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(54) **GAS SEPARATION LIQUEFACTION MEANS AND PROCESSES**

(57)

**ABSTRACT**

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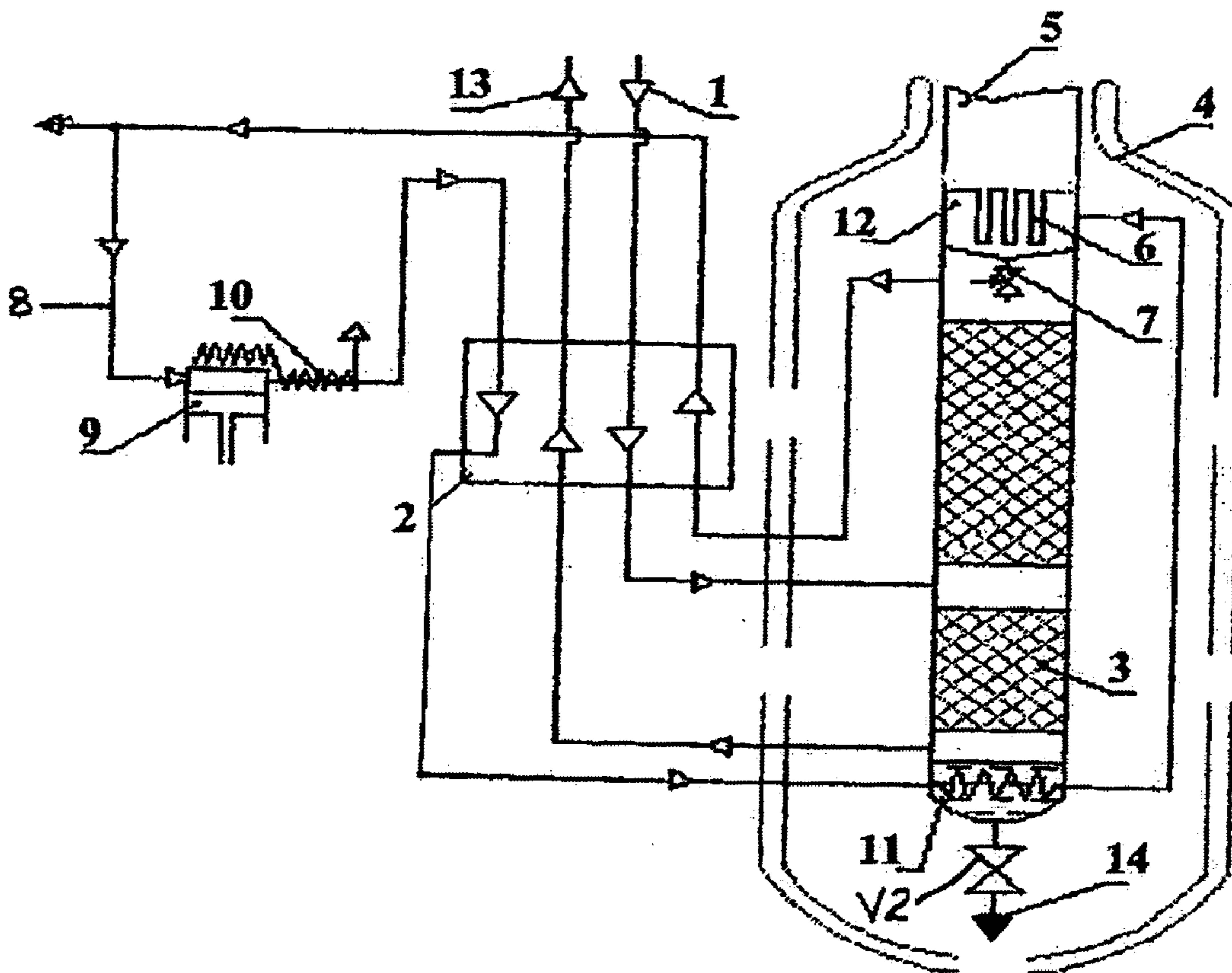
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Single or double column cryogenic gas-separation/liquefaction devices, where refrigeration to the device is supplied by a cryocooler alone or by a combination of a cryocooler and by a Joule-Thompson throttling process, where the gas condensation may occur directly on the cold portion of the cryocooler which may be located inside of the thermally insulated space of the distillation column(s) are disclosed. The system is particularly useful for medical applications, such as providing for safe and economical high-purity oxygen for at-home use. The invention principles include a combined column embodiment for simultaneous production of high-purity liquid or gaseous oxygen and nitrogen. Another double column design offers reduced temperature and pressure separation with easy switching between oxygen and nitrogen extraction or single component extraction. If both gaseous and liquid oxygen are required, oxygen purity of approximately 95% can be produced with good recovery, i.e., with nitrogen purity of approximately 91%.



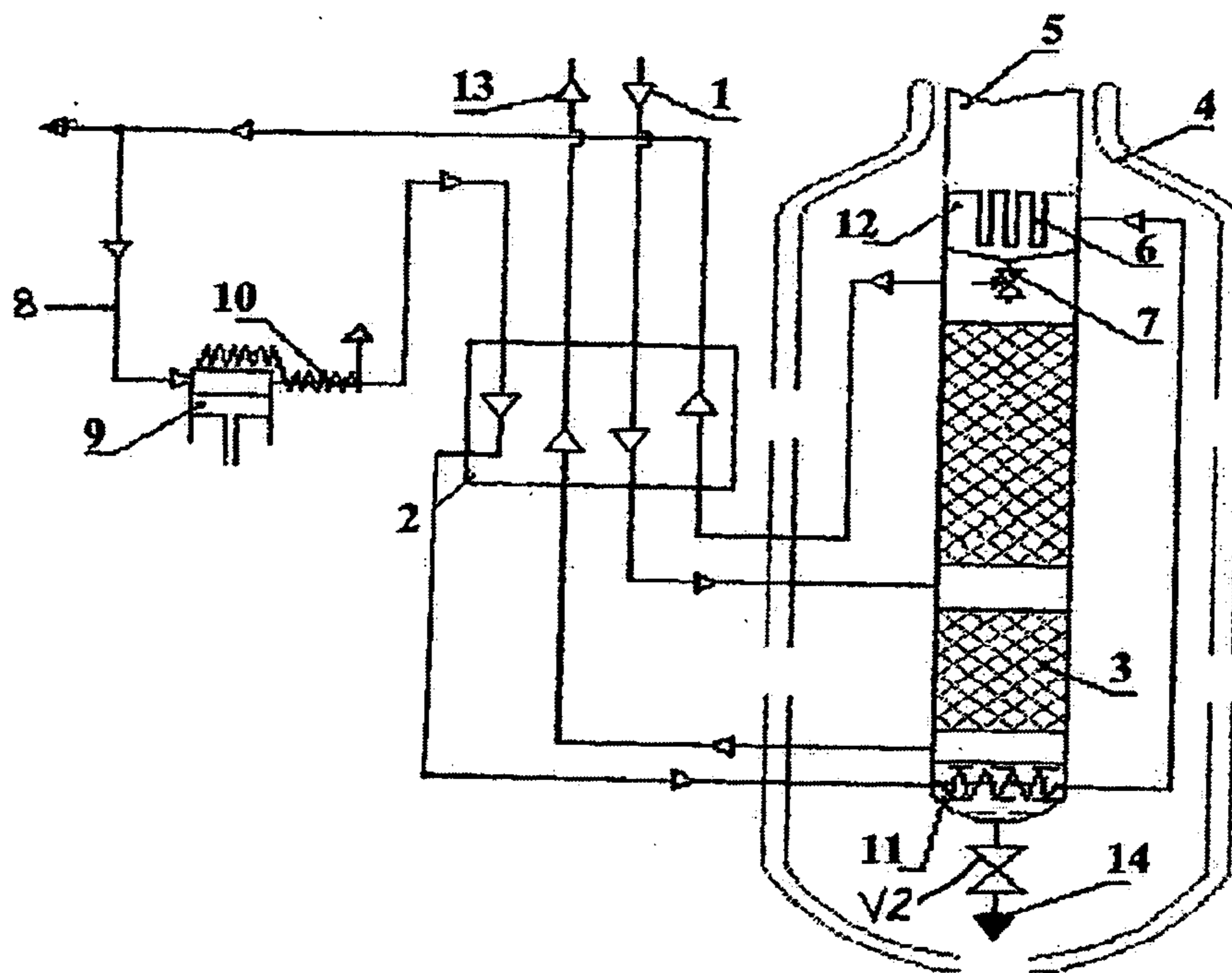


Fig. 1

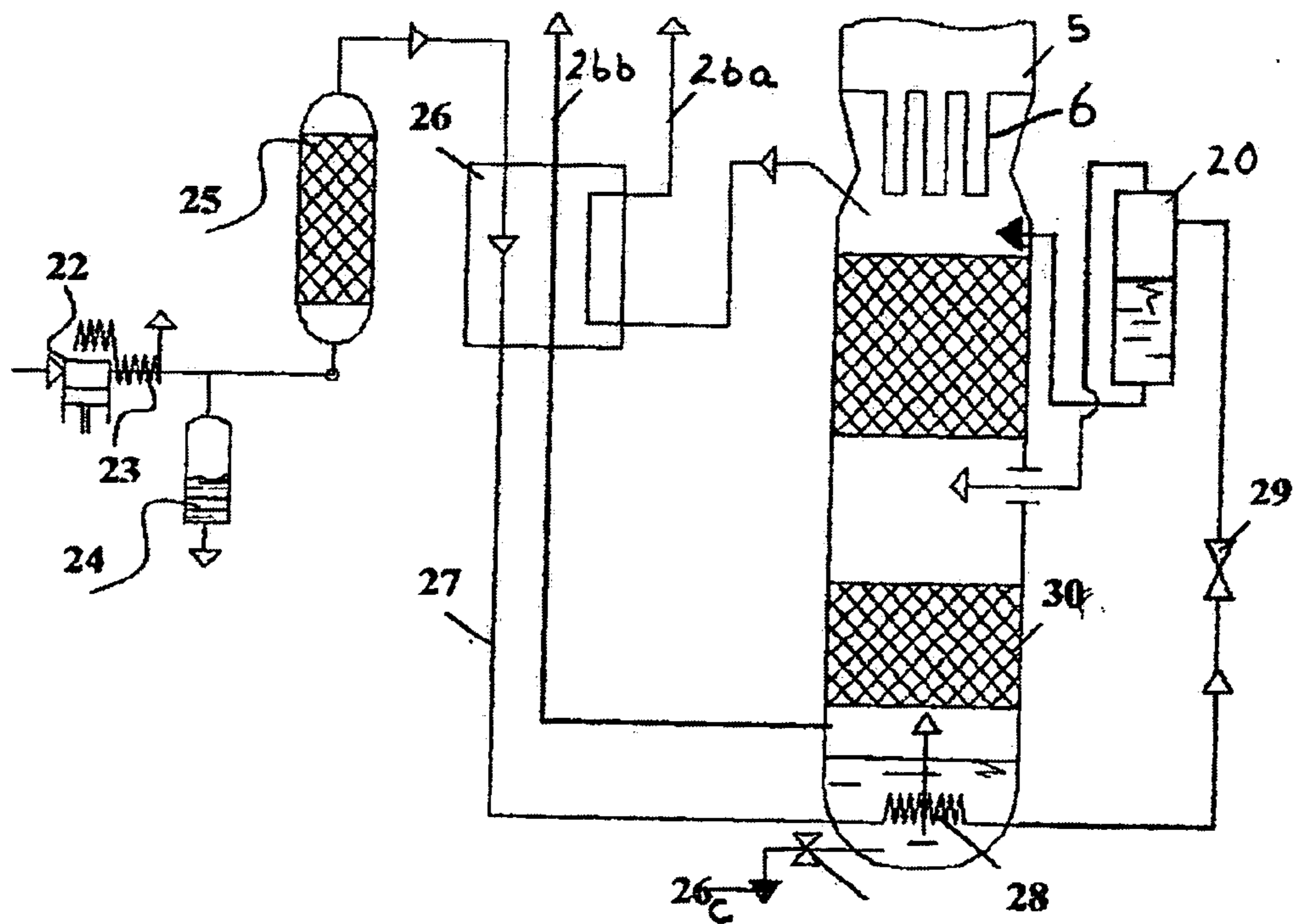


Fig. 2

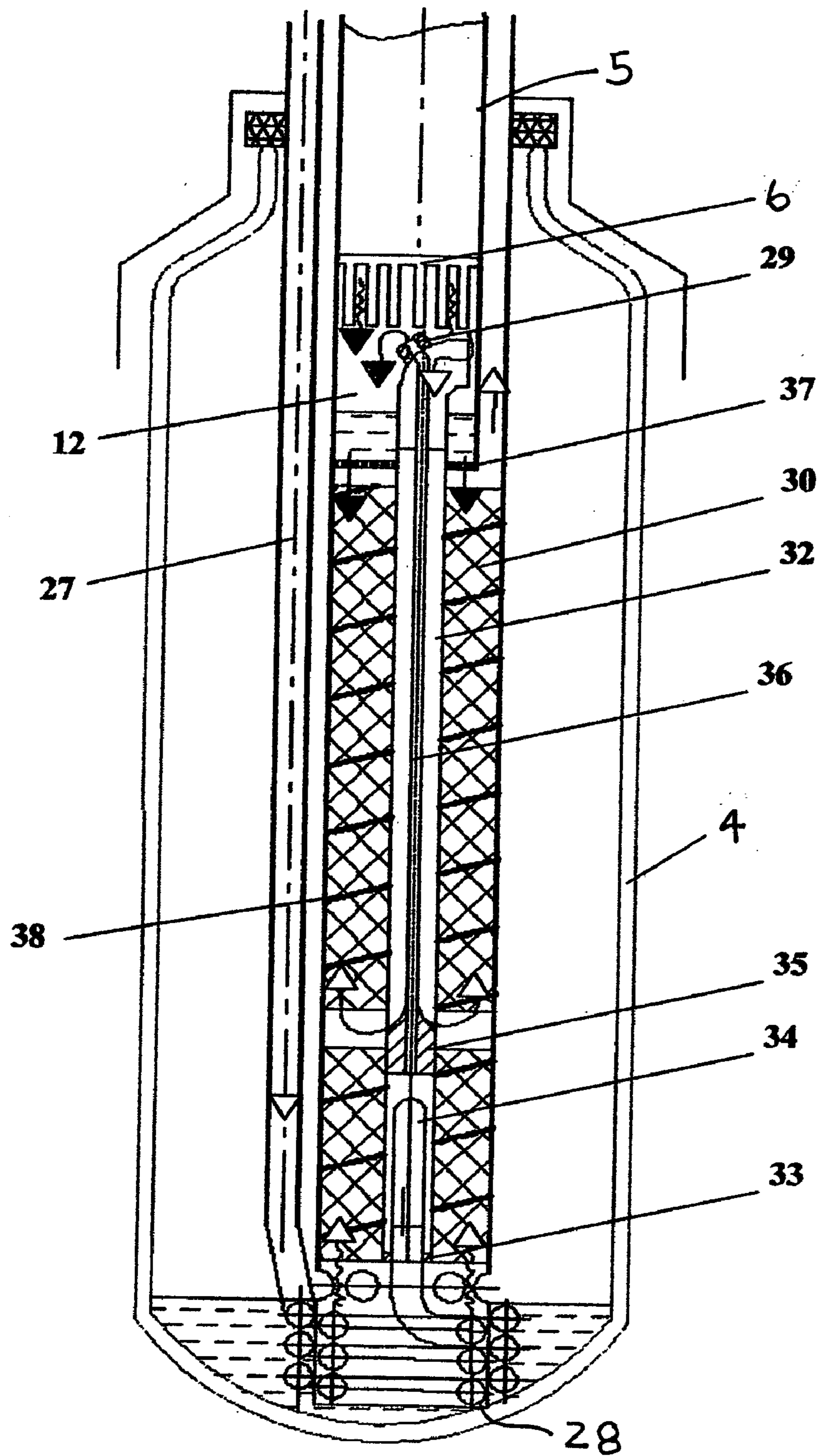


Fig. 3



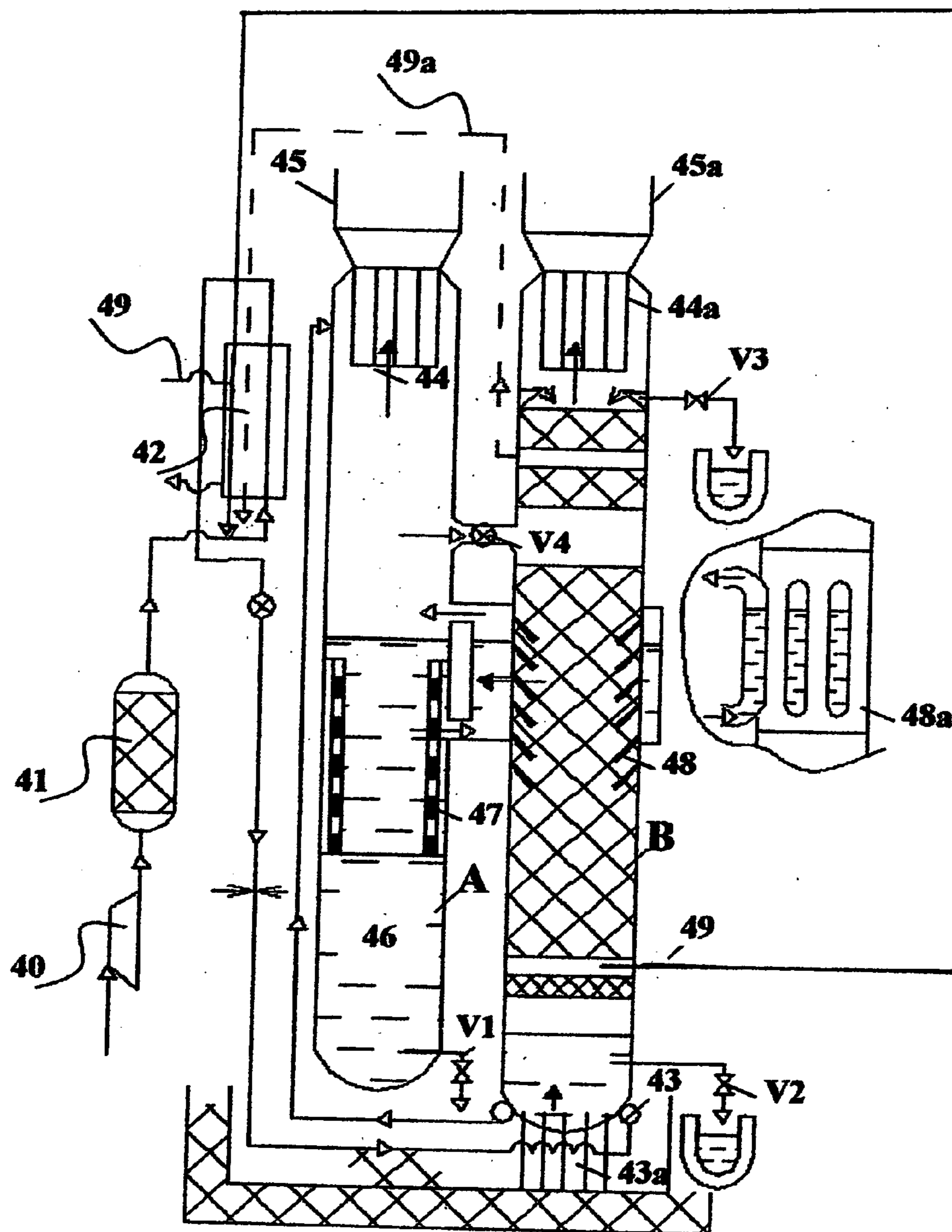
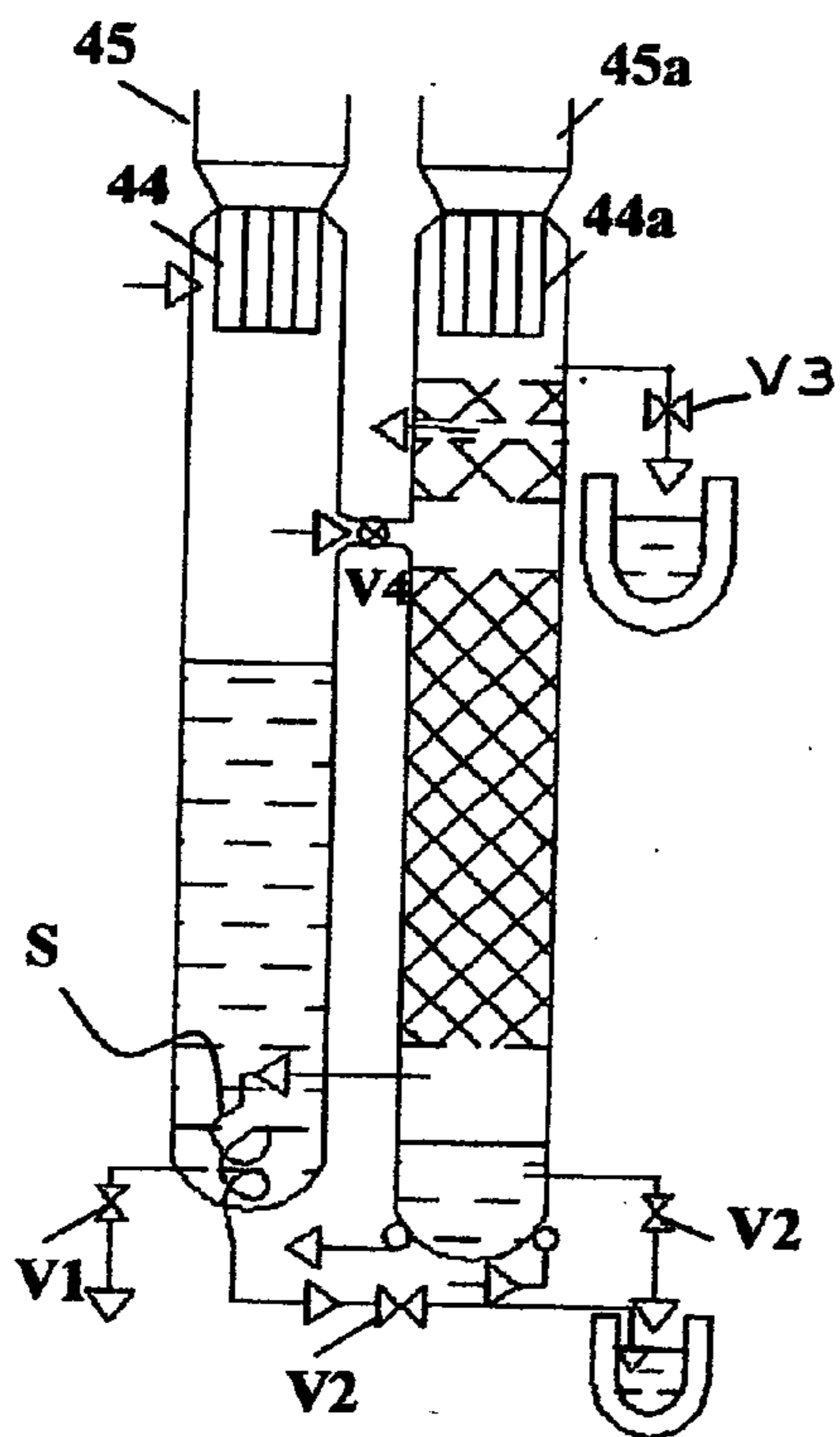
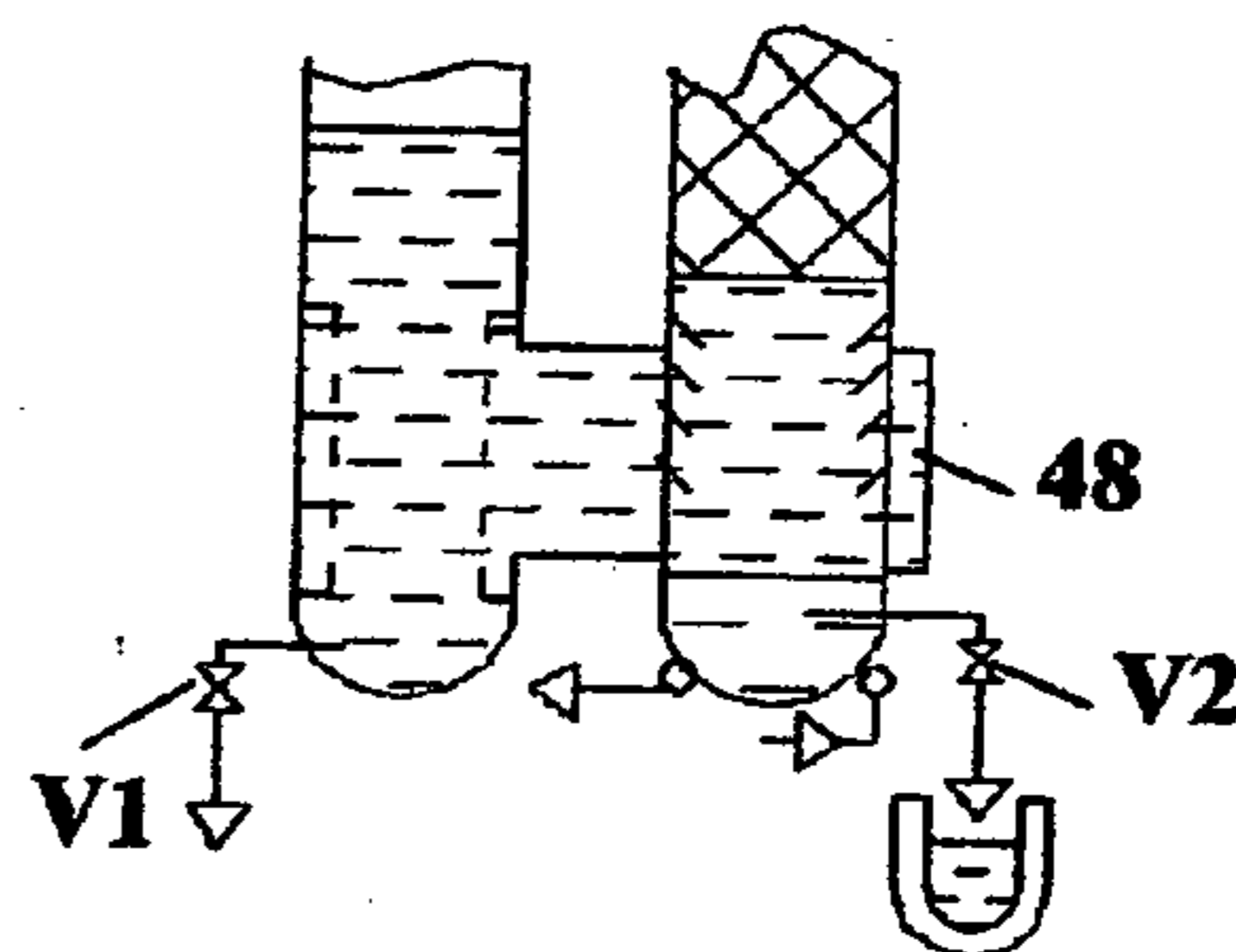


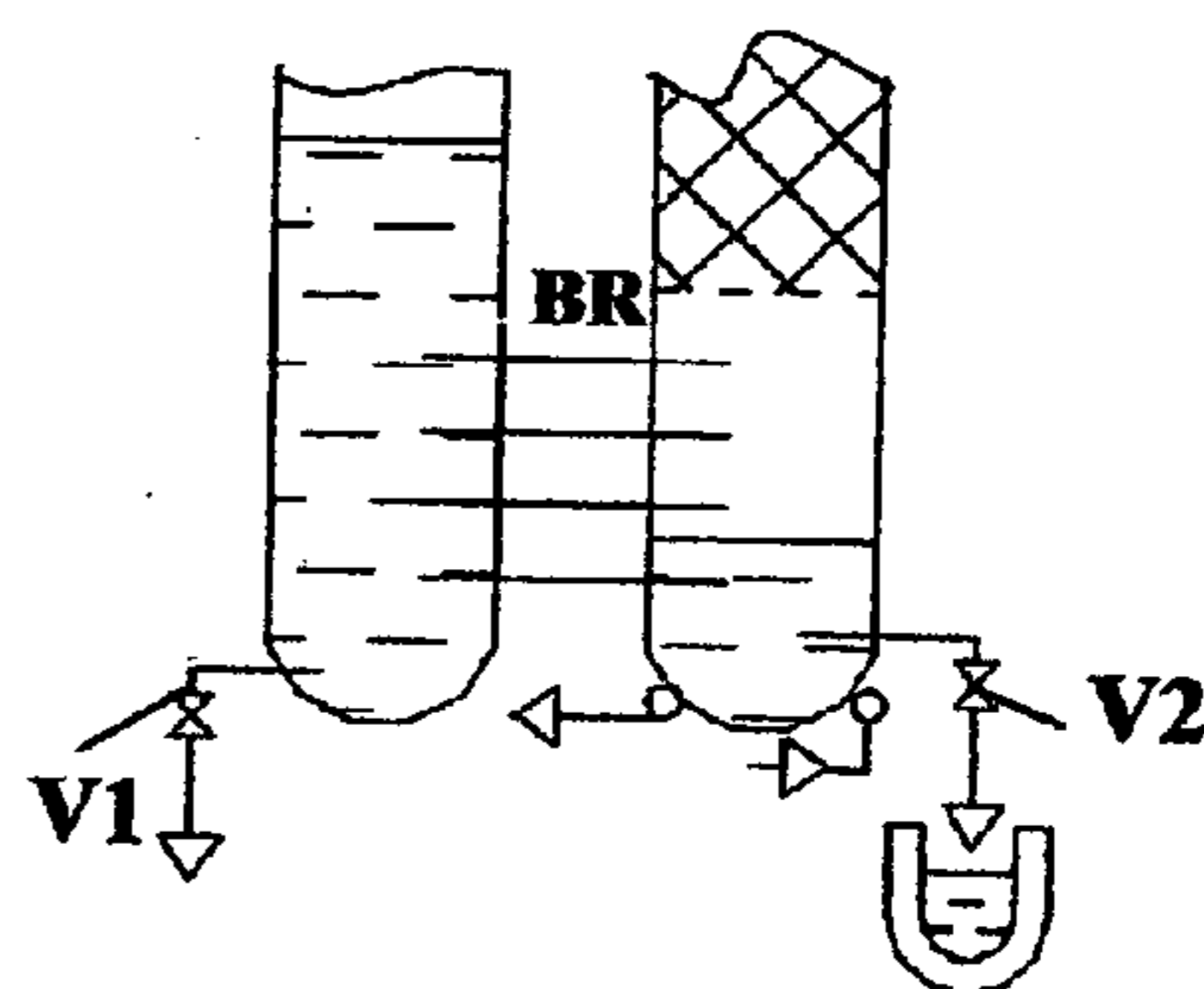
Fig. 4



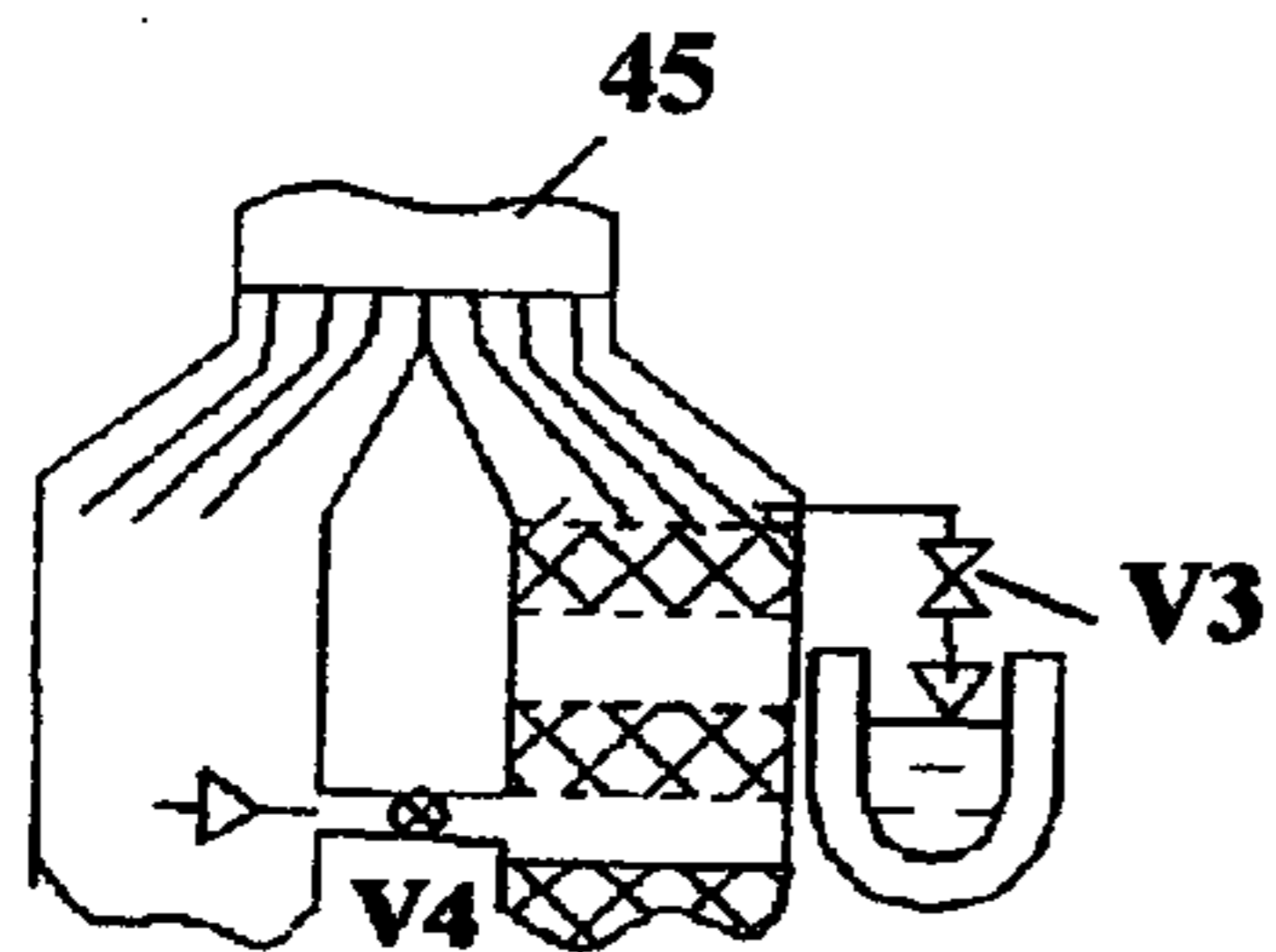
4a



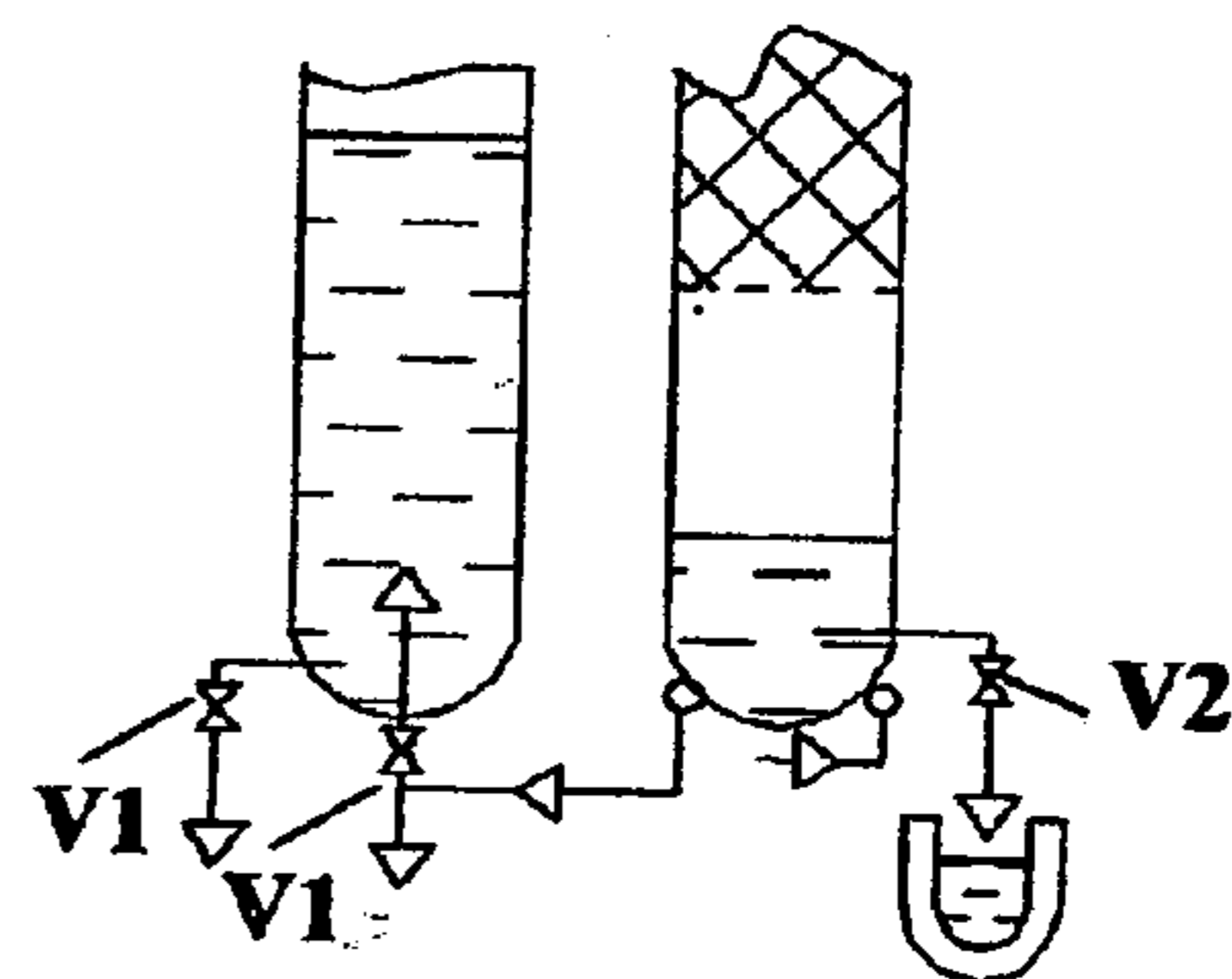
4c



4d



4b



4e

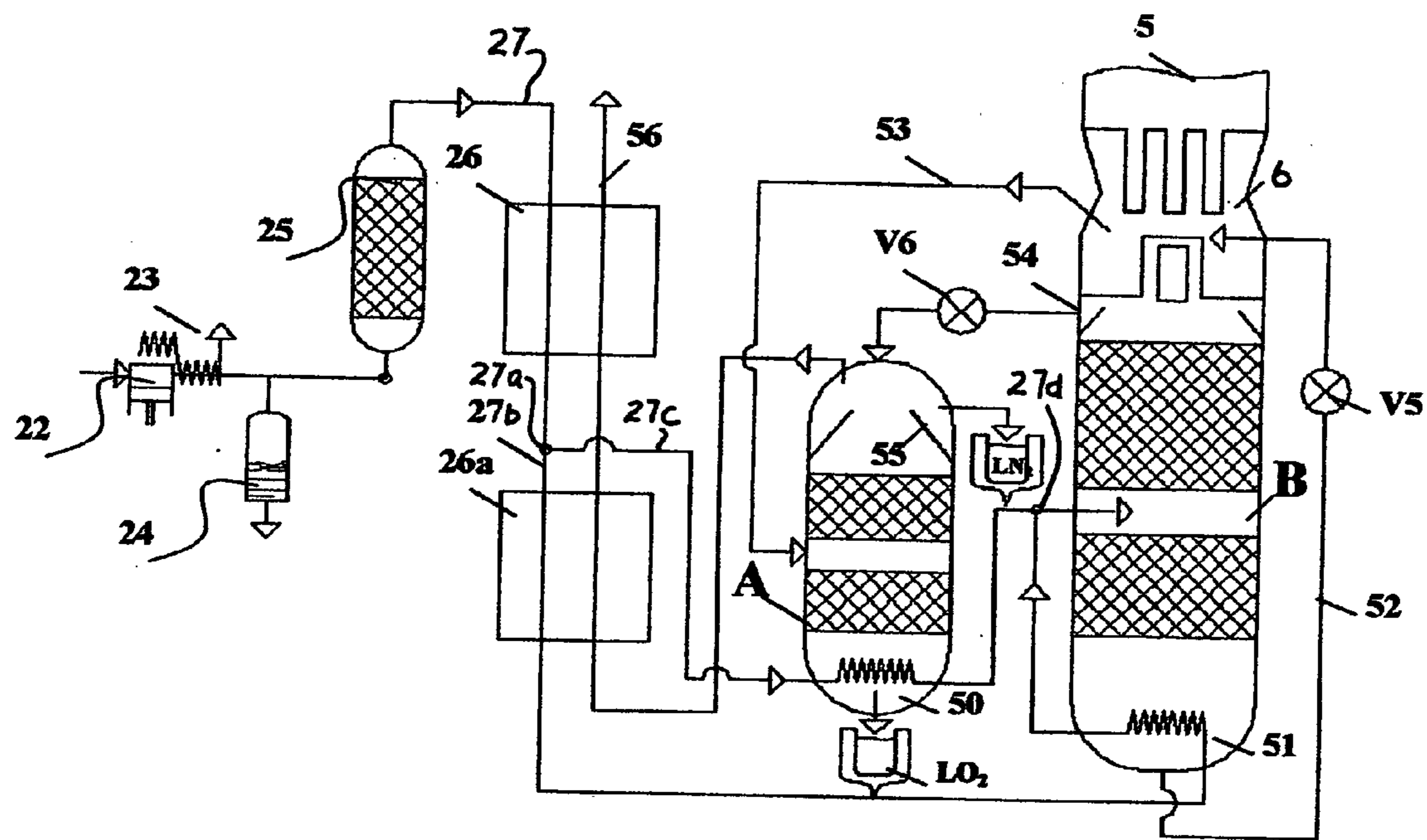


Fig.5

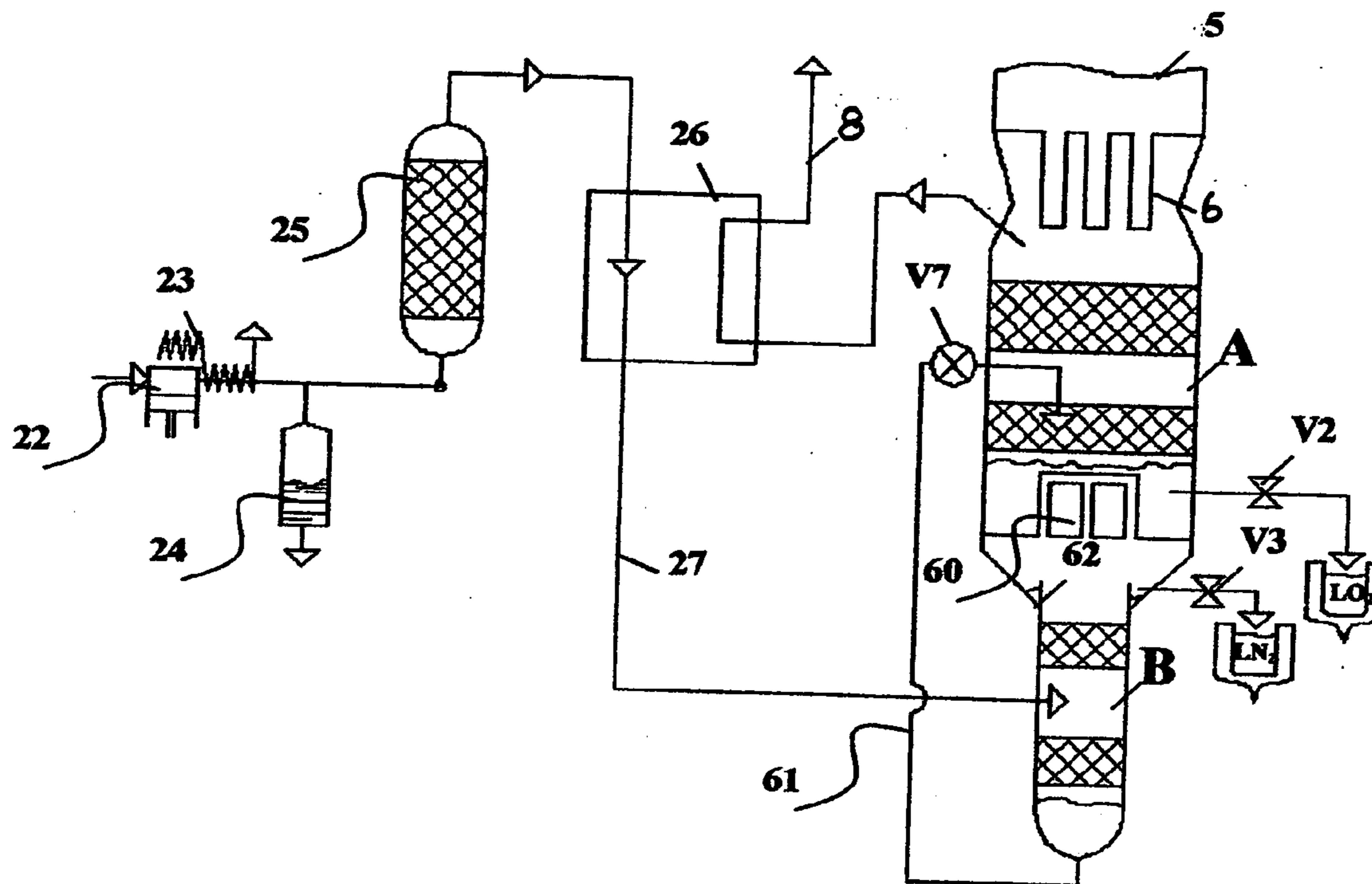


Fig.6

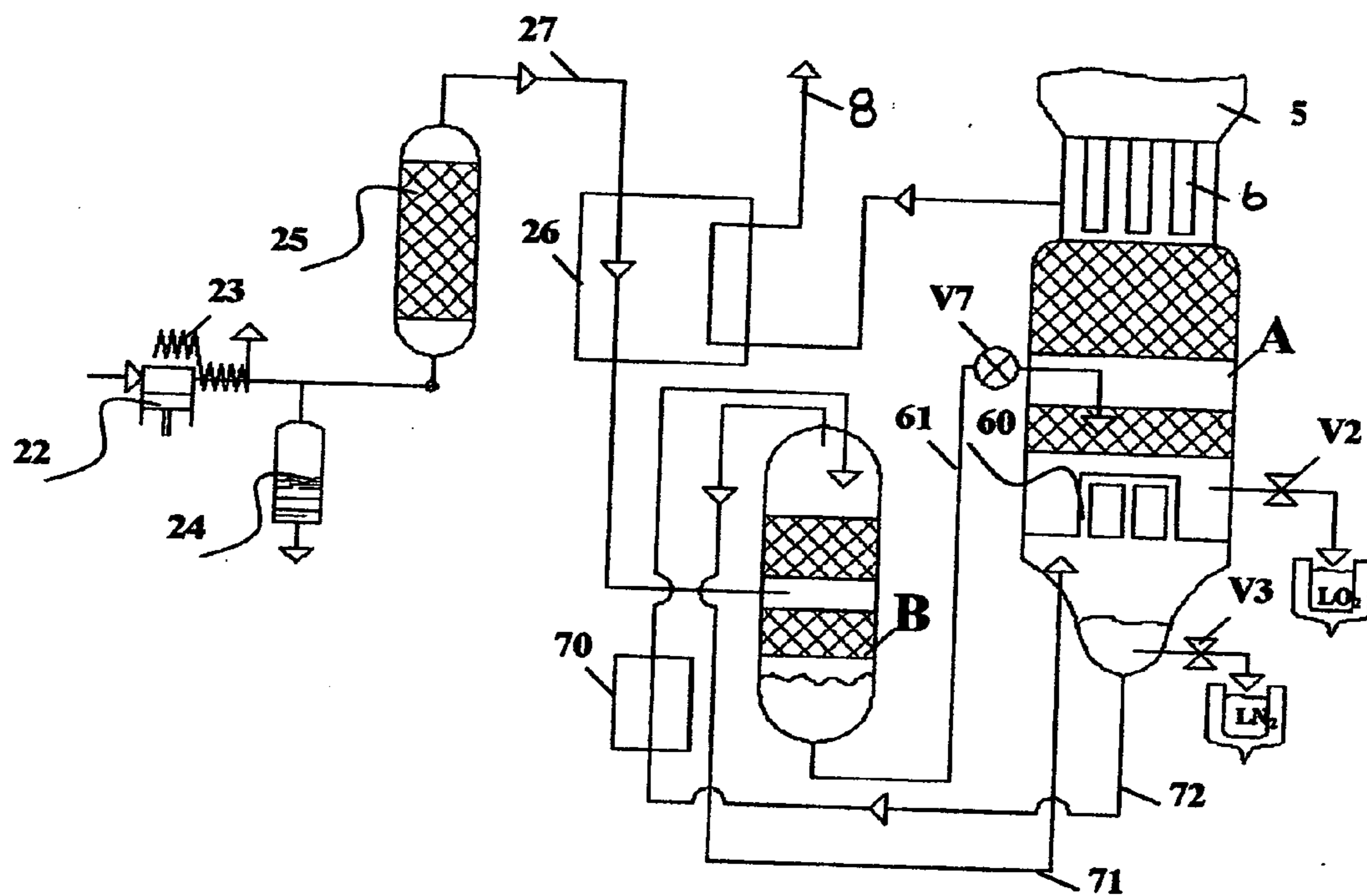


Fig.7



## GAS SEPARATION LIQUEFACTION MEANS AND PROCESSES

### CROSS-REFERENCE TO RELATED APPLICATIONS

[0001] Not Applicable

### STATEMENT REGARDING FEDERALLY SPONSORED RESEARCH OR

[0002] DEVELOPMENT

[0003] Not Applicable

### REFERENCE TO SEQUENCE LISTING, A TABLE OR A COMPUTER PROGRAM LISTING COMPACT DISK APPENDIX

[0004] Not Applicable

### BACKGROUND

[0005] The present invention relates generally to gas-separation/liquefaction and, more particularly, to a single and double column high-purity cryogenic gas-separation/liquefaction devices, where the refrigeration to the cryogenic gas-separation/liquefaction process is supplied by either a cryocooler alone or by a combination of a cryocooler and by a Joule-Thompson throttling process, and where the gas condensation may occur at least partially directly on the cold portion of the cryocooler which may be located inside of the thermally insulated space of the distillation column.

[0006] The background information discussed below is presented to better illustrate the novelty and usefulness of the present invention. This background information is not admitted prior art.

[0007] Cryogenic separation of gas mixtures is a well-established art. The processes used to separate the gaseous constituents of ambient air are well-known and understood. Although all of air's valuable components such as argon, neon, and xenon may be presently extracted from air in high-purity concentrations, the mainstay of the separation industry is the production of nitrogen and oxygen in various purities in gaseous or liquid form, as demanded by the particular application.

[0008] The first air separation plant for the commercial production of oxygen was designed and built by Dr. Carl von Linde in 1902. The plant had a single distillation column and refrigeration was obtained by throttling. Due to the plant's dependence on ineffective throttling and other inefficiencies, gaseous oxygen production required pressures of over 30 atmospheres, or higher. In the same year, Georges Claude improved on the Linde process by adding an expansion engine to the process. The expansion engine, however, proved to be an unreliable component. Therefore, in the late 1930's P. L. Kapitza proposed and developed expansion turbines for the separation of oxygen that proved to be far more reliable than the expansion engine. Moreover, the ability of the expansion turbines to handle large volumetric flow provided for cryogenic processing at much lower pressures, thus reducing the plant investment cost. Since that time, many variations and improvements have been made on these devices and their related processes.

[0009] A process for the synthesis of methane-oxygen mixtures gas has been described whereby the feeding natural gas and (preferably dry) compressed air into a distillation column at appropriate locations and at appropriate temperatures, produces nitrogen and heavier hydrocarbons as by-products. In this process, refrigeration is provided by multiple expansion machines and an expansion valve.

[0010] Soon after, a low temperature, single column, distillation process, where the refrigeration is provided by a reciprocating expansion machine and by a throttle valve both external to the distillation column, was described.

[0011] Gas-fractionating devices that provide the reflux for the distillation process and that have external refrigeration for condensation where the gas stream that provides the refrigeration and the gas stream to be separated are distinct, have been discussed, although, the design of such devices was not described.

[0012] A process of providing reflux in the distillation column in a reflux condenser that is refrigerated by a conventional Joule-Thompson throttling-work expanding, oxygen rich stream, external to the distillation column, was also taught.

[0013] Cryogenic separation of ethylene from a gaseous mixture at various temperature levels using refrigeration provided by an unspecified external refrigeration system was disclosed.

[0014] A process for the recovery of nitrogen from air within a single column, where refrigeration is provided by a turbo-expander, Joule-Thompson throttling, has been described.

[0015] A device providing for nitrogen rejection from a natural gas stream that utilizes a series of Joule-Thompson throttle valves to provide the necessary cooling, instead of external refrigeration, has also been introduced into the art. The use of a mixed refrigerant in a single loop refrigeration system providing for at least part of the heat duty of the reboiler is also known.

[0016] Most recently, a dephlegmator type separator where the refrigeration is also supplied by an external supply, was introduced.

[0017] It should be noted that even today the irreversible throttling process and the reversible, minus the losses, adiabatic expansion for cryogenic gas-separation/liquefaction, are still practiced almost exclusively. Moreover, it appears that for large tonnage capacity, cryogenic gas-separation/liquefaction plants will be using this technology for some time to come.

[0018] This is not the case, however, in the field of small-scale production of high-purity gases, such as therapeutic oxygen where the immense need for low-cost, small-scale production of high-purity breathing oxygen is currently generating interest in developing small-scale cryogenic-based gas-separation/liquefaction plants.

[0019] Until recently small-scale cryogenic-based gas-separation/liquefaction plants had to rely on periodic cryogenic liquid addition for their refrigeration needs. This type of refrigeration, however, is quite expensive. Lately, however, reliable cryocoolers of various designs capable of supplying refrigeration at, or below, the liquefaction tem-



perature of nitrogen have been made available. These cryo-coolers could be eminently suitable for small scale air separation as they eliminate the need for liquid nitrogen to be delivered to the gas-separation/liquefaction facility.

[0020] Applicant is not aware of any device or method wherein at least part of the refrigeration required to remove the heat of condensation from the distillation column reflux is achieved by a cryocooler wherein, in normal operation, a portion of the separated component(s) are condensed at least partially directly on the cold portion of the cryocooler.

#### SUMMARY

[0021] Accordingly, the present invention provides for means and processes that satisfy the hereto unmet need for small scale cryogenic air and/or other gas mixture separation where the needed refrigeration is provided wholly, or at least partially, by a cryocooler wherein during normal operation a portion of the enriched or separated component condenses at least partially directly onto the cold portion of the cryocooler.

[0022] Both single or double column high-purity cryogenic gas-separation/liquefaction devices are embodied within the principles of the invention where the refrigeration to the cryogenic gas-separation/liquefaction device is supplied by either a cryocooler alone or by a combination of a cryocooler and by a Joule-Thompson throttling process, and where the gas condensation may occur at least partially directly on the cold portion of the cryocooler which may be located inside of the thermally insulated space of the distillation column(s).

[0023] Using the embodiments described herein, gases, such as high-purity oxygen may be separated from, for example, ambient air in a device of the present invention, wherein that device is much smaller than presently available gas separation/liquefaction devices. Thus, these gas-separation/liquefaction systems made according to the principles of the present invention are particularly useful for medical applications, and especially for providing for safe and economical high-purity oxygen for at-home use.

[0024] The principles of the invention as taught herein include a combined column embodiment for the simultaneous production of high-purity liquid or gaseous oxygen and nitrogen. Another double column design offers a reduced temperature and pressure separation with an easy switch between oxygen and nitrogen extraction or single component extraction. If both gaseous and liquid oxygen are required, an oxygen purity of approximately 95% can be produced with good recovery i.e., with nitrogen purity of approximately 91%.

[0025] These advances in the art and the benefits they provide are accomplished by providing for a high-purity cryogenic gas-separation/liquefaction device for the production of liquid gases that comprises:

[0026] a) at least one means for supplying a feed gas;

[0027] b) at least one cryogenic means having a cold portion means for providing refrigeration for at least a process of condensing;

[0028] c) at least one condensation means for condensing at least one component of the feed gas, the condensation means thermally connected to the cold portion means;

[0029] d) at least one distillation means for providing for distillation of the condensed at least one component of the gas, and

[0030] e) at least one insulating means for thermally insulating the device,

[0031] wherein the refrigeration to the cryogenic gas-separation/liquefaction process may be provided by the at least one cryogenic means alone and where gas condensation may occur at least partially directly on the cold portion means of the at least one cryogenic means which may be located inside of the thermally insulated space of the at least one distillation means. It should be understood that in all contemplated applications the cold portion of the cryocooler may be equipped with extended surfaces for enhanced heat transfer.

[0032] It is further contemplated that the high-purity cryogenic gas-separation/liquefaction device further comprises:

[0033] a) wherein at least one cryogenic means is a cryocooler,

[0034] b) wherein at least one condensation means is a condenser, or the cold portion of the cryocooler.

[0035] c) wherein at least one distillation means is a distillation column, and

[0036] d) wherein at least one insulating means is a thermally insulated container,

[0037] wherein the refrigeration to the cryogenic gas-separation/liquefaction process may be provided by the at least one cryocooler alone and where gas condensation may occur at least partially directly on the cold portion of the at least one cryocooler which may be located inside or outside of the thermally insulated space of the at least one distillation column.

[0038] It is still further contemplated that the high-purity cryogenic gas-separation/liquefaction device further comprises:

[0039] a) wherein the cryogenic means is a cryocooler,

[0040] b) wherein the condensation means is a condenser, or the cold portion of the cryocooler.

[0041] c) wherein the distillation means is a distillation column, and

[0042] d) wherein the insulating means is a Dewar flask,

[0043] wherein the refrigeration to the cryogenic gas-separation/liquefaction process may be provided by the cryocooler alone and where gas condensation may occur directly on the cold portion of the cryocooler which may be located inside or outside of the Dewar flask of the distillation column.

[0044] Additionally it is contemplated that the high-purity cryogenic gas-separation/liquefaction device also may comprise wherein the feed gas, which may be ambient air or any other gas mixture of interest, may be driven into the device by a fan or compressor means, and further wherein water and carbon dioxide may be removed from the feed gas. The refrigeration to the cryogenic gas-separation/liquefaction device may be provided by a combination of the cryogenic means and a Joule-Thompson throttling process.



[0045] Moreover it is contemplated that the high-purity cryogenic gas-separation/liquefaction device, as recited above may further comprise wherein the feed gas passes through a multi-pass heat exchanger means for cooling and further where the cooled feed gas is introduced to the at least one distillation means at an appropriate composition point. It is also contemplated that an Interior volume of the at least one distillation means may be kept at an elevated pressure by a compressor.

[0046] Another contemplation comprises using the gas separation/liquefaction devices of this invention to achieve a high-purity cryogenic gas-separation/liquefaction process for the production of liquid gases, comprising the steps of:

[0047] a) supplying a feed gas;

[0048] b) providing at least one cryogenic means having a cold portion means for providing refrigeration for at least a process of condensing;

[0049] c) condensing at least one component of the feed gas using at least one condensation means thermally connected to the cold portion means; or directly on the cold portion means.

[0050] d) distilling the at least one component of the condensed gas using at least one distillation means, and

[0051] e) insulating the device using at least one thermally insulating means,

[0052] wherein providing the refrigeration to the cryogenic gas-separation/liquefaction process may be by the at least one cryogenic means alone and where gas condensation may occur at least partially directly on the cold portion means of the at least one cryogenic means which may be located inside or outside of the thermally insulated space of the at least one distillation means.

[0053] Furthermore, a high-purity double column cryogenic gas separation/liquefaction device for the simultaneous collection of a plurality of high-purity liquid gases is contemplated, wherein such a device comprise:

[0054] a) at least one means for supplying a feed gas;

[0055] b) a plurality of cryocoolers wherein each cryocooler has a cold portion to provide refrigeration,

[0056] c) a plurality of condensing means wherein each condenser means is a standard condenser thermally related to the cold portion of a cryocooler, or where the condensing means is a cold finger of the cryocooler.

[0057] d) a plurality of distillation columns, and

[0058] at least one insulating means for insulating the device,

[0059] wherein refrigeration to the cryogenic gas-separation/liquefaction process may be provided by the cryocoolers alone and where gas condensation may occur at least partially directly on the cold portions of the cryocoolers which may be located inside or outside of the thermally insulated space of the distillation column.

[0060] Still other benefits and advantages of this invention will become apparent to those skilled in the art upon reading and understanding the following detailed specification and related drawings.

#### BRIEF DESCRIPTION OF THE DRAWINGS

[0061] In order that these and other objects, features, and advantages of the present invention may be more fully comprehended and appreciated, the invention will now be described, by way of example, with reference to specific embodiments thereof which are illustrated in appended drawings wherein like reference characters indicate like parts throughout the several figures. The invention will be described and explained with additional specificity and detail using the accompanying drawings, in which:

[0062] **FIG. 1** is a schematic of a first embodiment of the present invention.

[0063] **FIG. 2** is a schematic of a second embodiment of the present invention.

[0064] **FIG. 3** is a plan view of a distillation column functionally situated inside of a cryogenic Dewar flask.

[0065] **FIG. 4** is a schematic of another embodiment of a gas separation device, made according to the principles of the invention described herein, where two cryocoolers are used for cooling and condensation.

[0066] **FIG. 4a** is a schematic of a coil evaporator that may be used as an alternative to a conventional condenser.

[0067] **FIG. 4b** is a schematic of an optional design for a combined cooling source to reduce total energy consumption.

[0068] **FIG. 4c** is a schematic of a more effective heat exchange where the condenser-evaporator is placed at the bottom section of column B.

[0069] **FIG. 4d** is a schematic of an alternative heat bridge that may be used in place of condenser 48.

[0070] **FIG. 4e** is a schematic of the optional use of direct blow-through flow, in which case the condenser could be eliminated.

[0071] **FIG. 5** is a schematic, in plan view, of double column design for the simultaneous production of high-purity oxygen and nitrogen according to the principle of the present invention.

[0072] **FIG. 6** is a schematic, in plan view, of a variation in design of a double column gas separator device according to the present invention.

[0073] **FIG. 7** is a schematic, in plan view, of another variation in design of a double column gas separator device according to the present invention.

#### REFERENCE NUMERALS AND THE PARTS OF THE INVENTION TO WHICH THEY REFER

[0074] **1** Conduit through which air may be driven into a gas separation device of this invention by a fan or compressor means located in an operative position.

[0075] **2** Multi-pass heat exchanger having a warm end (the top part of exchanger, as illustrated) and a cold end (the bottom part of the exchanger, as illustrated).

[0076] **3** Distillation column.

[0077] **4** Insulation about distillation column **3**.

[0078] **5** Cryocooler (see **FIGS. 1, 5, and 7**).



- [0079] **6** Condensing means as exemplified is the cold finger of cryocooler **5**.
- [0080] **7** Expansion valve providing for Joule-Thompson expansion.
- [0081] **8** Conduit for nitrogen-rich stream.
- [0082] **9** Compressor keeping interior volume **12** of distillation column **3** at appropriate pressure.
- [0083] **10** Heat exchanger for removing heat of compression.
- [0084] **11** Boiler.
- [0085] **12** Elevated pressure volume of distillation column **3**.
- [0086] **13** Exit point of the gaseous product.
- [0087] **14** Exit point of liquid product.
- [0088] **20** Phase separator.
- [0089] **22** Indicates, in **FIG. 2**, where air is driven into a gas separation device of this invention (equivalent to **9**).
- [0090] **23** Heat exchanger for the removal of the heat of compression.
- [0091] **24** Humidity removal device.
- [0092] **25** Dew point reduction and removal of carbon dioxide device.
- [0093] **26** Multi-pass heat exchanger having a warm end (the top part of exchanger, as illustrated) and a cold end (the bottom part of the exchanger, as illustrated).
- [0094] **26a** Exit pathway of gaseous nitrogen enriched stream.
- [0095] **26b** Exit pathway of the gaseous oxygen from vapor space above reboiler **28**.
- [0096] **26c** Exit pathway of liquid oxygen that passed through valve **V2** on its way from the liquid pool of reboiler **28**.
- [0097] **27** Compressed air conduit through which gas passes to reboiler (same function as seen in **FIGS. 2, 3, and 5**).
- [0098] **27a** Point where gas stream is separated into two parts.
- [0099] **27b** Conduit through which one part of stream is fed to heat exchanger **26a**.
- [0100] **27c** Conduit through which one part of stream is fed to boiler **50**.
- [0101] **27d** Point where the separated gas streams are rejoined.
- [0102] **28** Reboiler to vaporize liquid oxygen for operating distillation column **30**.
- [0103] **29** Expansion valve providing for Joule-Thompson expansion (same function in **FIGS. 2 and 3**).
- [0104] **30** Distillation column.
- [0105] **32** Tube located inside distillation column **30**.
- [0106] **33** Annular fitting bonded to both tube **32** and to conduit **27**.
- [0107] **34** Filter through which compressed air enters the lower section of the tube **32**.
- [0108] **35** Upper annular fitting.
- [0109] **36** Center tube.
- [0110] **37** Liquid distributor.
- [0111] **38** Spiral guide of packing section.
- [0112] **40** Indicates, in **FIG. 4**, compressor to drive air into a gas separation device of this invention.
- [0113] **41** Dew point reduction and removal of carbon dioxide device.
- [0114] **42** Multi-pass heat exchanger having a warm end (the bottom part of exchanger, as illustrated in **FIG. 4**) and a cold end (the top part of the exchanger, as illustrated in **FIG. 4**).
- [0115] **43** Boiler for vaporizing liquid oxygen.
- [0116] **44** Condensing means as exemplified is the cold finger of cryocooler **45**.
- [0117] **44a** Condensing means as exemplified is the cold finger of cryocooler **45a**.
- [0118] **45** First cryocooler.
- [0119] **45a** Second cryocooler.
- [0120] **46** Collector section of column A.
- [0121] **47** Filter for liquid air.
- [0122] **48** Condenser/evaporator.
- [0123] **48a** Detail of condenser/evaporator.
- [0124] **49** Conduit for enriched oxygen.
- [0125] **49A** Conduit for enriched nitrogen.
- [0126] **50** Boiler of distillation column A.
- [0127] **51** Boiler of distillation column B.
- [0128] **52** Conduit through which liquid from boiler **51** travels.
- [0129] **53** Conduit through which non-condensed vapor travels.
- [0130] **54** Collectors of column B.
- [0131] **55** Collectors from which high-purity liquid nitrogen may be removed.
- [0132] **56** Conduit through which highly enriched nitrogen vapor travels.
- [0133] **60** Condenser.
- [0134] **61** Liquid conduit.
- [0135] **62** Collector.
- [0136] **70** Liquid pump.
- [0137] **71** First conduit line providing flow connection between column A and column B.
- [0138] **72** Second conduit line providing flow connection between column A and column B.
- [0139] A First column.
- [0140] B Second column.



- [0141] BR Heat bridge.
- [0142] V1 Valve to periodically drain CO<sub>2</sub> impurities.
- [0143] V2 Valve for collection of oxygen gas containing most of the argon gas component of feed air.
- [0144] V3 Valve for collection of nitrogen liquid.
- [0145] V4 Valve.
- [0146] V5 Throttling valve.
- [0147] V6 Valve shown in **FIG. 5** through which enriched liquid oxygen flows from collectors **54** of column B to enter the top of column A.
- [0148] V7 Throttling valve.
- [0149] S Coil evaporator used in place of condenser/evaporator **48**.
- [0150] Cross-hatching denotes separation devices, such as trays, packing, etc.

#### Definitions

- [0151] Cold finger, as used herein, refers to a finger-shaped cooled and cooling protruding member. Where the cryocooler has adequate cooling capacity, condensation can be carried out on the cold portion of the cryocooler itself, without employing a separate condenser.
- [0152] Condensation, as used herein, refers to the conversion of a substance from its vapor or gaseous state to its liquid or solid state usually initiated by a reduction in temperature of the vapor.
- [0153] Condenser, as used herein, refers to that part of a distillation apparatus that cools vapor until it becomes a liquid. There are several types of condensers. One is simply an inner tube that is cooled by an outer jacket filled with a liquid like water, for example. The other type has the inner tube filled with small (usually glass) beads or other shaped small bits of material. The distillate is taken from the top. This type of condenser in effect does thousands of tiny condensations (they occur on each bead) and produces a much more pure product. To achieve a similar effect with the other type, you need to do multiple distillations. Where the cryocooler has adequate cooling capacity, condensation can be carried out on the cold portion of the cryocooler itself, without employing a separate condenser.
- [0154] Cryocooler, as used herein, refers to any device that can produce cryogenic temperatures with significant capacity for useful application. The term cryocooler may denote any of the following: Gifford-McMahon cryocoolers and any related variations, Stirling cryocoolers of the crank or linear motor driven variety, the many variations of the Pulse Tube cryocoolers and combinations of these with the Stirling refrigerators, reverse Brayton Cycle cryocoolers, Multi component Vapor Compression cryocoolers, and the like.
- [0155] Note that providing reflux in the distillation column by condensing a portion of the at least partially separated gas(s) on the cold part of the cryocooler would be a common characteristic for any cryocooler.

- [0156] Cryogenics, as used herein, refers to the science concerned with low-temperature phenomena. Temperatures less than -40 degrees Celsius are usually classified as cryogenic.
- [0157] Dephlegmator, as used herein, refers to a part of a distilling apparatus in which the separation of the vapors (gases) is effected.
- [0158] Dewar or Dewar flask, as used herein, refers to a glass or metal container made like a vacuum bottle that is used especially for storing liquefied gases.
- [0159] Distillation, as used herein, refers to the process of purifying, or separating the components of, a mixture by successive evaporation and condensation.
- [0160] Joule-Thompson expansion, as used herein, refers to the cooling that gas undergoes as it expands.
- [0161] It should be understood that the drawings are not necessarily to scale. In certain instances, details which are not necessary for an understanding of the present invention or which render other details difficult to perceive may have been omitted.

#### DETAILED DESCRIPTION

- [0162] Referring now, with more particularity, to the drawings, it should be noted that the disclosed invention is disposed to embodiments in various sizes, shapes, and forms. Therefore, the embodiments described herein are provided with the understanding that the present disclosure is intended as illustrative and is not intended to limit the invention to the embodiments described herein. **FIG. 1** schematically illustrates one device design and a related process of the present invention.
- [0163] In the schematic shown in **FIG. 1**, ambient air, or any other gas mixture of interest, from which water and carbon dioxide have been removed, is driven into conduit **1**, by a fan or compressor operatively located proximal to the entrance of conduit **1**. Conduit **1** connects to the warm end of multi-pass heat exchanger **2** (which warm end is located at the top of heat exchanger **2**, as illustrated). Low temperature air exits from the cold end of multi-pass heat exchanger **2** (which cold end is located at the bottom portion of heat exchanger **2**, as illustrated) and is introduced to distillation column **3** at the appropriate composition point.
- [0164] Distillation column **3** is insulated by insulator **4**. Cryocooler **5** is operatively installed about the top of distillation column **3**, as shown. The cold portion of cryocooler **5** is thermally connected to cold finger **6**. In the embodiment illustrated, the temperature of cold finger **6** is between about 77 K and about 88 K (roughly between the boiling point of liquid nitrogen, about 77 K, and the boiling point of liquid oxygen, about 90 K), or for other gases of interest, between the boiling points of the low and high boiling components of the other gas mixtures. The thermal design geometry and the insulation at the insertion of cryocooler **5** into column **3** should be carefully optimized to minimize heat input from the ambient.
- [0165] Interior volume **12** of distillation column **3** is kept at an elevated pressure by compressor **9** which is fed a nitrogen-enriched stream as its working fluid. The path of travel of the nitrogen-enriched stream is indicated by reference numeral **8**. The heat of compression is removed from



stream **8** by heat exchanger **10** before stream **8** enters the warm end of heat exchanger **2** where it is cooled to an intermediate temperature between the warm and cold end of the exchanger, as required by balancing (not shown). Stream **8** then enters boiler **11** to produce the oxygen-rich vapor that is used to operate distillation column **3**. Exiting boiler **11**, stream **8** is introduced into volume **12** of distillation column **3** where stream **8** condenses at least partially on cold finger surface **6** of cryocooler **5**. The condensate then undergoes a Joule-Thompson expansion in valve **7** and the liquid reflux is distributed to the top of the distillation column. The nitrogen-enriched stream exits distillation column **3** from the space between the low-pressure end of the Joule-Thompson expansion valve **7** and the top of the packing or trays of the distillation column (denoted by hatching) via conduit **8** and enters the cold end of the heat exchanger **2**. Heat exchanger **2** acts as a counterflow heat exchanger to cool the incoming feed gas against the outgoing waste and/or product gas; this heat exchanger may be of the regenerator type. Alternatively, high-pressure vapor space **12** at the top end of the distillation column may be connected to the cold end of conduit **8**, through which the nitrogen-enriched stream passes via an appropriately-sized capillary.

[0166] Oxygen-rich, gaseous product leaves the vapor space of reboiler **11**, enters the cold end of heat exchanger **2** at an appropriate temperature point, and is discharged at room temperature through exit **13**. Liquid oxygen product is discharged from boiler **11** at exit **14** through V2.

[0167] Although not shown, it will be readily appreciated by those skilled in the art, all of the cold conduits and the low temperature points of the heat exchanger **2** are kept well-insulated. The cold parts of heat exchanger **2** also may be positioned inside the Dewar flask that may also contain distillation column **3**. Conversely, the distillation column may be located inside of one Dewar while the heat exchanger and the cold conduits are kept in another Dewar.

[0168] In works by R. A. Gaggioli et al., K. D. Timmerhaus et al., and A. M. Arkharov et al. one, who is well-versed in the art, will find the fundamental physics and physical chemistry required for constructing one of the above described novel cryogenic separation systems according to the principles taught herein and will also find the procedures necessary for balancing the system around a given component, such as a cryocooler.

[0169] Another contemplated embodiment for the cryogenic separation of air using a cryocooler is illustrated schematically in FIG. 2. Ambient air, or any other gas mixture of interest, is compressed to a relatively low-pressure, typically, but not necessarily, less than around 0.8 MPa, by compressor **22**. The heat of compression is removed in heat exchanger **23** and the condensed humidity is removed in humidity exchanger **24**. The cooled and dried compressed air then enters dew point reduction device **25** (suggested types of available dew point reduction devices are given below) to reduce the dew point of the cooled and dried compressed air to a desired low level and to remove CO<sub>2</sub> to prevent plugging in the low temperature parts of the system. The humidity and carbon dioxide removal can be effected by any known, or yet to be known, device, such as semi-permeable membranes, adsorption devices, or by any combination of the known methods. After leaving device **25**, the compressed, clean air enters the warm end of heat

exchanger **26** where it will be cooled down against the cold outgoing product and waste gases. The cooled air is then introduced through conduit **27** to reboiler **28** where liquid oxygen is vaporized and collected in the bottom of the distillation column to enable proper functioning of the same. Partially condensed in the reboiler **28**, the compressed air undergoes a Joule-Thompson expansion in valve **29** where the temperature will be reduced further causing the liquid mass fraction to increase. The liquid and gas phases will be separated in phase separator **20**, and introduced into the distillation column **30** at the appropriate composition points. Cold finer **6** of cryocooler **5** will provide part of the liquid required for the operation of distillation column **30**. The gaseous nitrogen enriched stream is withdrawn from the gas space at the top of the column along pathway **26a**, gaseous enriched oxygen is withdrawn from the vapor space above reboiler **28** along pathway **26b**, and liquid oxygen from the liquid pool of the reboiler, after passing through trough valve V2, is withdrawn through pathway **26c**. The separated gases (enriched oxygen and enriched nitrogen) will be warmed up concurrently against incoming air in heat exchanger **26**. If both gaseous and liquid oxygen are required, an oxygen purity of approximately 95% can be produced with good recovery i.e., with nitrogen purity of approximately 91%.

[0170] FIG. 3 illustrates a distillation column functionally positioned inside of insulating cryogenic Dewar flask **4**, which flask may be made of metal or glass. Compressed air enters the device via conduit **27** (having the same function as the conduit **27** in FIG. 2) at the top of the flask, which conduit is in close proximity to the length of distillation column **30**. The compressed air travels down conduit **27** forming a spiral in the liquid pool affecting reboiler **28**. Distillation column **30** is constructed in an annular fashion with tube **32** located inside distillation column **30**. As illustrated in FIG. 3, the packing (denoted by hatching) is divided into two portions. Compressed air from conduit **27** enters tube **32** near the bottom of the distillation column. Annular fitting **33** is bonded to both tube **32** and to conduit **27** so that compressed air enters the lower section of the tube **32** first through filter **34**, which is optional, then through upper annular fitting **35** to finally enter into center tube **36**. Tube **36** ends in capillary fitting **29** (same function as mentioned in discussion relating to FIG. 2) and will discharge a mixture of gaseous vapor and liquid into elevated pressure volume **12** of the distillation column. The liquid phase will join the condensate obtained on cold finger **6** of cryocooler **5** and will be distributed through sieve **37** to provide the reflux. The gaseous phase will be returned downward in the annulus formed by tube **32** and the center tube **36** to an appropriate concentration location of the distillation column packing or trays. Drillings or slots provided in tube **32** (not shown) will let this gas portion join the gas phase of the distillation column at the appropriate concentration height.

[0171] Compactness of the unit is achieved by the optional use of spiral guide **38** of packing sections in the distillation columns. In such a design the vapor goes upward along a spiral path as the reflux is distributed downward through each loop by gravity and capillary forces.

[0172] Yet another embodiment, as illustrated in FIG. 4, offers a double column design for the simultaneous production of high-purity oxygen and nitrogen, where enhanced performance of the system is achieved using two cryocool-



ers for cooling and condensation. Here, feed air is compressed by 40. Water vapor and carbon dioxide are then removed from the compressed air in device 41. The cleaned and compressed air then travels through multi-pass heat exchanger 42, which exchanger has a warm end (the bottom part of exchanger, as illustrated in FIG. 4) and a cold end (the top part of the exchanger, as illustrated in FIG. 4) (these parts are similar to parts 22, 25, and 26 of FIG. 2, respectively). The air is then conveyed to boiler 43 where the liquid oxygen is vaporized in the bottom section of column B. The partially condensed stream is fed to cold finger 44 at the upper section of column A where additional condensate will be formed. Cold finger 44 is part of the cold end of first cryocooler 45. Alternatively, condensation may occur at least partially directly on the cold end of cryocooler 45. The condensed air with the remaining CO<sub>2</sub> impurities will drip down into the collector section 46 of column A wherefrom it may be periodically drained through valve V1. The liquid air, after passing through filter 47 will be introduced to the condenser-evaporator 48 (which may be of the common type, as illustrated in as 48A) of column B where it is partially vaporized. The gaseous phase is fed to the appropriate concentration point of the upper section of column B through V4 and separated in a gas-liquid contacting device due to rectification with the downstream liquid. The liquid stream/ vapor balance in column B is maintained by cold finger 44a which is cold part of second cryocooler 45a. Alternatively, condensation may occur at least partially directly on the cold end of cryocooler. Separated oxygen gas containing most of the argon gas component of the feed air and nitrogen liquid may leave the column through valves V2 and V3, respectively. Enriched oxygen and nitrogen gas flows through conduits 49 and 49A respectively and will be utilized in heat exchanger 42 to pre-cool the incoming air. This design allows high-purity co-production of oxygen in gaseous and liquid form and of nitrogen.

[0173] FIGS. 4a, 4b, 4c, 4d, and 4e show alternate designs for selected structural parts of the invention as described in connection with FIG. 4. FIG. 4a illustrates coil evaporator S used in place of condenser/evaporator 48 (as illustrated in FIG. 4). FIG. 4b illustrates the use of a combined cooling source which will reduce the total energy consumption. More effective heat exchange could be achieved using the alternative design illustrated in FIG. 4c where the condenser-evaporator is placed at the bottom section of column B. Condenser 48 (as illustrated in FIG. 4) may be replaced by heat bridge BR as depicted in FIG. 4d or completely eliminated from the device if direct blow-through flow is utilized per FIG. 4e. The schemes presented in 4d and 4e may necessitate utilization of a heat bridge 43a.

[0174] FIG. 5 illustrates a double column design for the simultaneous production of high-purity oxygen and nitrogen, using any cooling device wherein the cold portion of the cooling device is installed directly into the distillation column. H<sub>2</sub>O and CO<sub>2</sub> are first removed from compressed air as described above. The cleaned and compressed air then enters the system through conduit 27 and is then sent through a two part heat exchanger, having sections 26 and 26a. At point 27a between heat exchanger sections 26 and 26a, the air stream is split into two streams. One stream is fed through conduit 27c to boiler 50 of distillation column A where it is partially condensed. The second stream is fed through conduit 27b to heat exchanger 26a where it will exchange heat with the countercurrent flow of enriched

nitrogen vapor that travels through conduit 56 before entering boiler 51 of distillation column B. The first and second streams then will be rejoined at 27d and fed into column B at the appropriate concentration point. Liquid from boiler 51, travels through conduit 52 to throttling valve V5 entering cold finger 44 at the top of column B (cold finger 44 part of the cryocooler cooling device 45 is functionally the same as the cold finger 44a in FIG. 4). Liquid reflux generated by valve V5 and cold finger 6 will irrigate the packing or the feed trays, as appropriate. The non-condensed vapor in conduit 53 will be fed to the appropriate concentration section of column B. The liquid from collectors 54 of column B travels through valve V6 to enter the top of column A. High-purity liquid oxygen will be removed from the bottom of column A and high-purity liquid nitrogen may be removed from collectors 55. Column A is operated at near atmospheric pressure while column B is at the pressure provided by the compressor.

[0175] FIG. 6 shows another combined column embodiment designed for the simultaneous production of liquid or gaseous oxygen and nitrogen. After the temperature of the compressed and purified feed air is appropriately reduced in heat exchanger 26, the air is fed to the mid-section of distillation column B by conduit line 27. Distillation columns A and B, in this embodiment are combined into one, two-par, unit where the two parts are separated by condenser 60, which is functionally positioned on the bottom section of column A. Condensate from high-pressure distillation column B passes through liquid line 61 and throttle valve V7 to the mid-section of low-pressure distillation column A. Enriched nitrogen gas is withdrawn from the top of column A through conduit 8 and exchanges heat in heat exchanger 26 against the incoming compressed, purified air. High-purity liquid oxygen exits at the bottom section of column A through valve V2 and liquid nitrogen passes through collectors 62 to exit through valve V3 located in this example near the top of column B.

[0176] Another double column design is illustrated in FIG. 7. The main advantage of this design is that this system provides for a reduction of both temperature and pressure process conditions, as well as an easy switch between the production of both oxygen and nitrogen or single component extraction. Condenser 60 is operatively placed at the bottom section of column A. Liquid pump 70 located on conduit line 72 provides the flow connection between condenser 60 and distillation column B. Products are extracted in the same manner as described above in the discussion relating to the embodiment illustrated in FIG. 6.

[0177] The foregoing description, for purposes of explanation, uses specific and defined nomenclature to provide a thorough understanding of the invention. However, it will be apparent to one skilled in the art that the specific details are not required in order to practice the invention. Thus, the foregoing description of the specific embodiment is presented for purposes of illustration and description and is not intended to be exhaustive or to limit the invention to the precise form disclosed. Those skilled in the art will recognize that many changes may be made to the features, to the way that some of the parts of the device may be arranged relative to one another creating various embodiments, as well as methods of making the embodiments of the invention described herein without departing from the spirit and scope of the invention. Thus, it is to be understood that the



present invention is not limited to the described exemplary methods, embodiments, features or combinations of features but include all the variation, methods, modifications, and combinations of features within the scope of the appended claims. The invention is limited only by the claims.

What is claimed is:

1. A high-purity cryogenic gas-separation/liquefaction device for the production of liquid gases, comprising:

- a) at least one means for supplying a feed gas;
- b) at least one counterflow heat exchanger to cool the incoming feed gas against the outgoing waste and/or product gas;
- c) at least one cryogenic means having a cold portion means for providing refrigeration for at least a process of condensing;
- d) at least one condensation means for condensing at least one enriched component of the feed gas, said condensation means thermally connected to said cold portion means; or directly on the cold portion means.
- e) at least one distillation means for providing for distillation of the condensed at least one component of the gas;
- f) at least one insulating means for thermally insulating said device, and
- g) liquid collecting means to collect and store one or two liquid products, as desired,

wherein the refrigeration to the cryogenic gas-separation/liquefaction process may be provided by said at least one cryogenic means alone and where gas condensation may occur at least partially directly on the cold portion means of said at least one cryogenic means which may be located inside of the thermally insulated space of the said at least one distillation means.

2. The high-purity cryogenic gas-separation/liquefaction device, as recited in claim 1, further comprising:

- a) wherein at least one cryogenic means is a cryocooler,
- b) wherein at least one condensation means is a condenser, or directly on the cold portion of the cryocooler.
- c) wherein at least one distillation means is a distillation column, and
- d) wherein at least one insulating means is a thermally insulated container,

wherein the refrigeration to the cryogenic gas-separation/liquefaction process may be provided by said at least one cryocooler alone and where gas condensation may occur directly on the cold portion of said at least one cryocooler which may be located inside or outside of the thermally insulated space of the said at least one distillation column.

3. The high-purity cryogenic gas-separation/liquefaction device, as recited in claim 2, further comprising:

- a) wherein said cryogenic means is a cryocooler,
- b) wherein said condensation means is a standard condenser thermally related to the cold portion of a cryocooler, or where the condensing means is a cold finger of the cryocooler.

c) wherein said distillation means is a distillation column, and

d) wherein said insulating means is a Dewar flask,

wherein the refrigeration to the cryogenic gas-separation/liquefaction process may be provided by said cryocooler alone and where gas condensation may occur directly on the cold portion of said cryocooler which may be located inside or outside of the Dewar flask of said distillation column.

4. The high-purity cryogenic gas-separation/liquefaction device, as recited in claim 1, further comprising wherein refrigeration to the cryogenic gas-separation/liquefaction device may be provided by a combination of said cryogenic means and a Joule-Thompson throttling process.

5. The high-purity cryogenic gas-separation/liquefaction device, as recited in claim 1, further comprising wherein said feed gas may comprise ambient air or any other gas mixture of interest.

6. The high-purity cryogenic gas-separation/liquefaction device, as recited in claim 1, further comprising wherein water and carbon dioxide are removed from said feed gas.

7. The high-purity cryogenic gas-separation/liquefaction device, as recited in claim 1, further comprising wherein said feed gas is driven into said device by a fan or compressor means.

8. The high-purity cryogenic gas-separation/liquefaction device, as recited in claim 1, further comprising wherein said feed gas passes through a multi-pass heat exchanger means for cooling.

9. The high-purity cryogenic gas-separation/liquefaction device, as recited in claim 1, further comprising wherein said cooled feed gas is introduced to said at least one distillation means at an appropriate composition point.

10. The high-purity cryogenic gas-separation/liquefaction device, as recited in claim 1, further comprising wherein an interior volume of said at least one distillation means is kept at an elevated pressure by a compressor.

11. A high-purity cryogenic gas-separation/liquefaction process for the production of liquid gases, comprising the steps of:

- a) supplying a feed gas;
- b) at least one counterflow heat exchanger to cool the incoming feed gas against the outgoing waste and/or product gas;
- c) providing at least one cryogenic means having a cold portion means for providing refrigeration for at least a process of condensing;
- d) condensing at least one enriched component of the feed gas using at least one condensation means thermally connected to said cold portion means or condensing at least one enriched component of the feed gas directly on the cold portion means.
- e) distilling said at least one component of the condensed gas using at least one distillation means;
- f) insulating said device using at least one thermally insulating means, and
- g) collecting liquid in liquid collecting means to collect and store one or two liquid products, as desired,



wherein providing the refrigeration to the cryogenic gas-separation/liquefaction process may be by said at least one cryogenic means alone and where gas condensation may occur at least partially directly on the cold portion means of said at least one cryogenic means which may be located inside or outside of the thermally insulated space of the said at least one distillation means.

**12.** The high-purity cryogenic gas-separation/liquefaction process, as recited in claim 11, further comprising wherein providing refrigeration to the cryogenic gas-separation/liquefaction device may be by a combination of said cryogenic means and a Joule-Thompson throttling process.

**13.** The high-purity cryogenic gas-separation/liquefaction process, as recited in claim 11, further comprising wherein said feed gas may comprise ambient air or any other gas mixture of interest.

**14.** The high-purity cryogenic gas-separation/liquefaction process, as recited in claim 11, further comprising removing water and carbon dioxide from said feed gas.

**15.** The high-purity cryogenic gas-separation/liquefaction process, as recited in claim 11, further comprising driving said feed gas into said device by a fan or compressor means.

**16.** The high-purity cryogenic gas-separation/liquefaction process, as recited in claim 11, further comprising passing said feed gas through a multi-pass heat exchanger means for cooling.

**17.** The high-purity cryogenic gas-separation/liquefaction process, as recited in claim 11, further comprising introducing said cooled feed gas to said at least one distillation means at an appropriate composition point.

**18.** The high-purity cryogenic gas-separation/liquefaction process, as recited in claim 11, further comprising wherein an interior volume of said at least one distillation means is kept at an elevated pressure by a compressor.

**19.** The high-purity cryogenic gas-separation/liquefaction process, as recited in claim 11, further comprising utilizing at least one boiler to produce an enriched vapor to operate said at least one distillation column.

**20.** A high-purity double column cryogenic gas-separation/liquefaction device for the simultaneous collection of a plurality of high-purity liquid gases, comprising:

- a) at least one means for supplying a feed gas;
- b) at least one counterflow heat exchanger to cool the incoming feed gas against the outgoing waste and/or product gas;
- c) a plurality of cryocoolers wherein each cryocooler has a cold portion to provide refrigeration,
- d) a plurality of condensers wherein each condenser is thermally related to the cold portion of a cryocooler,
- e) a plurality of distillation columns;
- f) at least one insulating means for insulating said device, and
- g) liquid collecting means to collect and store one or two liquid products, as desired,

wherein refrigeration to the cryogenic gas-separation/liquefaction process may be provided by said cryocoolers alone and where gas condensation may occur at least partially directly on the cold portions of the cryocoolers which may be located inside or outside of the thermally insulated space of the distillation column.

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