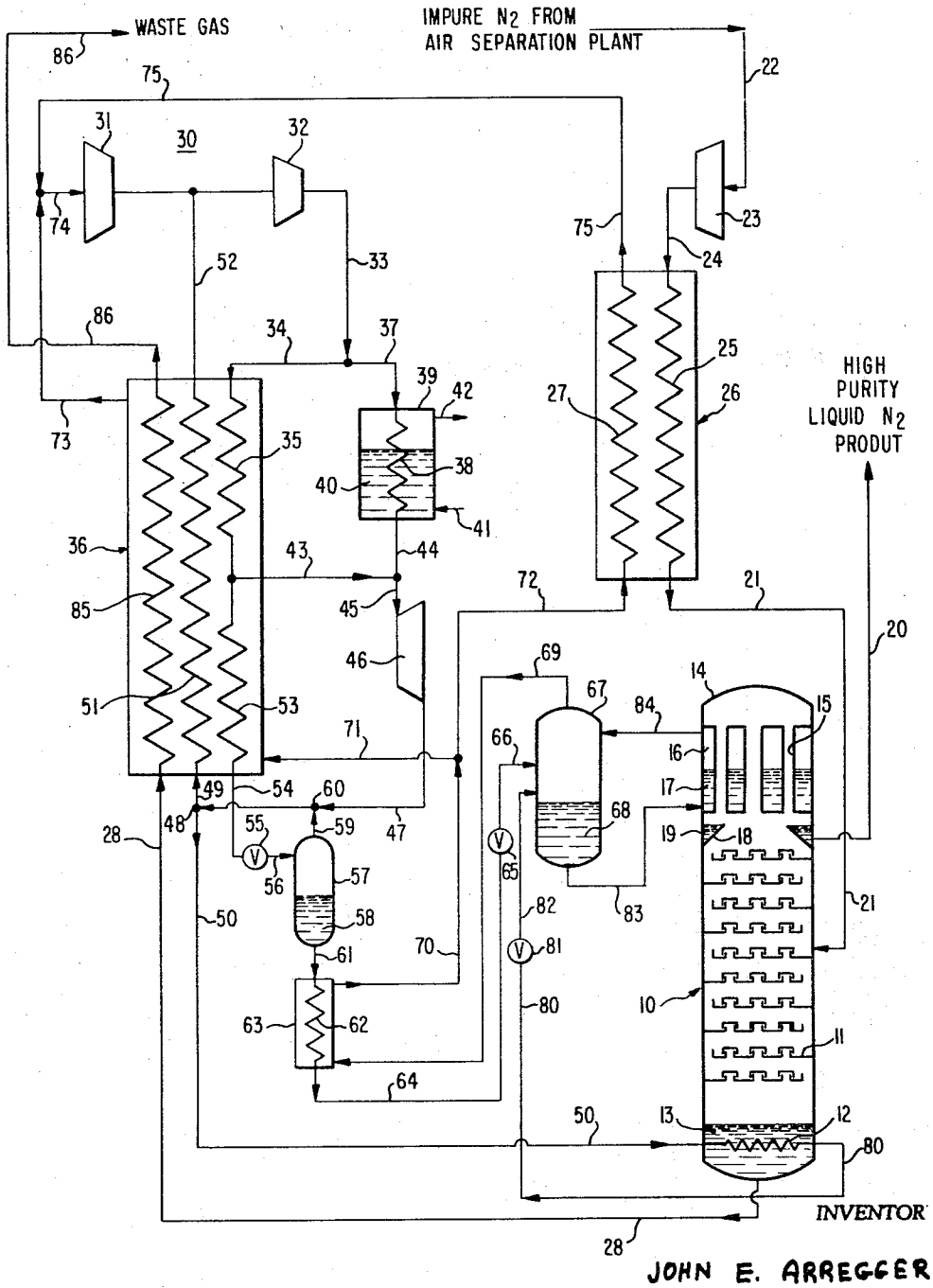
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PROCESS FOR SEPARATING GASEOUS MIXTURES

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PROCESS FOR SEPARATING GASEOUS MIXTURES

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ABSTRACT OF THE DISCLOSURE

Process for separating gaseous mixture in a fractionating operation producing low boiling point gaseous fraction and high boiling point liquid fraction in which a compressed refrigerant is cooled and expanded to effect partial liquefaction providing a liquid part and a gaseous part and in which the gaseous part is passed in heat interchange with the high boiling point liquid fraction to provide reboil for the fractionating operation and then with the liquid part used to effect liquefaction of low boiling point gaseous fraction as reflux for the fractionating operation.

This invention relates to low temperature separation of gaseous mixtures and more particularly to a low temperature process for separating from a gaseous mixture a component thereof of high purity.

Conventional low temperature processes for the separation of binary mixtures are capable of providing the high boiling point component or the low boiling point component of high purity. Arrangements have been suggested in the past for modification of conventional processes to make it possible to provide the high boiling point component of high purity and also a limited quantity of the low boiling point component of high purity. For example, in the separation of air into oxygen and nitrogen components, conventional two-stage fractionating cycles are designed to provide oxygen component of a high degree of purity and also permit the withdrawal, usually from the high pressure zone of the fractionating column, of a quantity of nitrogen also of high purity. However, since the high purity nitrogen separated in the high pressure fractionating zone is the source of liquid reflux required to effect separation of oxygen of high purity, the quantity of high purity nitrogen that may be so withdrawn is limited to but a small part of the total nitrogen available. Accordingly, in order to provide separation from a gaseous mixture of unlimited quantities of components of high purity, it is necessary to employ an auxiliary fractionating system. The present invention relates to such a system and provides a novel arrangement which makes it possible to effect the separation of a low boiling point component of high purity and in liquid phase with minimum power reqnuirements.

Accordingly, it is an object of the present invention to provide a novel low temperature process for separating from a gaseous mixture a component of high purity.

Another object is to provide a novel low temperature separation process embodying a fractionating zone and a refrigeration cycle which makes it possible to effect separation of a component of high purity, which may be in liquid phase, with minimum expenditure of power.

Still another object is to provide a novel low temperature separation process of the character described above in which the high purity component may be produced under superatmospheric pressure.

Other objects and features of the present invention will appear from the following detailed description considered in connection with the acompanying drawing, the single figure of which discloses diagrammatically a low temperature separation process embodying the principles of

the present invention. It is to be expressly understood the drawing is designed for purposes of illustration only and not to define the limits of the invention, reference for the latter purpose being had to the appended claims.

Although the present invention is disclosed and described in the environment of a system for providing high purity low boiling point component of a gaseous mixture, particularly the production of high purity nitrogen in liquid phase from waste nitrogen of an air separation plant, it is to be expressly understood that the present invention may be employed to provide from other gaseous mixtures high purity, high boiling point component as well as high purity, low boiling point component.

With reference more particularly to the drawings, there is shown therein a low temperature process embodying the principles of the present invention comprising a fractionating column 10 providing a fractionating zone containing liquid-vapor contact means in the form of a series of bubble-cap type fractionating trays 11 which may be of conventional construction. The column 10 includes a reboiler 12 immersed in a pool 13 of liquid collected in the bottom of the column and a refluxing condenser 14 having a plurality of tubes 15 communicating with the zone of the fractionating column and a shell space 16 surrounding the tubes 15 and containing a body of liquid refrigerant 17. The column 10 further includes a liquid collecting shelf 18 located below the refluxing condenser 14 and above the uppermost fractionating tray for the purpose of collecting in a pool 19 a part of liquid material condensed in the refluxing condenser 14. A conduit 20 is provided for withdrawing from the column liquid collected in the pool 19. Feed to the column is introduced at a midpoint by conduit 21. In the illustrated embodiment of the invention, the column feed comprises impure nitrogen produced from an air separation plant and delivered by conduit 22. After compression to the desired pressure in compressor 23, the compressed feed is conducted by conduit 24 for flow through passageway 25 of heat exchange device 26 in countercurrent heat interchange with relatively cold fluid flowing through passageway 27 of the heat exchange device, as described below, and thereby cooled to a lower temperature and then conducted by the conduit 21 as feed to the fractionating column 10.

The fractionating column 10 operates in a conventional manner with the vapor leaving the uppermost fractionating tray comprising low boiling point component of high purity, such as nitrogen, and with the liquid collected in the pool 13 comprising substantially the total high boiling point component of the gaseous feed mixture. The high purity low boiling point component vapor flows upwardly through the tubes 15 of the refluxing condenser 14 and is there liquefied upon heat interchange with liquid refrigerant of the body 17. The resulting liquid low boiling point component of high purity is collected in part in the pool 19 from which it is withdrawn by way of conduit 20 as product, and the remainder comprises reflux for the fractionating column which flows downwardly into the fractionating column in contact with upwardly flowing vapor derived by vaporization of liquid in the pool 13 upon flowing through the boiling coil 12 of a relatively warm fluid. Liquid bottoms are withdrawn from the column 10 through conduit 28.

The refrigeration system includes a compressor 30 including at least two stages 31 and 32, as shown. The high pressure stage 32 delivers gaseous refrigerant, such as nitrogen, under relatively high pressure to conduit 33. The compressed gaseous refrigerant in conduit 33 is divided with a major portion being conducted by conduit 34 for flow through passageway 35 of heat exchange device 36 and with the remainder fed by conduit 37 for flow

through heat exchange passageway 38 of heat exchange device 39 for heat interchange with a pool 40 of a low level extraneous refrigerant. The low level refrigerant may be derived from a closed system, not shown, connected to the heat exchange device 39 by liquid conduit 41 and vapor conduit 42. The compressed refrigerant is cooled, upon flowing through the heat exchange passageway 35, to a temperature that may correspond to the temperature of the low level refrigerant, and a portion of the cool compressed refrigerant is withdrawn from the heat exchange device 36 by conduit 43 and merged with the cool compressed gaseous refrigerant withdrawn from the heat exchange passageway 38 by conduit 44. The merged stream of cool compressed refrigerant is fed by conduit 45 to an expansion engine 46 wherein the cool gaseous refrigerant is expanded with production of external work to a lower superatmospheric pressure, hereinafter referred to as "the intermediate superatmospheric pressure," with a resulting decrease in temperature. The effluent from the expansion engine 46 is conducted by conduit 47 to juncture 48 where the effluent is divided between conduits 49 and 50. Effluent of the expansion engine 46 is conducted by the conduit 49 for flow through passageway 51 of the heat exchange device 36 in countercurrent heat interchange with compressed gaseous refrigerant flowing through the passageway 35 as described above. Such expander effluent leaves the passageway 51 at about ambient temperature and is conducted by conduit 52 to the inlet of the second stage 32 of the compressor 30 which operates at a suction pressure substantially corresponding to the pressure of the expander effluent, i.e., the intermediate superatmospheric pressure.

The remaining portion of the compressed gaseous refrigerant not withdrawn from the heat exchange device by the conduit 43 flows through heat exchange passageway 53 in countercurrent heat interchange with relatively cold fluid including the fluid flowing through the passageway 51, and leaves the heat exchange device 36 by conduit 54 at a lower temperature which may be at or close to the saturation temperature of the refrigerant at the existing pressure. The cool refrigerant is then conducted by conduit 54 to an expansion valve 55 by which its pressure is reduced to a lower superatmospheric pressure corresponding to the pressure of the effluent of the expansion engine 46, i.e., the intermediate superatmospheric pressure. Such expansion in the valve 55 results in further cooling and partial liquefaction of the refrigerant. The stream of partially liquefied refrigerant is fed by conduit 56 to phase separator 57 wherein the liquefied portion collects in a pool 58 and the vapor portion is removed by conduit 59 and merged at junction 60 with effluent from the expansion engine 46 in conduit 47. Liquefied refrigerant is withdrawn from the phase separator by conduit 61, flowed through passageway 62 of subcooler heat exchanger 63 and then conducted by conduit 64 to a pressure reducing valve 65 where its pressure is reduced to a lower superatmospheric pressure, hereinafter referred to as the "low superatmospheric pressure," and then conducted by conduit 66 to a phase separator 67 to provide a part of a pool 68 of liquid refrigerant therein. Refrigerant flashed upon flow through the pressure reducing valve 65 is withdrawn from the phase separator 67 by conduit 69 and conducted thereby for flow through the shell side of the subcooler heat exchanger 63 and then conducted by conduit 70 to conduits 71 and 72 for flow in part through the shell side of the heat exchange device 36 and for flow in part through passageway 27 of the heat exchange device 26, respectively. The gaseous refrigerant leaves the shell side of the warm end of the heat exchange device 36 by conduit 73 at substantially atmospheric temperature and is conducted thereby to the inlet 74 of the first stage 31 of the compressor 30 which operates at a suction pressure corresponding substantially to the pressure of the phase separator 67, i.e., the low superatmospheric pressure, while the gaseous refrigerant leaves the passageway 27 of the heat exchange device 26 also at substantially ambient temperature, and is conducted by conduit 75 to the inlet 74 of the first compression stage 31.

Gaseous refrigerant under intermediate superatmospheric pressure in the conduit 50, derived in part from effluent of the expansion engine 46 and from the phase separator 57, is conducted thereby for flow through the boiling coil 12 of the fractionating column 10 wherein the gaseous refrigerant is liquefied upon vaporization of liquid in the pool 13. The so liquefied refrigerant is conducted by conduit 80 to a pressure reducing valve 81 whereby the pressure of the liquid is reduced to the low superatmospheric pressure and then conducted by conduit 82 to the phase separator 67 to provide a part of the liquid of the pool 68. Refrigerant flashed in the valve 81 is withdrawn from the phase separator 67 by conduit 69 along with the vapor flashed in the pressure reducing valve 65. Liquid refrigerant is withdrawn from the pool 68 by conduit 83 and fed thereby to the shell side of the refluxing condenser 14 to provide the pool 17 of liquid refrigerant and liquid refrigerant which is vaporized upon liquefaction of nitrogen vapor flowing through the tubes 15 of the refluxing condenser is withdrawn from the shell side 16 of the refluxing condenser by conduit 84 and fed thereby to the phase separator 67 above the pool of liquid 68. Such refrigerant vapor leaves the phase separator 67 by way of the conduit 69 for flow through the shell side of the subcooler 63 and then is divided and fed through the shell side of the heat exchanger 36 and passageway 27 of the heat exchanger 26 and then returned to the inlet of the low stage compresor 31 along with the refrigerant vapor flashed in the pressure reducing valves 65 and 81. Liquid bottoms collecting in the fractionating column 10 are withdrawn from the pool 13 by conduit 28 and conducted thereby for flow through a heat exchange passageway 85 of the heat exchange device 36 in countercurrent heat interchange with the compressed gaseous refrigerant flowing through the passageways 35 and 53, such liquid being vaporized upon flowing through the passageway 85 and being withdrawn from the heat exchange device 36 by conduit 86 as a waste gas.

The fractionation and refrigeration systems described above are interrelated in a novel manner which obtains the separation of high purity component with minimum power requirements. In accordance with the present invention, a portion of the refrigerant vapor at the intermediate superatmospheric pressure is conducted by the conduit 50 for flow through the reboiler coil 12 to provide the required reboil for the fractionating column 10 upon vaporization of liquid of the pool 13 with concomitant liquefaction of the refrigerant which is subsequently reduced in pressure in the valve 81 to provide a portion of the liquid refrigerant of the pool 68 which in turn provides the refrigeration requirements of the refluxing condenser 14. Furthermore, a portion of the cold refrigerant vapor at the low superatmospheric pressure is used to cool the incoming feed mixture in the heat exchange device 26 and liquid bottoms withdrawn from the column 10 are flowed through the heat exchange device to aid in cooling the compressed refrigerant. The novel inter-arrangement and coaction of the refrigeration and fractionating systems make it possible to obtain a high level of usable refrigeration for a given energy input and provide an overall process which operates at high efficiency as compared to prior arrangements.

As an operating example of the present invention, gaseous mixture comprising impure nitrogen from an air separation plant under a pressure of about 18 p.s.i.a. and containing 99% nitrogen and 1% oxygen and other impurities is compressed in the compressor 23 to about 55 p.s.i.a., and thereafter cooled to about —287° F. in the heat exchange device 26 and then introduced into the fractionating column 10. The fractionating column 10 operates at a pressure of about 50 p.s.i.a. and liquid nitrogen of a purity of 99.99+ collects in the pool 19 at a tem-

perature of about —298° F. while the liquid in the pool **13**, which contains substantially the total oxygen and other impurities, is at a temperature of about —288° F. High purity nitrogen, in liquid phase, is withdrawn from the column by conduit **20** under a pressure of about 55 p.s.i.a. and at a temperature of about —298° F. On the basis of 350 mol per hour of impure nitrogen entering the cycle by conduit **22**, about 343 mol per hour of high purity liquid nitrogen will be withdrawn by the conduit **20** and about 7 mol per hour of liquid fraction will be withdrawn from the column by conduit **28**. The refrigeration system employs nitrogen as the refrigerant which is compressed to about 3,000 p.s.i.a. in high pressure stage **32**. A major portion, about 60%, of the compressed refrigerant flows through the heat exchange passageway **35** and the remaining portion flows through the passageway **38** of the heat exchange device **39** wherein it is cooled to about —50° F. in heat exchange with a liquid refrigerant such as Freon. The gaseous refrigerant withdrawn from the heat exchange device **36** by way of conduit **43**, about 20% of the total compressed refrigerant, is at a temperature of about —50° F. and the combined streams are expanded in expansion engine **46** to an intermediate superatmospheric pressure of about 96 p.s.i.a. and thereby cooled to about —278° F. The remaining portion of the gaseous refrigerant leaves the passageway **53** at about —280° F. and upon expansion in the valve **55** to the intermediate pressure of about 96 p.s.i.a. is partially liquefied with the resulting liquid collecting in the pool **58** being at a temperature of about —284° F. and with the unliquefied vapor leaving the phase separator **57** by a conduit **59** at substantially the same temperature. Liquid withdrawn from the pool **58**, comprising about 33% of the total compressed refrigerant, is further cooled to about —297° F. in the subcooler heat exchanger **63** and upon pressure reduction in the valve **65** to the low superatmospheric pressure of about 41 p.s.i.a. forms a part of the liquid of the pool **68** at a temperature of about —302° F. The refrigerant vapor in conduits **47** and **59**, which comprises the remainder of the total compressed refrigerant, is divided at junction **48** with a portion thereof sufficient to provide the reboil requirements of the fractionating column **10** being passed by conduit **50** for flow through the reboiler **12** and there liquefied. For the operating condition of the column **10** set forth in this example, about 595 mol per hour of refrigerant vapor at a pressure of about 95 p.s.i.a. and a temperature of about —282° F. is flowed through the reboiler coil **12**. Liquefied refrigerant leaves the reboiler **12** at a temperature of about —284° F. and is reduced in pressure in valve **81** to the low superatmospheric pressure of about 41 p.s.i.a. to provide the remainder of the liquid forming the pool **68**. Liquid refrigerant from the pool **68** provides refrigeration for the refluxing condenser **14** to effect liquefaction of the high purity nitrogen formed in the fractionating column **10** operating under a pressure of 50 p.s.i.a. Liquid bottoms from the fractionating column **10**, the remaining refrigerant vapor at the intermediate superatmospheric pressure and a part of the refrigerant vapor at the low superatmospheric pressure flow outwardly though the heat exchange device **63** to effect cooling of the compressed gaseous refrigerant as described above, while a portion of the cold refrigerant vapor at the low superatmospheric pressure, about 350 mol per hour, flows through the passageway **27** of the heat exchange device **26** to effect cooling of the compressed feed mixture to about —287° F. The refrigerant vapors leave the heat exchange devices at substantially ambient temperature and the vapors at the low superatmospheric pressure enter the suction inlet of the first stage of compression **31** a substantially 41 p.s.i.a. while the refrigerant vapors at the intermediate superatmospheric pressure enter the suction inlet to the second stage compression **32** at substantially 95 p.s.i.a.

There is thus provided by the present invention a novel system for separating from gaseous mixtures a component of high purity, such as waste nitrogen gas from an air separation plant, which is capable of providing the desired component without limitation on quantity at any desired wide range of pressures and in liquid phase.

While only one embodiment of the invention has been disclosed and described herein, it is to be expressly understood that changes and substitutions may be made therein without departing from the spirit of the invention as well understood by those skilled in the art. Reference therefore will be had to the appended claims for a definition of the invention.

What is claimed is:

1. Process for producing high purity component of a gaseous mixture in a low temperature operation employing a fractionating zone, comprising the steps of

compressing and cooling gaseous mixture and feeding cool compressed gaseous mixture to the fractionating zone under a first pressure to effect separation of the gaseous mixture into a gaseous low boiling point fraction and a liquid high boiling point fraction,

cooling compressed gaseous refrigerant and expanding cooled compressed gaseous refrigerant to a second pressure to effect partial liquefaction of the refrigerant,

separating the unliquefied refrigerant of the partially liquefied refrigerant from the liquefied refrigerant thereof to provide a first liquid portion and a first vapor portion,

passing first liquid portion in heat interchange with gaseous low boiling point fraction produced in the fractionating zone to effect liquefaction of gaseous low boiling point fraction,

passing first vapor portion in heat interchange with liquid high boiling point fraction produced in the fractionating zone to effect vaporization of liquid high boiling point fraction and liquefaction of the first vapor portion to provide a second liquid portion,

passing second liquid portion in heat interchange with gaseous low boiling point fraction produced in the fractionating zone to effect liquefaction of gaseous low boiling point fraction,

utilizing vaporized liquid high boiling point fraction as reboil for the fractionating zone,

utilizing liquefied gaseous low boiling point fraction as reflux for the fractionating zone,

and withdrawing low boiling point fraction from the fractionating zone as high purity component of the gaseous mixture.

2. Process for producing high purity component of a gaseous mixture as defined in claim **1** in which liquefied low boiling point fraction is withdrawn from the fractionating zone.

3. Process for producing high purity component of a gaseous mixture as defined in claim **1** including the steps of

reducing the pressure of the first liquid portion and of the second liquid portion to a third pressure lower than the second pressure prior to passing the first liquid portion and the second liquid portion in heat interchange with gaseous low boiling point fraction.

4. Process for producing high purity component of a gaseous mixture as defined in claim **3** in which the first pressure is less than the second pressure.

5. Process for producing high purity component of a gaseous mixture as defined in claim **1** in which first liquid portion and second liquid portion is vaporized upon heat interchange with gaseous low boiling point fraction,

and in which such vaporized first liquid portion and second liquid portion is utilized to cool the compressed gaseous mixture and thereafter used to form the compressed gaseous refrigerant.

6. Process for producing high purity component of a gaseous mixture as defined in claim **5** including the steps of

withdrawing liquid fraction from the fractionating zone and passing withdrawn liquid fraction in heat interchange wth compressed refrgerant to effect cooling of the compressed refrigerant.

7. Process for producing high purity component of a gaseous mixture as defined in claim 6 including the step of

utilizing a part of the first vapor to cool the compressed gaseous refrigerant and thereafter to use such vapor to form the compressed refrigerant.

8. Process for producing high purity component of a gaseous mixture as defined in claim 7 including the steps of

expanding a part of cooled compressed gaseous refrigerant with production of work to provide a part of the first vapor portion.

9. Process for producing high purity component of a gaseous mixture as defined in claim 8 including the step of

cooling by an extraneous refrigeration a portion of the cooled compressed gaseous refrigerant prior to the expansion with production of external work.

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