2. The gas passes through the final cooler passage and emerges from it at a temperature T_3 and a pressure P_3. The gas then passes through the pure gas passage where it is reheated and leaves the heat exchanger at a temperature T_4 and a pressure P_4.

The initial temperature profile used at the start of the separation process can be obtained either by a direct cooling process where the final cooling passage is cooled by a countercurrent heat exchanger which is integrated in the separator block or by an indirect cooling process wherein a separately arranged final cooler is used.

According to the present invention there is provided apparatus for separating a desublimatable component from a gas mixture containing an inert gas in a countercurrent heat exchanger comprising:

(a) a raw gas separator passage in which the desublimatable component of the gas mixture desublimates;

(b) a pure gas passage in which cooled inert gas which emerges from the separator passage is reheated to its starting temperature and which is parallel in countercurrent flow and thermally connected to the separator passage;

(c) at least one auxiliary passage through which an auxiliary stream can flow and which is thermally connected to the raw gas passage and the pure gas passage;

(d) means for supplying cooling energy to the auxiliary passage and providing a nonstationary temperature distribution profile in the heat exchanger;

(e) a final cooler having a passage connecting the separator passage and pure gas passage on the cold end, and in which it is possible to reduce supersaturation produced in the separator passage; and for the case of snow formations

(f) a filter downstream of the separator passage for collecting crystallites of the desublimate which are carried along by the gas stream.

Generally, the heat exchange apparatus contains a precooling portion before the separator passage and the process gas enters the heat exchanger in the precooling portion. The precooling portion of the apparatus further includes that portion of the apparatus where the reheated pure gas leaves the heat exchanger. The precooling portion and the separator passage form part of a heat exchanger block.

The particular advantages of the present invention are therefore that a countercurrent separator system is formed with which it is possible to operate with a temperature profile moving in but one direction, the dew point of the component being shifted in a controlled manner from the cold to the warmer end. Preferably, this shift of the dew point is obtained by means of a controlled additional or auxiliary cooling stream. The present invention achieves a practically fixed final separator temperature so that a uniform layer formation of the desublimating component (e.g. UF_6) is produced in the separator passage. At the beginning of the separation process, the separator preferably is operated with a very steep temperature gradient at the cold end so that it operates with high driving temperature differences. Then the initially possibly-occurring supersaturation of the component (e.g. UF_6) in the pure gas can be reduced to no measurable traces (e.g. less than 0.1 ppm for UF_6) in the final cooler region of the separator in a flow region having extremely short diffusion paths. By controlling the speed of travel of the dew point the buildup of the layer of the desublimatable component (UF_6) can be controlled so that the desublimatable component produces a uniform cross-sectional covering on 70-80% on the free cross section of the separator passage and the there-occurring slight increase in pressure loss is compensated in the other parts of the separator apparatus as a result of charges in density and viscosity with decreasing temperature, to such an extent that the reduction in pressure in the total system during the charging process remains practically constant.

The accompanying drawings, in which like numbers indicate like parts, illustrate examples of presently preferred embodiments of the invention and, together with the description, serve to explain the principles of the invention.

Figure 1a shows the temperature profile (separator temperature T) as a function of the separator length L at various times during a typical separation process performed according to the present invention, including the temperature profile at 0 hours, 2.5 hours, and 5 hours.

Figure 1b shows the corresponding time sequence of the resulting pressure reduction $\Delta P_A = P_1 - P_2$ as a function of the separator length L, for the embodiment of Figure 1a.

Figure 1c shows a corresponding time sequence of the coverage of the surface of the free separator cross section F at every point in the separator passage, which has the temperature T_1 at the warmer end and the temperature T_2 at the cold end, for the embodiment of Figure 1a.

The values measured for the pressure drop and the charges in Figures 1a, 1b and 1c apply to a specific separator charge of 135 kg UF_6/h and 15,000 m^3 of gas mixture/h per m^2 cross section of the separator passage at an average gas pressure of 170 Torr.

Figure 2a shows the time curve of the temperatures at various points in a separator apparatus during a separation process which is performed with controlled deformation of the temperature profile in accordance with a typical embodiment of the present invention. The temperature T_3 is -126°C.

Figure 2b shows two time curves for the differential pressure $\Delta P_A = P_1 - P_2$ and the differential pressure $\Delta P_A = P_1 - P_4$, during the same separation operation as represented by Figure 2a and which is performed with controlled deformation of the temperature profile.

Figure 3a shows, in full line, the cooling energy "Q" as a function of the initial cooling period "t" before starting a typical separation process performed according to the present invention wherein the initial cooling time is 60 minutes and the cooling energy during the initial cooling of a separator block is supplied by a stream of pure hydrogen of 9.5 kMol/h.

Figure 3b shows, in full line, a measured temperature profile at the begining of the separation process, obtained during the initial hydrogen cooling cycle shown in Figure 3a.

Figure 4 shows a schematic view of one embodiment of a separator heat exchanger apparatus according to the present invention.

Figure 5 shows a front elevational view of another embodiment of a separator heat exchanger apparatus according to the present invention.

The present invention is based on the surprising realization that a separator can be operated during a separating process with changing temperature gradients and changing driving temperature differences and that this mode of operation can be varied within wide limits even with mixtures having a great tendency to supersaturate, which are of particular interest in practice, without a reduction in the separating efficiency as a result of condensation in the gaseous phase and snow formation.

Experimental tests with stationary temperature distribution have shown that a mode of operation with an almost constant temperature profile for the separator and a shift of this profile toward lower temperatures does not permit favorable charging of the separator. However, experiments with nonstationary temperature gradients in accordance with the present invention have shown that a very high charging capacity can be realized with low pressure losses in an intentionally unilaterally cooled separator which receives constant additional cooling energy during the separator operation.

If, as in the case of a UF_6 mixture, supersaturation occurs during the separation process and if - as has been demonstrated in experiments - no snow formation occurs in the core of the stream during any conceivable modes of separator operation and in spite of high supersaturation, the separating project can be divided into two steps:

1. The predominant portion of the desublimate is separated in the separator itself in an optimum manner with respect to high capacity and low pressure losses without consideration of a high residual content of the supersaturated component in the discharged gas.
2. Thereafter, the reduction or elimination of the supersaturation in a flow area having sufficiently short diffusion lengths.

In order to realize pressure losses ΔP_A which are as low as possible with a high charging capacity in the separator passage, a separation profile is produced in the process of the present invention which is distinguished by a uniform reduction in size of the separator passage over as long a length of the separator block as possible. The optimum lower profile of the remaining cross section may here be selected, either with respect to a constant pressure loss per unit length or a constant flow speed of the process gas, so that acceleration and deceleration phenomena in the separator passage are avoided. In order to fully utilize this basic optimization rule, it is necessary to consider the change in density of the process gas as a result of changes in temperature along the separator passage, and achieve a correspondingly controlled shift in the dew point, and with it of the entire separation profile, toward the warm end of the separator during the separator operation.

Experiments have shown that a technically particularly simple mode of operation results, which is very effective respect to charging capacity and resulting pressure losses, if during separator operation a controlled cooling energy Q_A is fed to the separator system, which cooling energy goes beyond the sum of the cooling requirement needed to remove the desublimation head Q_D and the cooling requirement needed to cover temperature losses which occur in the system due to such factors as insulation losses. The use of such a controlled cooling energy Q_A results in the desired continuous shift of the dew point and of the separation profile as it is shown in Figures 1 and 2, which show measuring results from a typical separation experiment. In this way, it is possible to limit the rise in the differential pressure $\Delta P_A = P_1 - P_2$ in the separator passage during the entire separating process by about 7 Torr with a charge of about 50% of the available separator volume; where P_1 is the pressure at the warmer end of the separator and P_2 is the pressure at the cold end of the separator passage. This rise is additionally compensated by a reduction in the differential pressures produced in the remaining passages of the separator system which are free of UF_6 as a result of a reduction in temperature during the separation process. This means, as shown in Figure 2b in the difference between the entrance pressure and the exit pressure $P_1 - P_4$ over the entire separator system, that with this type of control of the separation process, there results practically no rise in the differential pressure $\Delta P = P_1 - P_4$ over the entire separator system.

Since with the practiced nonstationary mode of operation of the separator, the temperatures of the gas streams in the separator system are changing continuously, prior calculation of the required cooling energy necessitates measurement or computation, respectively, of all quantities of heat fed to and removed from the separator system. The heat energy balance is determined from the measured values obtained and from that the additional cooling energy required in the cooling stream to produce the desired continuous drop in the temperature profile in the separator is calculated. With a permanent separator program routine, an optimum time sequence of the cooling energy requirement can be determined once, and this optimum time sequence then can be used for technical operation of the separator system.

With a separator which is well insulated in a known manner, insulation losses need not be considered. In that case, the total cooling energy Q_{total} required to be introduced into the separator is then a combination of (1) the desublimation heat energy Q_D to be dissipated and (2) the cooling energy Q_A which is identified as the excess cooling energy and which serves to continuously lower the temperature in the separator block. The desublimation heat energy Q_D is proportional to the flow intensity of the component to be separated, i.e., to the product of the total flow intensity (flowrate) "m" and the concentration "N" of the component to be separated. Likewise, the cooling energy Q_A is proportional to this product, i.e., to the concentration N of the component to be separated and the total flow intensity "m", so that with equal quantities separated the same temperature state will always result in the separator. This assures that with changing contents of the component to be separated in the raw process gas, the same final state of charge is attained in the full separator. The total cooling energy results from the following equation:

$$Q_{total} = K_A \cdot \dot{m} \cdot N \qquad (1)$$

During separator operation, the temperatures T_1 and T_4 at the warm end of the precooler remain practically constant and generally no cooling energy is supplied in this portion of the apparatus. A portion of the total cooling energy, however, is fed to the cold end of the separator system through the process gas stream $\dot{m}$. The process gas stream at the cold end of the separator has a temperature T_2 as it emerges from the separator passage and is then further cooled in a final cooling passage to a colder temperature T_3 which is the lowest temperature produced in the separator apparatus. The amount of cooling energy introduced at the cold end is referred to as $Q_{mainstream}$ and is proportional to the flow intensity of the process gas and the temperature difference ΔT which is created at the cold end between the temperature T_2 at the end of the separator passage and the temperature T_3

in the final cooler passage. Thus,

$$Q_{mainstream} = \dot{m} \cdot c_p \cdot \Delta T \qquad (2)$$

where $\Delta T = T_2 - T_3$.

The rest of the required total cooling energy must be supplied through an additional auxiliary cooling stream in a third passage of the separator. The auxiliary cooling stream has a cold entrance temperature T_{K1}, and a warmer exit temperature T_{K2}. This auxiliary cooling stream generally is introduced at the lowest separator temperature T_3. The cooling energy supplied through the auxiliary cooling stream is referred to as Q_K, and can be regulated by a change in the flow intensity $\dot{m}_K$ of the auxiliary cooling stream under consideration of the difference ΔT_K between its exit temperature T_{K2} and its entrance temperature T_{K1} according to equation (3)

$$Q_K = \dot{m}_K \cdot c_{pK} \cdot \Delta T_K \qquad (3)$$

The required auxiliary cooling energy Q_K is also given according to equation (4) as follows:

$$Q_K = Q_{total} - Q_{mainstream} \qquad (4)$$

The required auxiliary cooling stream intensity $\dot{m}_K$ can then be calculated from the required auxiliary cooling energy $Q_K = Q_{total} - Q_{mainstream}$.

Thus, by introducing equations (1), (2) and (3) into equation (4), and solving for $\dot{m}_K$, it is possible to determine the cooling stream intensity $\dot{m}_K$ required for the desired cooling speed of the separator according to equation (5) as follows:

$$\dot{m}_K = \frac{K_A \cdot \dot{m} \cdot N - \dot{m} \cdot c_p \cdot \Delta T}{c_{P,K} \quad T_K} \qquad (5)$$

where $\dot{m}$ is the measured values for the total throughput of the process gas stream, ΔT is the temperature difference in the process gas at the cold end of the separator, N is the concentration in the process gas of the component to be separated at the entrance, and ΔT_K is the temperature difference of the auxiliary cooling stream. The constant K_A determines the desired cooling speed of the separator and is thus determinative for the thickness of the coating layer. In the course of experiments, this parameter K_A has been optimized with a view toward reaching a compromise between maximum separator capacity and low pressure losses. A favorable compromise is a maximum charge of the given separator up to 50% of the separator volume with a total pressure reduction at the separator passage of 7 Torr, as is shown in the typical experimental embodiment of Figure 2. This value for constant K_A, once determined, is given for the calculation of the intensity of the auxiliary stream $\dot{m}_K$. In experiments regulated by means of direct flow intensity control of $\dot{m}_K$, tests were made with variations of the UF_6 concentration in the raw gas within a range from N = 0.1% to 0.5%, and operating pressures in a range from 100 to 300 Torr. Although the separating times in these cases were inversely proportional to the UF_6 intake in a range from 3 to 15 hours, the separation profile was always practically the same.

The initial temperature profile used at the start of the separation process can be obtained either by a direct cooling process where the final cooling passage is cooled by a cross-current heat exchanger which is integrated in the separator block or by an indirect cooling process wherein a separately arranged final cooler is used. Initial cooling of the separator to obtain the starting temperature profile is advisably effected by cooling the final cooler passage by a cross-current heat exchanger which is integrated in the separator block, and by transfer of the cooling energy by heat conduction into the separator block when the process gas stream is shut off.

With a separately arranged final cooler, the separator can be cooled with the aid of a stream of inert gas which receives the corresponding cooling energy in the final cooler. This cooling method furnishes a starting cooling energy of similar magnitude and just as favorable temperature profiles for the start as the direct cooling process. This can be seen in the comparisons shown in Figures 3a and 3b. Figure 3a shows in full line, the cooling energy "Q" as a function of the initial cooling period "t" for a typical separation process performed according to the present invention wherein the initial cooling time is 60 minutes and the cooling energy during initial cooling of a separator block is supplied by a stream of hydrogen of 9.5 KqMol/h. As a comparison, calculated results for cooling using an integrated cross-current heat exchanger for cooling the final cooler passage and applicable for purely nonstationary heat conduction along the same separator block, is shown in dashed line. Figure 3b shows, in full line, a measured temperature profile at the beginning of the separation process obtained during the initial hydrogen cooling cycle shown in full line in Figure 3d. As a comparison, the dashed line in Figure 3b shows a calculated temperature profile for the purely nonstationary longitudinal heat conduction in the separator block during direct cooling starting from room temperature.

In the process of the present invention, after the desired layer thickness profile of the desublimate is obtained in the separator passage, the process gas stream is cut off so that it no longer flows into the heat exchanger apparatus. The heat exchanger apparatus is then heated to discharge the desublimate from it. During this heating, the desublimate is heated to a temperature which causes the desublimate to change to its liquid or vapor state and can then be removed from the heat exchanger in the liquid or vapor state. The heating of the heat exchanger can be performed, for example, by passing a heating medium through the auxiliary passage of the heat exchanger. In one embodiment of the present invention, the heat exchanger separator is disposed in a pressure vessel, and the pressure vessel can be heated to provide further heat imput for discharge of the desublimate.

After the desublimate is discharged, the separator is warm and empty, and can now be cooled again in the manner discussed previously to obtain a starting temperature profile for a further separation cycle.

Turning now to the drawings, there is shown in Figure 4 a countercurrent heat exchanger in the form of a compact heat exchanger block 25 which is mounted in a pressure vessel 31 (shown in broken line). Heat exchanger block 25 includes a raw gas separator passage 26 and a pure gas passage 21 which is in countercurrent relationship to separator passage 26 and which communicates with separator passage 26 at their lower ends via a final cooler passage 24. The lower ends of separator passage 26 and pure gas passage 21 are their colder ends. Heat exchanger block 25, in the case of UF_6 separation, is provided with multi-entry ribs (not shown) (rib height 11.8m; serrated; 10 ribs per inch with a rib thickness of 0.2mm) over the entire length of separator passage 26. Pure gas passage 21 has smooth ribs (not shown) over its entire length and does not require a distributor at its cold end.

The heat exchanger block 25 further includes a pre-cooling portion 40 which is at the top of block 25 and outside of pressure vessel 31. Pre-cooling portion 40 contains a segment 41 which communicates with separator passage 26 and a segment 42 which communicates with pure gas passage 21. Pre-cooling portion 40 and the actual separator portion below it are combined to form a homogeneous compact heat exchanger block 25.

The heat exchanger apparatus further includes at least one auxiliary flow passage 27 which is in heat exchange relation with separator passage 26, but does not communicate therewith. Auxiliary flow passage 27 can extend the full length of separator passage 26 or a portion thereof. As shown in the embodiment of Figure 4, auxiliary flow passage 27 extends substantially the entire length of separator passage 26. Auxiliary passage 27 can function as an auxiliary cooling passage for supplying the needed auxiliary cooling energy Q_K to the separator system. In addition, auxiliary passage 27 can serve further as a heating passage to heat the separator during the emptying operation where the desublimate is removed from the separator. When auxiliary passage 27 functions as an auxiliary coolant passage, the coolant enters passage 27 at its lower end via an auxiliary cooling stream entrance 28 at a temperature T_{K1} and leaves via lines 37′ or 37″ at its upper end at a temperature T_{K2}.

Raw process gas, which contains the desublimatable component, enters the heat exchanger at its top without use of a distributor (full end tank) from a process gas inlet line 1 which is connected with pre-cooling segment 41. Pure process gas from which the desublimatable component has been removed leaves the heat exchanger during the separation process via a gas outlet line 2 which is connected with pre-cooling segment 42. When the separator becomes fully charged, the process gas inlet line 1 is cut off, and any remaining inert gas is evacuated through pure gas passage 21 and an evacuation line 4 so as not to unnecessarily remove desublimate. After evacuation of any remaining inert gas is complete, the heat exchanger is heated to evaporate the desublimate. The evaporated desublimate leaves the heat exchanger by flowing through separator passage 26 to an outlet gas line 11 which is connected on one end to a desublimate discharge line 3 and on its other end to raw gas inlet line 1. This arrangement of the outlet gas line 11 and desublimate discharge line 3 prevents backflow of the desublimate through non-tight valves of the discharge line 3 to the pure gas side of the separator apparatus. As can be seen in Figure 4, valves are provided to control flow in gas lines 1, 2, 3, 4, and 11 and all of these valves are located at the warm end of the separator, and thus these valves do not have to be in the form of low temperature valves. Further, all of these valves which control flow in gas lines 1, 2, 3, 4 and 11 do not have to meet high tightness requirements during passage.

As shown in Figure 4, a filter 23 in the form of an integrated wire mesh is provided downstream of separator passage 26 in a lower end tank portion 22 of the separator, and the gas coming from separator passage 26 flows through filter 23 and thereafter enters into the full cross section of pure gas passage 21. Crystallites of the desublimate which are carried along by the gas stream are collected in filter 23. According to previous operating experience, filter 23 need only be capable of retaining a maximum of 1 part per thousand of the separated UF_6 quantity. Therefore, only a small filter surface and minimum filter volume are required which can be integrated in a simple manner.

In the final cooler 24, a passage 18 is connected to the separator passage 26, and a passage 19 to the pure gas passage 21. Passages 18 and 19 are cooled by a flow of heat transfer medium flowing in a third passage 20. In this embodiment, the heat exchanger 24 is integrated in block 25 and is provided with side tanks 29 and 30. The passage requirement for the crosscurrent cooling heat exchanger preferably is selected to be identical with the number of auxiliary passages 27. This will result in only relatively small side tanks 29 and 30 at the cold end of the separator apparatus for the coolant supply needed for cooling final cooler passages 24.

Cooling of the warm, empty separator is effected with the aid of the crosscurrent heat exchanger which provides cooling of final cooler passage 24 and which is charged with a liquid coolant (e.g., refrigerant 12, i.e., CCl_2F_2). The coolant is disposed in a closed circuit formed by a coolant supply line 6 and a coolant outlet line 5, under the respective vapor pressure given by the highest occurring temperature, e.g., 10 atmospheres absolute for T_{max} = 40°C at the start of the cooling period and about 1 Torr = −120°C at the end of the cooling period. The coolant is introduced into side tank 29 of the crosscurrent heat exchanger from coolant supply line 6 through a coolant inlet line 34 and discharged from side tank 30 of the crosscurrent heat exchanger through a discharge line 35 to coolant out line 5.

In one embodiment of the present invention, the coolant supplied to the crosscurrent heat exchanger which is used to cool final cooler passage 24 may advantageously also be supplied to auxiliary passage 27 as a liquid auxiliary cooling stream, since it would in any case be at the separator temperature (T_3) when it is introduced into auxiliary passage 27 at entrance 28. The coolant is supplied from coolant supply line 6 to entrance 28 of auxiliary passage 27 through a line 36 which branches off from line 34. In this embodiment of the invention, the coolant circuit for cooling the final cooler passage 24 and that for the auxiliary passage are, in part, portions of the same circuit.

In another embodiment of the present invention, the coolant circuit for cooling the final cooler passage 24 and that for the auxiliary cooling passage 27 are completely separated from each other. A particular reason for the use of separate coolant circuits is given if a two-phase mixture of a suitable coolant is introduced into auxiliary passage 27 for cooling in order to produce a specially shaped temperature profile in separator block 25.

If the auxiliary cooling stream supplied to auxiliary passage 27 is to be regulated completely decoupled from the cooling circuit used to cool final cooler passage 24, the line connections 9 and 10 shown in dashed lines in Figure 4 are applicable if the coolant from the heating system also is not to be used.

To empty the separator after it becomes charged with desublimate, heat energy must be supplied to the separator. Heat energy can be supplied to the apparatus by passing a warm gaseous coolant through auxiliary passage 27. The warm gaseous coolant enters the top of auxiliary passage 27 through a line 37′ or a line 37″. Lines 37′ and 37″ are supplied with warm gaseous coolant from a heating vapor line 7. In addition, heating energy can be supplied to the separator through pressure vessel 31 which acts as a heat jacket which surrounds heat exchanger block 25. Thus, warm gaseous coolant from heating vapor line 7 can be passed into the top of pressure vessel 31 through an inlet line 38 to provide jacket heating of the separator. The warm gaseous coolant which enters inlet line 38 and line 37′ or line 37″ can be steam. As the warm gaseous coolant passes through pressure vessel 31 and auxiliary passage 27, it condenses to its liquid state. A condensate discharge 32 is located at the bottom of pressure vessel 31 and feeds into a condensate outlet line 8. Similarly, condensate can be discharged from auxiliary passage 27 through a discharge line 45 which feeds into condensate outlet line 8.

Heating and discharging of the desublimate by charging auxiliary passage 27 and pressure vessel 31 with warm, gaseous coolant through lines 37′ or 37″ and 38 from heating vapor line 7, with controlled discharge of the condensate through condensate discharge lines 32 and 45 and condensate outlet line 8, produces rapid heating of the desublimate to the desired temperature level, and proper intake of the required evaporation heat to points which assure evaporation of the desublimate.

An optimum solution for providing cooling energy to cool auxiliary passage 27, for providing cooling energy to auxiliary passage 27, and for providing heat energy to auxiliary passage 27 during heating, is the use of a single heat transfer medium for all three objectives. This should be possible with low maximum system pressures (about 1 atmosphere absolute) if instead of the conventional cooling media ("Freon" Registered Trade Mark), there is used butane or pentane or similar low-boiling-point substances which are liquid at the final separator pressure under very low gas pressures.

Turning now to Figure 5, there is shown an embodiment of a separator apparatus which is similar to that shown in Figure 4, and where the same reference numerals as in Figure 4 are used to indicate like parts. Auxiliary passage 27 is not shown in Figure 5, but line 36 for supplying a cooling liquid auxiliary stream and entrance 28 to auxiliary passage 27 are shown in Figure 5. In view of the extremely compact configuration of the separator, jacket heating of it and the connected lines can be effected during emptying of the separator by a combination of steam heat and vacuum insulation as shown in Figure 5. The part of the separator block 25 disposed below the process connections is suspended in pressure vessel 31 which receives warm gaseous coolant through line 38 as heating means. In addition, warm gaseous coolant is introduced into auxiliary passage 27 to effect heating. The temperature of the warm gaseous coolant introduced through line 38 is regulated above the condensation point of the warm gaseous coolant by maintaining a suitable pressure in pressure vessel 31, and any steam condensate which may form flows out at the bottom of pressure vessel 31 through condensate discharge 32. When the heating period is terminated, line 38 is closed to prevent further quantities of warm gaseous coolant from entering pressure vessel 38, but there is a quantity of warm gaseous coolant which then remains in pressure vessel 31. During the cooling period which follows heating and emptying of the separator, the warm gaseous coolant remaining in pressure vessel 31 after blocking off of pressure vessel 31 is frozen out on condenser surfaces 33 which are provided on cooler inlet line 34. For this purpose, a coolant fluid is required which has a vapor pressure less than 10^{-3} Torr at the separator temperature.

In the method of the present invention, the controllable change of the nonstationary temperature profile, together with a likewise controllable displacement speed of the dew point, produce a layer thickness profile of the desublimate along the surfaces of the separator with which there preferably is realized an optimized ratio of maximum charging quantity to occurring pressure decrease (P_1 - P_4) at the end of the charging step. During the separation process, the temperature T_2 at the cold end of the separator is kept at values which are necessary for the degree of purity of the remaining gas mixture or for the degree of separation of the desublimate. Supersaturations of the component to be desublimated occurring in gas mixtures having a tendency to supersaturate can be isothermally reduced in flow regions provided for this purpose and having sufficiently short diffusion paths in the range of the final separator temperature (T_2, T_3). The temperature profile before the onset of the separation process preferably is shaped with the use of an auxiliary cooling stream which is fixed in time according to quantity, temperature, composition and direction of flow and which is conducted over the entire length of the separator or parts thereof in auxiliary passage 27. On the other hand, the controllable change in the temperature profile and the controllable displacement speed of the dew point along the surfaces of the separator during the actual separation process can be controlled with the aid of an auxiliary cooling stream which can be regulated in time with respect to quantity, temperature and composition and which flows in auxiliary passage 27 over the entire length of the separator (L) or parts thereof. The quantity, temperature and composition of the auxiliary cooling stream can be controlled under consideration of the momentary heat flow balance of the heat exchanger separator 25 in such a manner that a predetermined excess cooling energy is introduced in such quantities that the desired cooling periods for warmer portions of the separator and the desired displacement speeds of the dew point result therefrom. The auxiliary cooling stream can be a stream of inert gas, or can consist of a liquid heat transfer medium, or can be a two-phase mixture with a measured quantity of liquid component.

The desired layer thickness profile in all sections of the charged raw gas separator passage 26 preferably is determined as an optimized function of the local flow speed, the local density and the local Reynolds number Re of the process gas mixture with utilization of the effective hydraulic diameter of the separator passage 26 as it results from the free cross section at full charge. Preferably, the optimized layer thickness profile produces a constant value for the specific pressure reduction dp/dL along the charging length of separator passage 26, or a constant average flow energy along separator passage 26. The excess cooling energy introduced into the separator 25 can be programmed dynamically on the basis of the momentary heat flow balance and the given values for the concentration and flow intensity under consideration of the momentary values of concentration and flow intensity of the desublimatable mixture component of the gas mixture, the actually-realized charging state and the already-occurred pressure reduction.

Upon completion of the separation process, any remaining pure gas is evacuated to line 4 through pure gas passage 21 and the charged separator is then heated with the aid of an auxiliary heating stream in auxiliary passage 27 to empty the separator of the charged desublimate. The auxiliary heating stream can be in inert gas, or a condensable heat transfer medium, or a liquid heat transfer medium. Together with the heating process that results from the use of an auxiliary heating stream, a heat transfer medium can be introduced into pressure vessel 31 through line 38 so as to externally heat the separator and its connecting lines to aid in emptying the separator of the charged desublimate. The auxiliary heating stream can be regulated in time according to flow intensity and direction by conducting it over the entire length of auxiliary passage 27 or parts thereof so as to provide heat transfer. The desublimate can be discharged from the heated separator 25 by evaporation under pressures which lie below the pressure P_4 of the process gas which leaves through line 4. Emptying of separator 25 can be effected by extracting the desublimate in the form of a gaseous phase or as a liquid phase, after the triple point conditions have been exceeded. During emptying of the separator 25 under high internal pressure, pressure vessel 31 assumes a protective function against discharge of desublimate.

In the apparatus of the present invention, the auxiliary passage 27 is provided with taps or openings to which various lines are connected for supplying cooling or heating streams to auxiliary passage 27. The heat exchanger 25 preferably is a compact heat exchanger and contains a final cooler passage 24 which is cooled by a crosscurrent heat exchanger that is integrated in the heat exchanger. In addition, a filter can be integrated in the heat exchanger.

The portion of the heat exchanger where the actual separation takes place preferably is built into a pressure vessel 31 and preferably is fixed to the cover of the pressure vessel 31 at its upper part and is freely suspended at the lower part. Preferably, all process valves, i.e., those valves which control the flow of the raw process gas, the pure gas and the evaporated desublimate, are disposed outside pressure vessel 31. Pressure vessel 31 can be evacuated so as to provide heat insulation. Further, pressure vessel 31 can be charged with an inert or a condensable heat transfer medium to heat the separator. When a condensable heat transfer medium is used in pressure vessel 31, after the heating operation is completed, any remaining condensable heat transfer medium which remains in the pressure vessel can be frozen out on a ribbed surface 33 of a suitable line that is in pressure vessel 31. This suitable line can be coolant inlet line 34 through which coolant is supplied to cool the final cooler passage 24. The number of auxiliary passages 27 preferably is identical with the number of passages of the integrated crosscurrent heat exchanger used to cool final cooler passage 24.

The process gas separator passage 26 for the raw gas preferably is provided with high ribs of short length with small spacing between ribs in the direction of flow. The length and design of the countercurrent passages 21 and 26 preferably are selected so that with constant gas throughput, the reduction in pressure loss occurring in the empty separator between the temperature distribution at the start of the separating process and the temperature distribution at the end of the separation is equal to the increase in pressure losses from charging of the separator. Suitable design of the ribs in the final cooler passage 24 produces flow regions with sufficiently short diffusion paths in which possibly occurring supersaturation is reduced.

WHAT WE CLAIM IS:

1. Method for separating a desublimatable component from a gas mixture containing an inert gas onto the surface of an indirect heat exchanger apparatus having a heat exchanger separator containing a separator passage, said separator having a cold end and a warmer end, comprising: producing a desired starting temperature profile in the heat exchanger separator, flowing the gas mixture through the separator from the warmer end toward the cold end to build up a desired layer thickness profile in a direction opposite the direction of flow of the gas mixture by providing a nonstationary temperature distribution profile in the heat exchanger separator to shift the position of the dew point at a controlled speed from a given point near or at the cold end of the heat exchanger separator at the beginning of the separation process to the warmer end of the separator and thereby build up the desired layer thickness profile and charge the separator with the desublimate, controlling change in the nonstationary temperature profile and thereby controlling the displacement speed of the dew point along the surface of the separator by supplying additional cooling energy to the separator, the additional cooling energy being regulated in time and regulated to produce a constant final temperature in the separator to achieve a desired purity for the inert gas or to achieve a desired degree of separation of the desublimate, terminating the flow of the gas mixture into the separator after full charging of the separator, then evacuating any remaining inert gas in the separator, and thereafter recovering the desublimate by heating up the separator and exhausting the desublimate.

2. A method as claimed in claim 1, wherein the change of the nonstationary temperature profile and the displacement speed of the dew point are controlled to build up the desired layer thickness profile with which there is realized at the end of the charging step an optimized ratio of maximum charging quantity to occurring pressure decrease over

the entire heat exchanger apparatus.

3. A method as claimed in claim 1 or 2, comprising isothermally reducing any occurring supersaturation by flowing the gas mixture through a flow region located in the cold end of the separator having sufficiently short diffusion paths if the gas mixture has a tendency to supersaturate with respect to the component to be desublimated.

4. A method as claimed in claim 1, 2 or 3, wherein the temperature profile before the onset of the separation process is shaped with the use of an auxiliary cooling stream which is fixed in time according to quantity, temperature, composition and direction of flow and which is conducted over at least a portion of the entire length of the separator in an auxiliary passage.

5. A method as claimed in claim 4, wherein the auxiliary cooling stream is conducted over the entire length of the separator.

6. A method as claimed in claim 4, or 5, wherein the auxiliary cooling stream is a stream of inert gas.

7. A method as claimed in claim 4 or 5, wherein the auxiliary cooling stream consists of a liquid heat transfer medium.

8. A method as claimed in claim 4 or 5 wherein the auxiliary cooling stream is a two-phase mixture with a measured quantity of liquid component.

9. A method as claimed in any preceding claim, wherein the controllable change in the temperature profile and the controllable displacement speed of the dew point along the surface of the separator is controlled with the aid of an auxiliary cooling stream which is regulated in time with respect to quantity, temperature and composition and which flows in an auxiliary passage over at least a portion of the entire length of the separator passage.

10. A method as claimed in claim 9, wherein the auxiliary stream is conducted over the entire length of the separator.

11. A method as claimed in claim 9 or 10, wherein the quantity temperature and composition of the auxiliary cooling stream is controlled under consideration of the momentary heat flow balance of the heat exchanger separator in such a manner that a predetermined excess cooling energy is introduced in such quantities that the desired cooling periods for warmer portions of the separator and the desired displacement speeds of the dew point result therefrom.

12. A method as claimed in claim 9, 10 or 11, wherein the auxiliary cooling stream is a stream of inert gas.

13. A method as claimed in claim 9, 10 or 11, wherein the auxiliary cooling stream consists of a liquid heat transfer medium.

14. A method as claimed in claim 9, 10 or 11, wherein the auxiliary cooling stream is a two-phase mixture with a measured quantity of liquid component.

15. A method as claimed in any preceding claim, wherein the layer thickness profile in all sections of the charged separator passage is determined as an optimized function of the local flow speed, the local density and the local Reynolds number (Re) of the gas mixture with utilization of the effective hydraulic diameter of the separator passage as it results from the free cross section of the separator passage at full charge.

16. A method as claimed in any preceding claim, comprising controlling the build-up of the desired layer thickness profile to produce a constant value for the specific pressure reduction dp/dL along the charging length of the separator passage.

17. A method as claimed in any one of claims 1 to 15, comprising controlling the build-up of the desired layer thickness profile to produce a constant average flow energy along the separator passage.

18. A method as claimed in any preceding claim, wherein the additional cooling energy introduced into the separator to bring about the shift in dew point is controlled dynamically on the basis of the momentary heat flow balance and the given values for the concentration and flow rate under consideration of the momentary values of concentration and flow rate of the desublimatable component of the gas mixture, considering the actually realized charging state, and the already occurred pressure reduction.

19. A method as claimed in any preceding claim, wherein upon completion of the separation process, the heat exchanger is evacuated to remove any remaining inert gas, and the separator is then heated with the aid of an auxiliary heating stream in the auxiliary passage to empty the separator of the charged desublimate.

20. A method as claimed in claim 19, wherein the auxiliary heating stream is an inert gas.

21. A method as claimed in claim 19, wherein a condensable heat transfer medium is used as the auxiliary heating stream.

22. A method as claimed in claim 19, wherein the auxiliary heating stream is a liquid heat transfer medium.

23. A method as claimed in claim 19, wherein the heat exchanger is mounted in a

pressure vessel, and a heat transfer medium is introduced into the pressure vessel to externally heat the separator to aid in emptying the separator of the charged desublimate.

24. A method as claimed in claim 19, wherein the auxiliary heating stream is regulated in time according to flow intensity and direction by conducting it over at least a portion of the length of the auxiliary passage so as to provide heat transfer.

25. A method as claimed in claim 24, wherein the auxiliary heating stream is conducted over the entire length of the auxiliary passage.

26. A method as claimed in claim 19, wherein the desublimate is discharged from the heated separator by evaporation under a pressure which lies below the pressure which exists in the heat exchanger at the point where the gas resulting from the desublimate leaves the heat exchanger.

27. A method as claimed in claim 19, wherein the heat exchanger is mounted in a pressure vessel, the separator is emptied of desublimate under high internal pressures, and the pressure vessel assumes a protective function against discharge of desublimate.

28. A method as claimed in any preceding claim, for separating a desublimatable component from a gas mixture containing an inert gas onto the surface of an indirect heat exchanger apparatus having a heat exchanger separator containing a separator passage, said separator having a cold end and a warmer end, substantially as hereinbefore described and illustrated.

29. A desublimatable component whenever separated from a gas mixture containing an inert gas by a method as claimed in any preceding claim.

30. Apparatus for separating a desublimatable component from a gas mixture containing an inert gas in a countercurrent heat exchanger comprising

(a) a raw gas separator passage in which the desublimatable component of the gas mixture desublimates;

(b) a pure gas passage in which cooled inert gas which emerges from the separator passage is reheated to its starting temperature and which is parallel in countercurrent flow and thermally connected to the separator passage;

(c) at least one auxiliary passage through which an auxiliary stream can flow and which is thermally connected to the raw gas passage and the pure gas passage;

(d) means for supplying cooling energy to the auxiliary passage and providing a nonstationary temperature distribution profile in the heat exchanger;

(e) a final cooler having a passage connecting the separator passage and pure gas passage on the cold end, and in which it is possible to reduce supersaturation produced in the separator passage; and for the case of snow formations.

(f) a filter downstream of the separator passage for collecting crystallites of the desublimate which are carried along by the gas stream.

31. Apparatus as claimed in claim 30, wherein the auxiliary passage is provided with taps to which lines are connected for supplying a heat transfer medium to the auxiliary passage.

32. Apparatus as claimed in claim 30 or 31, wherein the means for supplying cooling energy to the auxiliary passage is a compact heat exchanger.

33. Apparatus as claimed in claim 32, wherein the final cooler is cooled by a crosscurrent heat exchanger which is integrated with the compact heat exchanger.

34. Apparatus as claimed in claim 32, wherein the filter is integrated with the compact heat exchanger.

35. Apparatus as claimed in any one of claims 30 to 34, wherein the separator passage and pure gas passage and the auxiliary passage are arranged in thermal contact to form a compact heat exchanger block which heat exchanger block contains an integrated filter and an integrated final cooler.

36. Apparatus as claimed in any one of claims 30 to 35, wherein the raw gas separator passage and the pure gas passage comprise a desublimator which is built into a pressure vessel to provide thermal insulation by vacuum or for outside heating with heat transfer fluid.

37. Apparatus as claimed in claim 36, wherein the desublimator is fixed to the cover of the pressure vessel and is freely suspended at the lower part.

38. Apparatus as claimed in claim 33, wherein the number of auxiliary passages is identical with the number of passages of the compact heat exchanger used to cool the final cooler passage.

39. Apparatus as claimed in claim 30, wherein the length and design of the countercurrent passages are selected so that with constant gas throughput, the reduction in pressure loss occurring in the empty separator between the temperature distribution at the start of the separating process and the temperature distribution at the end of the separation is equal to the increase in pressure losses from charging of the separator.

40. Apparatus for separating a desublimatable component from a gas mixture

containing an inert gas, substantially as hereinbefore described with reference to and as illustrated in the accompanying drawings.

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Fig.1a

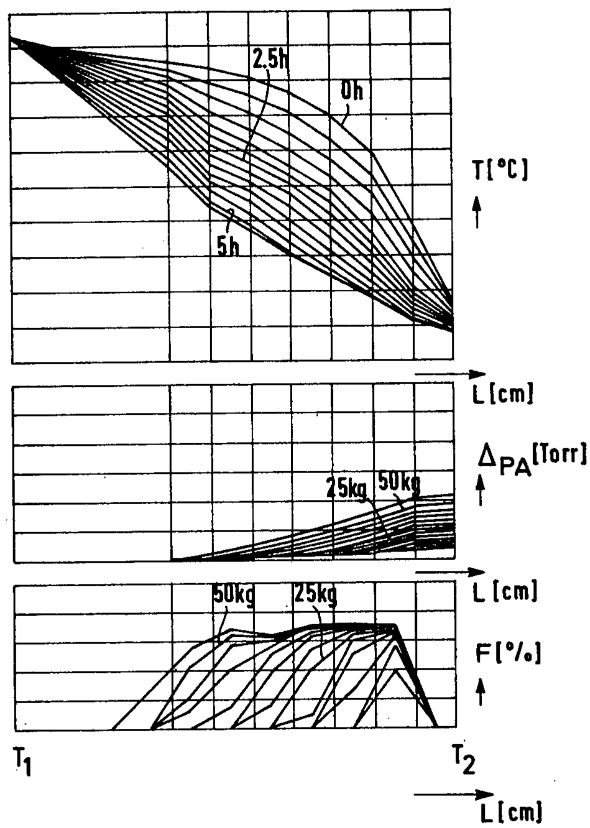


Fig.1b

Fig.1c

Fig.2a

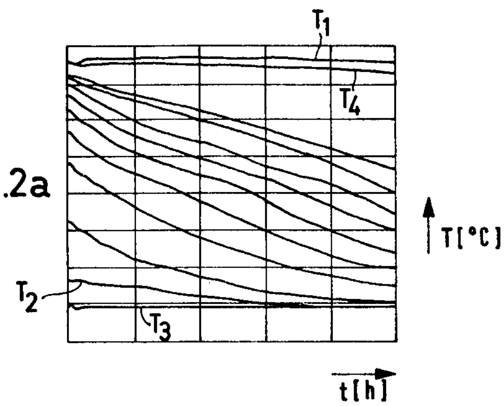


Fig.2b

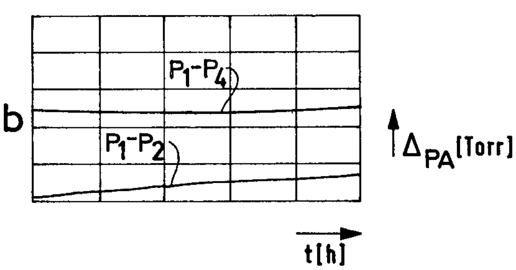


Fig.3a

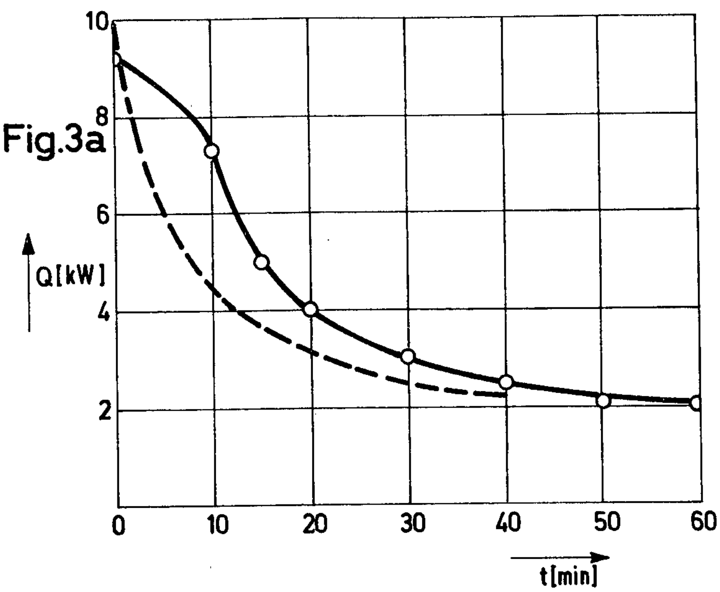


Fig. 3b

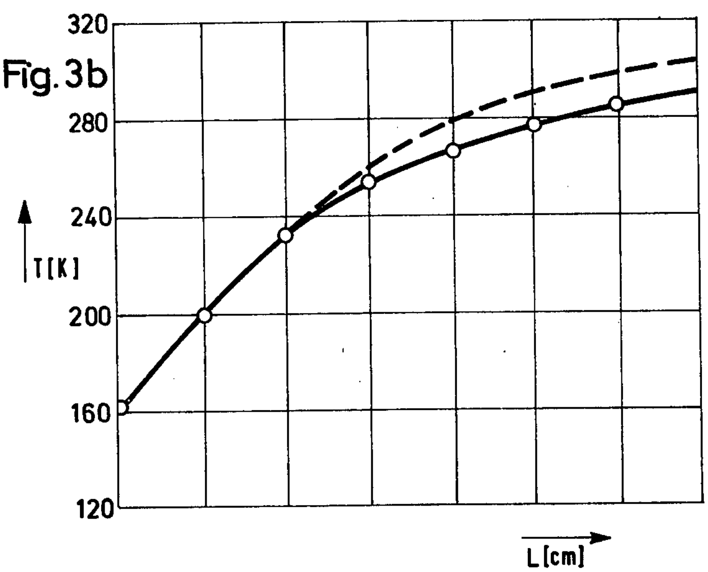
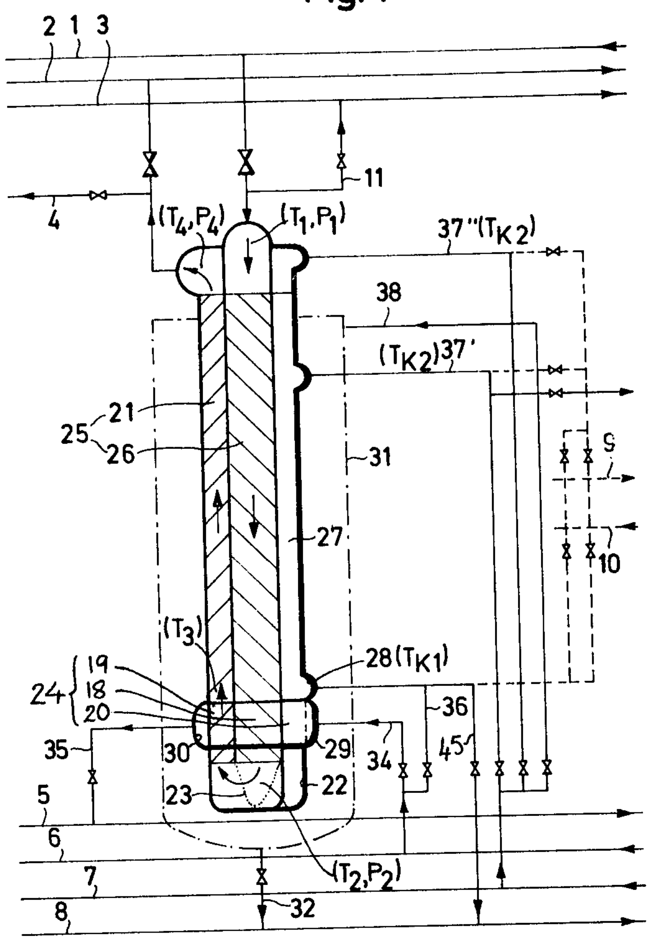


Fig.4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1580169 COMPLETE SPECIFICATION

5 SHEETS *This drawing is a reproduction of the Original on a reduced scale*

Sheet 5

Fig.5