

[54] **PROCESS FOR PURIFICATION OF NATURAL GAS**

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[51] Int. Cl. .... **F25j 1/00, F25j 3/06**

[58] Field of Search ..... **62/911, 23, 24, 27, 28, 29, 62/22, 41**

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[57] **ABSTRACT**

Process for the purification of natural hydrocarbon gases which, as may be produced at a wellhead, are contaminated by substantial proportions of incombustible constituents such as nitrogen. Contaminated gas at elevated pressure is condensed by cooling in two steps, thereafter expanded to approximately atmospheric pressure to effect phase separation between a hydrocarbon-rich liquid and a contaminant-rich vapor. The hydrocarbon-rich liquid is partially vaporized and a second phase separation yields a liquid enriched in hydrocarbon. The vapor from the second separation is cooled and returned to the first separation while the vapor product from first separation along with the liquid product from the second separation are separately employed to effect condensation of the contaminated hydrocarbon.

**13 Claims, 3 Drawing Figures**

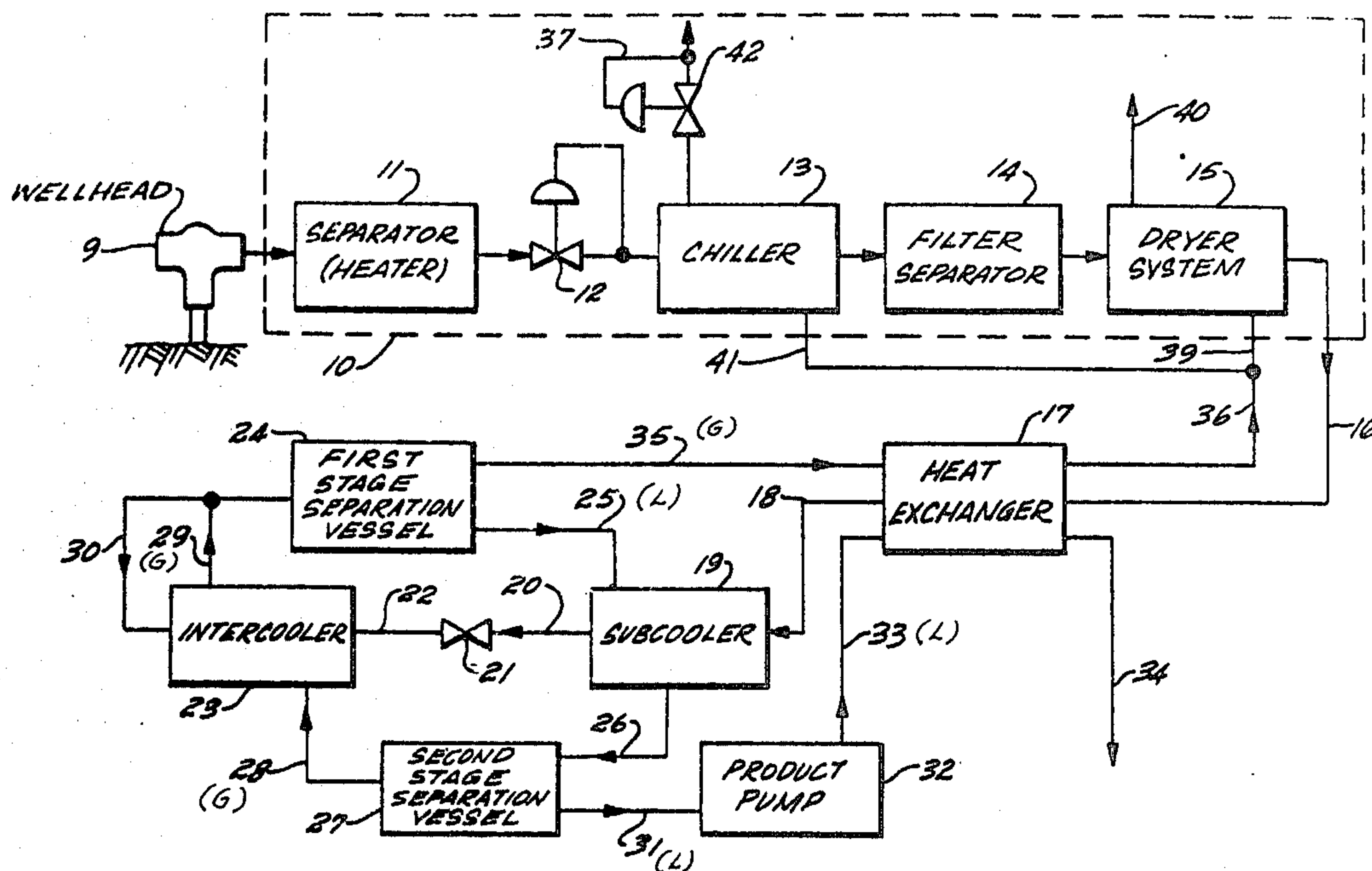


FIG. 1

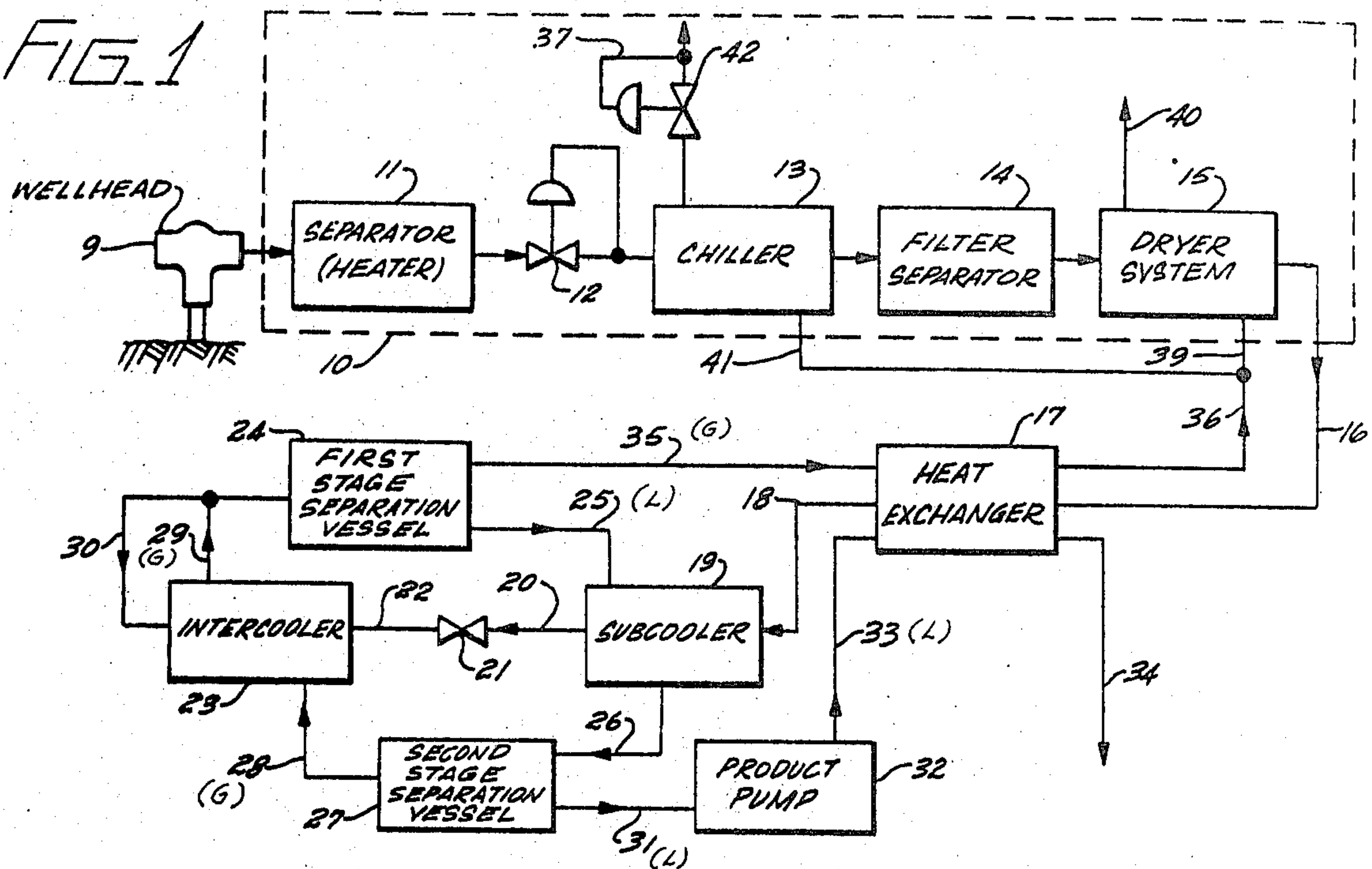
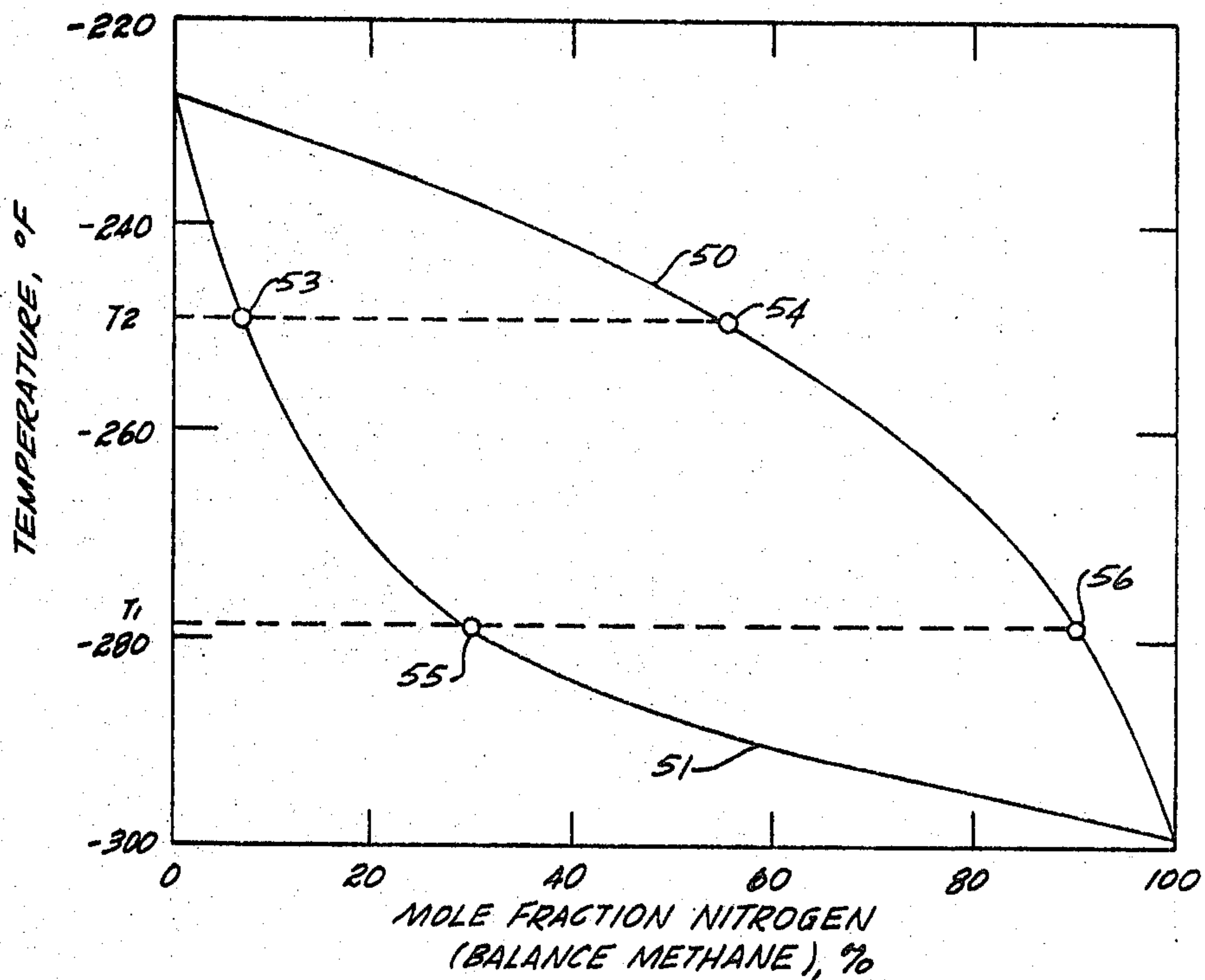


FIG. 2



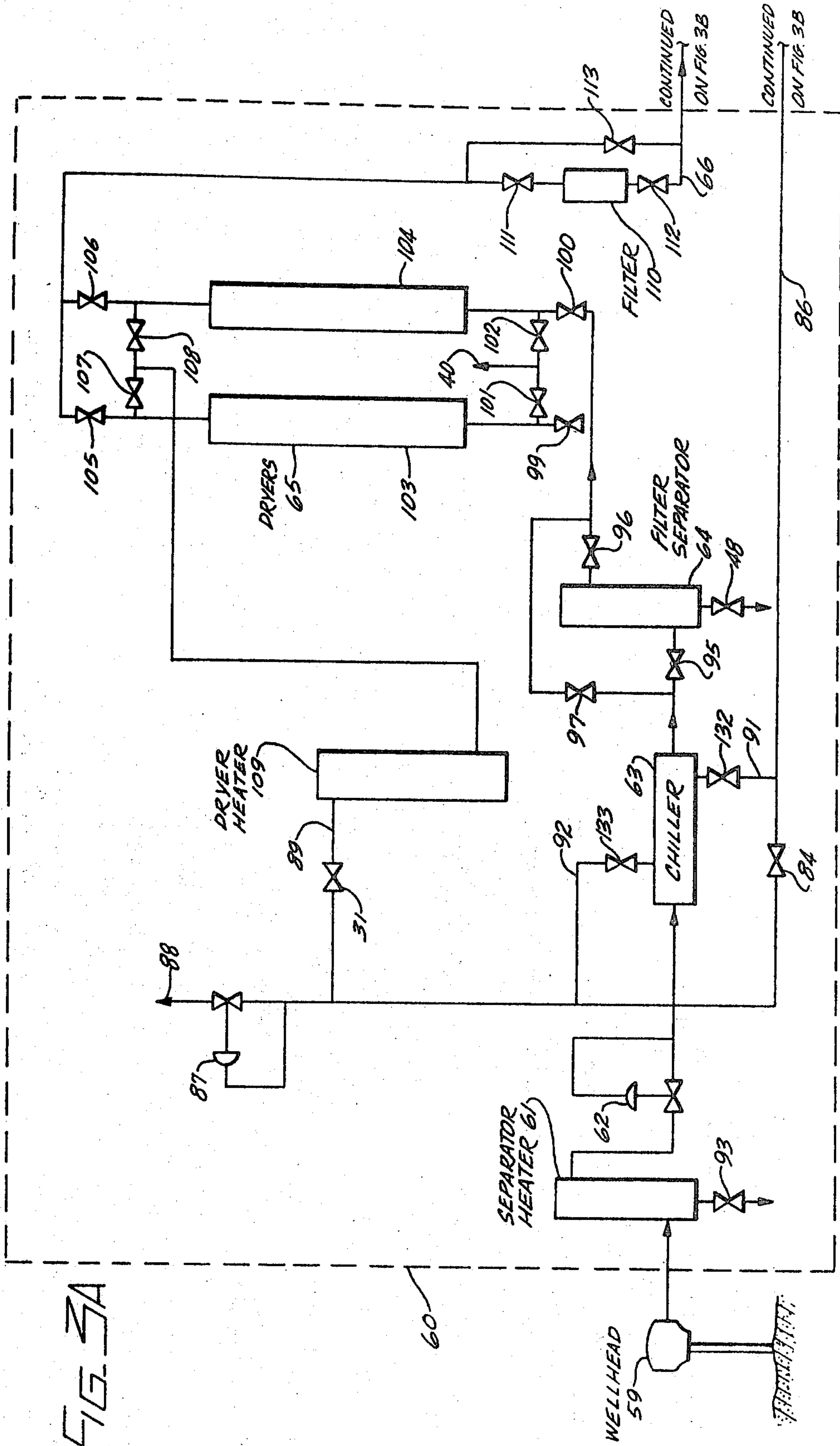
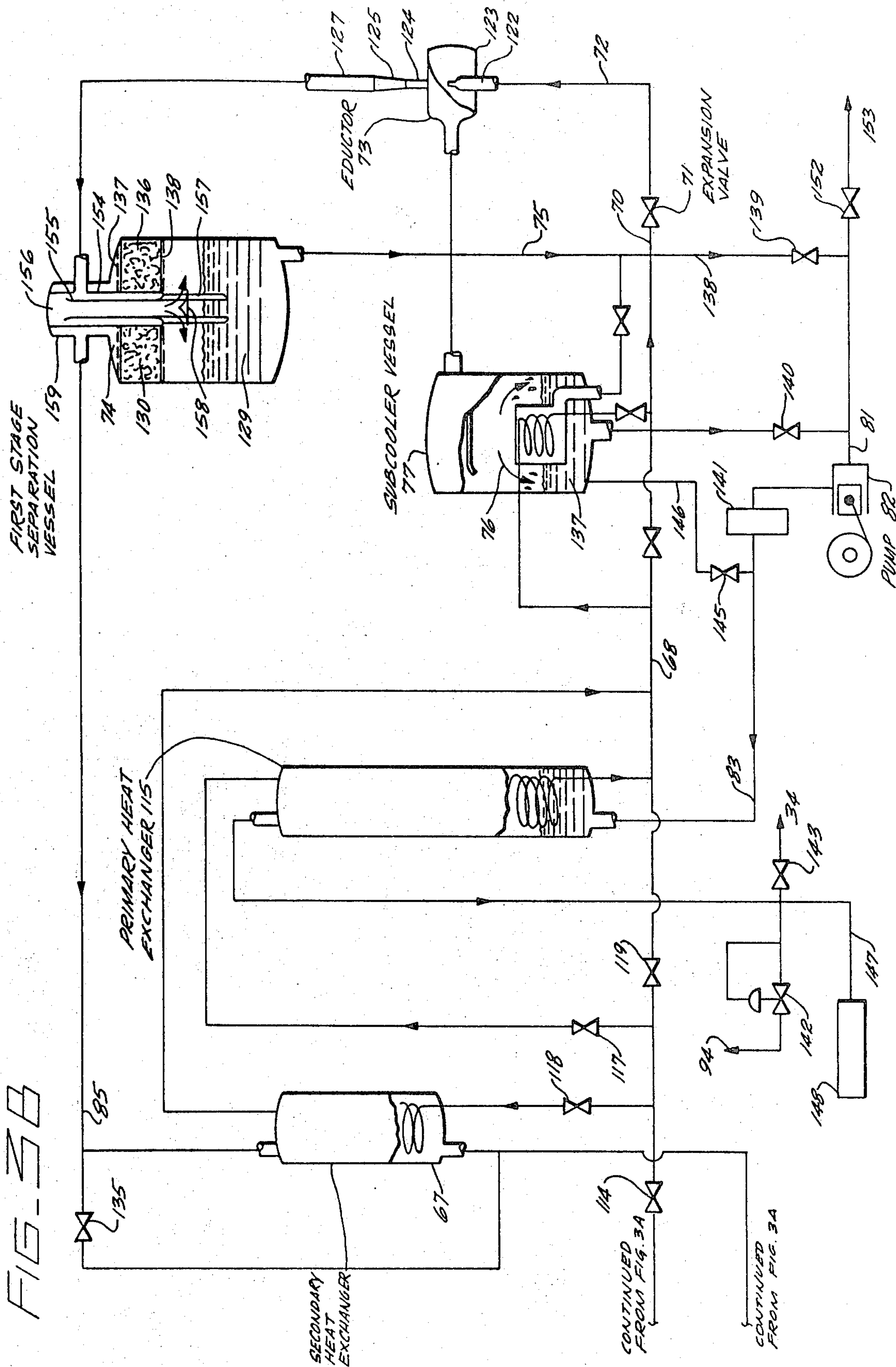


FIG. 3A

CONTINUED  
ON FIG. 3B

CONTINUED  
ON FIG. 3B







## PROCESS FOR PURIFICATION OF NATURAL GAS

## BACKGROUND OF THE INVENTION

The presence of substantial proportions of incombustible constituents in produced natural hydrocarbon gases from wellheads or other sources can render such gases commercially useless since the contaminant content reduces the heating value (BTU) of the gases below the minimum level acceptable for fuel purposes by gas distribution utility companies. The presence in the United States and other countries of known gas reservoirs which are substantially unexploited because the formation hydrocarbons are mixed with excessive quantities of nitrogen, generally in the range of 20 percent or more by volume, of itself indicates the need for an economical and effective process for purifying such natural gases to the extent required to meet commercial standards for its use as a fuel. In our U.S. Pat. No. 3,488,677, we describe a process for accomplishing this objective. The present invention represents an improvement over our patented process.

## SUMMARY OF THE INVENTION

Through the present invention, there is provided a process which is economical with respect to operational and installation costs and yet enables purification of high-pressure contaminated hydrocarbon gases to the extent that such gases are made marketable. The process comprises cooling the high-pressure contaminated natural gas in two successive steps to condense at least a substantial part of the gas stream by passing it in indirect heat exchange relationship first with at least two separate fluid streams and second with a boiling liquid. The cooled feed mixture is then expanded to substantially atmospheric pressure in a first separation zone to produce a single hydrocarbon-rich liquid phase and a single contaminant-rich vapor phase. The liquid and vapor phases are separated and the liquid is further enriched by partial vaporization in heat exchange with the natural gas in the second step of cooling referred to earlier. The partially vaporized stream from the first separation thus yields a second hydrocarbon-rich liquid phase and a second contaminant-rich vapor phase in a second separation zone. These are in turn separated, and the second vapor phase is cooled and reintroduced into the first separation zone. The liquid from the second separation and the vapor from the first are then separately passed as the fluid streams which cool the feed mixture by indirect heat exchange in the first cooling step before being withdrawn from the process as first and second product streams, respectively.

In our U. S. Pat. No. 3,488,677, we pointed out that by expanding the mixture to substantially atmospheric pressure it is possible to obtain effective separation of the constituents without requirement of conventional fractionation. We stated further that we had found by this expansion of the chilled fluid to substantially atmospheric pressure, the methane entrained and discharged along with the contaminant in the gaseous phase is comparable in terms of energy value to the power requirement of reflux refrigeration in a conventional fractionation system.

We have now found that a further increase in the effectiveness of the separation of the constituents is possible, still without the requirement of conventional fractionation and still without the necessity for any ad-

ditional consumption of power. This increase in separation effectiveness is achieved by performing a second separation of the liquid phase withdrawn from the first separation in a second separation zone following heating and partial vaporization of the liquid by indirect heat exchange with the cool feed stream in a subcooler. The vapor phase from this second separation contains an appreciably higher hydrocarbon content than the vapor phase from the first because of the higher temperature of the liquid and vapor in the second separation zone. However, this hydrocarbon content is not lost but recovered by first cooling the vapor phase from the second separation in an intercooler and then returning it to the first separation zone. It will be apparent that there is an additional cooling imparted to the contaminated feed mixture prior to its expansion when it is passed in the second step of cooling through the subcooler to provide the heat required to partially vaporize the liquid phase from the first stage of separation. We have found that this additional cooling is sufficient, after expansion of the contaminated feed mixture, to provide the cooling required to partially condense the vapor phase from the second stage of separation in the intercooler. The hydrocarbon content of the vapor phase from the first separation is less than that from the second, as explained above. Thus, the liquid phase from the second separation and the vapor phase from the first separation have the highest and lowest hydrocarbon content, respectively, and these are withdrawn as the separated products from the process. Clearly, the effectiveness of the separation is enhanced beyond that achieved in withdrawing the liquid and vapor phases from a single separation as the separated products.

The term "high pressure" as applied to the contaminant gas to be purified is used herein with reference to the pressures at which natural gas is typically produced at a wellhead or at which it is maintained in contemporary transmission systems. These pressures, however, do not represent critical limitations on the process which may accept gaseous feed at higher or lower pressures down to at least several atmospheres. Although the process is applicable to various contaminants, it is described herein with reference to nitrogen as the contaminant, an incombustible found in many hydrocarbon reservoirs. It will also be understood that the term "purification" as used herein refers to removal of a sufficient amount of contaminant to render the hydrocarbon gas marketable under the prevailing local conditions.

The process will be better understood from the following description of the appended drawings.

## BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a block diagram of a process system showing one embodiment for the practice of the invention;

FIG. 2 is an isobaric curve showing methane-nitrogen phase separation at 50 p.s.i.a.; and

FIG. 3 is a schematic flow sheet of a process system depicting another embodiment falling within the scope of the invention.

## DESCRIPTION OF THE PREFERRED EMBODIMENTS

With particular reference to FIG. 1, high pressure gaseous natural gas mixture contaminated by a large proportion of nitrogen, as for example 60 percent by



volume, produced at a wellhead 9 at a high pressure, which is typically in excess of 100 atmospheres, is passed first through a feed conditioning system 10 shown in the upper part of the Figure and then through the two-stage separation process shown in the lower part of the Figure, the process system encompassing both aspects.

The conditioning system is comprised of a separator 11, to remove free liquids from the produced gas, and a pressure regulator 12 to reduce feed gas pressure, if necessary. The separator may also include a heater, so that the temperature of the feed gas following pressure reduction in regulator 12 will remain above the hydrate formation temperature of between about 45°F and 65°F. A chiller 13 may be provided to cool the feed gas in cases where the temperature following the pressure regulator 12 is appreciably higher than the hydrate formation temperature. In the chiller, the feed gas is cooled by one of the product streams, such as the nitrogen-rich stream as shown here entering chiller at 41 and discharging at 42. Suspended water and hydrocarbons condensed from the feed gas in the chiller are collected in a filter-separator 14. The feed gas is passed through a dryer system 15 to remove vaporized water and troublesome heavier hydrocarbons. The dryer system typically comprises beds of desiccant or molecular sieve as well as activated charcoal, which must be periodically regenerated using hot, dry gas heated by gas or electrical heaters included as part of the dryer system. Gas for this purpose may be drawn from the nitrogen-rich stream through a line 39 and discharged from the dryer system through line 40.

The conditioning system functions to stabilize the pressure and temperature of the feed mixture and to remove therefrom unwanted components which would freeze in the subsequent processing.

The conditioned feed mixture leaves the conditioning system through line 16 and enters a heat exchanger 17 which may consist of one or more units arranged so as to take full advantage of the cooling capacity of the separate streams of hydrocarbon-rich product and nitrogen-rich product as they are warmed from the low temperatures at which they are separated in the process, as will be described later. Typically, the feed stream passes in parallel counterflow heat exchange relation to each of the product streams, which in turn flow through separate passages or else through separate heat exchange units, as is the case in a preferred embodiment to be described in connection with FIG. 3.

The cooled feed mixture is passed through a line 18 to the subcooler 19 where it is further cooled by heat exchange with a stream of low temperature boiling liquid drawn from the first stage separation vessel. The subcooler is typically a tube-and-shell heat exchanger with the cooled feed passing through coiled tubes in counterflow relation to the boiling liquid in the shell.

The subcooled feed mixture leaves the subcooler through line 20 still at a substantial fraction of its original pressure as controlled by the regulator 12. The small loss of pressure arises from the nominal pressure drop in each of the units of the preconditioning system and in passage through the tubes of the heat exchanger and subcooler. The feed stream is normally cooled at a pressure above its critical pressure, in which case the stream is liquidized during condensation without passing through a two-phase region. However, in the case of a feed mixture below its critical pressure, the sub-

cooled feed from the subcooler could be either a liquid or a two-phase mixture, depending on the pressure and other conditions of operation.

The subcooled feed is expanded through expander 21 to a pressure as near atmospheric as is feasible and consistent with design requirements of the various parts of the system. In the typical case considered here (see Table I, page 11), the pressure, after the final expansion, is about 35 p.s.i.g. As will be explained in more detail subsequently, the choice of a separation pressure is a compromise between improved efficiency of separation favored by pressures near, or even below, atmospheric and the reduced size of pipes, valves, vessels and heat exchangers needed to handle the reduced volume of the gaseous streams of the process at a pressure of several atmospheres. A further advantage of relatively higher separation pressure is that the increased pressure, at which the nitrogen-rich product is available after passage through the heat exchanger 17, provides a usable source of dry gas for regeneration of the dryers. In comparison to the single-stage process described in our U.S. Pat. No. 3,488,677, the present two-stage process exhibits a markedly reduced dependence of separation efficiency on the pressure at which the separations are performed, within a range of pressures of up to several atmospheres, absolute.

The expander 21 is typically a throttle valve which produces a Joule-Thompson, or constant enthalpy expansion. Alternatively, an expansion engine or turbine could be used to achieve a nearly isentropic expansion affording greater cooling, still within the scope of the invention. Such an isentropic expander achieves improved cooling of the subcooled natural gas mixture by extraction of energy from the stream for performance of external work. The expanded feed from the expander passes in line 22 to an intercooler 23 where a portion of its low temperature is used to cool a stream of gas returning from the second stage separation vessel to the first stage. The intercooler provides passages for efficient transfer of heat from the interstage stream to the expanded feed stream.

The expanded feed stream and the interstage gas stream approach thermal equilibrium at the discharge of the intercooler and are passed through lines 30 and 20, respectively, into a first stage separation vessel 24. The streams may be mixed before entering the vessel or, in a preferred embodiment as shown in FIG. 3, the streams are mixed in the intercooler to achieve the desired thermal equilibrium by direct heat exchange.

The two streams enter the vessel and are separated into a single liquid phase and a single vapor phase which approach thermal equilibrium at the reduced separation pressure. The fraction of nitrogen in the liquid phase and in the vapor phase at equilibrium depends solely upon the pressure and temperature of the two phases; thus, for a given constant pressure in the first stage separation vessel, the composition of the two phases is governed by the temperature alone. In effect, since the second product stream containing principally nitrogen is derived from the vapor phase in the first stage separation vessel, the hydrocarbon discharged along with the nitrogen-rich second product stream, and also therefore the overall process efficiency, is directly controlled by the degree of cooling of the two streams introduced into the first stage of separation.

The nitrogen-rich gaseous phase separated in the first stage is withdrawn through line 35 and passed through



separate passages of the heat exchanger 17 where it is warmed to near the temperature of the conditioned feed and then discharged through lines 36 and 41 to chiller 13 and back-pressure regulator 37. The back-pressure regulator maintains a controlled, nearly constant pressure in the first stage separation vessel and affords a positive pressure at which the nitrogen-rich gas may be withdrawn through line 39 for dryer regeneration.

The hydrocarbon-rich liquid phase separated in the first stage comprises a first interstage stream which is withdrawn through line 25, partially vaporized by passage in heat exchange relation to the relatively warmer feed stream entering the subcooler 19, and then introduced as a two-phase mixture into the second stage separation vessel 27 and through line 26. As in the case of the first stage, the two-phase mixture is separated into a second single hydrocarbon-rich liquid phase and a second single nitrogen-rich gaseous phase in approximate thermal equilibrium, the liquid phase being richer in hydrocarbon than the first interstage stream from which it was derived. As recounted previously, the nitrogen content of the liquid and vapor phases depends only upon the separation temperature, for a given controlled pressure. Consequently, and as hereinafter explained in relation to the phase diagram of FIG. 2, the purity of the hydrocarbon-rich first product stream derived from the liquid phase in the second separation vessel is governed by the amount of warming of the first interstage stream in passing through the subcooler prior to introduction into the second separation vessel.

The gaseous phase separated in the second separation vessel comprises a second interstage stream and passes through line 28 to the intercooler 23, and thence back to the first stage separator 24, as previously described.

The liquid withdrawn from the second stage separation vessel through line 31 comprises the hydrocarbon-rich first product stream. If delivery of this product is to be at a pressure above the separation pressure, it can be advantageously pumped to its final delivery pressure while still in a liquid state by means of a product pump 32. Power required to pressurize the first product stream as a liquid is many times less than that required to compress the product in the gaseous state.

The pressurized first product liquid is passed through line 33 into separate passages of heat exchanger 17 where it is vaporized and warmed to near the temperature of the entering conditioned feed gas mixture. The gaseous hydrocarbon-rich first product stream is then discharged through discharge line 34 at final delivery pressure as provided by pump 32, but slightly reduced by nominal pressure drop in the heat exchanger and related discharge piping.

In the case of the natural gas mixture considered in connection with the description of this Figure, namely one containing about 60 percent by volume of nitrogen contaminant and flowing from the conditioning system into the process system at a rate of about 12 million standard cubic feet per day, the process yields a hydrocarbon-rich product containing, for example, less than 7 percent of nitrogen at a rate of about 4,300,000 standard cubic feet per day. Table I shows the approximate pressure, temperature, nitrogen content, and weight flow rate at various locations in the process system operating under one typical set of controlled conditions.

TABLE I

	Station per fig.1	Pressure (psia)	Temp. (°F)	Composition (Vol. %N <sub>2</sub> )	Weight Flow (lb./sec.)
5	Line 16	1500	65	60	8.00
	Line 18	1470	-182	60	8.00
	Line 20	1450	-255	60	8.00
	Line 22	50+	-286	60	8.00
	Line 29 and 30 combined	50	-279	60	10.62
10	Line 25	50	-279	30	4.70
	Line 26	50+	-249	30	4.70
	Line 28	50+	-249	55	2.62
	Line 31	50+	-249	6.5	2.08
	Line 33	600	-244	6.5	2.08
15	Line 34	590	33	6.5	2.08
	Line 35	50	-279	90	5.92
	Line 36	40	32	90	5.92
	Line 42	14.7	32	90	5.92

The different temperatures of the first and second separations, as for example line 25 withdrawing the first interstage liquid stream at -279°F and line 28 withdrawing the second interstage vapor stream at -249°F, illustrate a 30° difference in temperature between these separations. It is this difference between the temperature at which the two separations are performed that results in the improved efficiency of separation. The example presented does not represent a limiting, but rather a typical case. The nitrogen content of the hydrocarbon-rich product (lines 31, 33 and 34) can be higher or lower as required by the gas user. Such variation is accomplished by lower or higher temperature in the second stage separation vessel 27, respectively. Likewise, the residual methane content of the nitrogen-rich product stream can be varied; for example, increasing subcooler heat transfer capacity lowers the temperature of the subcooled feed in line 20, resulting in a lower temperature in the first stage separation vessel 24 and a corresponding reduction of methane in the vapor leaving the first stage through line 35 and the process system through line 36.

We have found that the hydrocarbon content of the vapor from the first stage of separation and that of the liquid from the second stage of separation can be controlled nearly independently over a wide, practically useful, range of values by controlling the temperature in each of the separation zones as independently as possible, as will be described. One theoretical limitation on this otherwise nearly independent control of the two separation temperatures is reached when the hydrocarbon content of the liquid phase from the first stage of separation is equal to that of the vapor phase from the second. This limit can be understood by reference to FIG. 2, which shows an isobar of the methane-nitrogen system with composition of vapor given by curve 50 when in equilibrium with liquid at any temperature. Liquid composition at each such temperature is given by curve 51. In any particular case the methane-rich product composition is assumed to be specified, thus fixing the point 53 on the liquid composition curve. Accordingly, the temperature  $T_2$  of the second stage of separation is determined and, therefore, the composition of the vapor from the second stage at 54. Similarly, it can be seen that decreasing the methane content of the first stage vapor, shown by the point 56, requires a reduction of the first stage separation temperature  $T_1$  and an accompanying increase in nitrogen content of the first stage liquid, shown by the point 55. A material and species balance clearly shows that when the composition of the first stage liquid, point 55, and second



stage vapor, point 54, are identical, all of the interstage liquid flowing from the first stage must be vaporized in the second stage and none is available as a methane-rich liquid product from the system. In order to approach the limit referred to previously, the nitrogen content of the liquid from the first stage of separation would be increased from 30 percent to 55 percent (assuming the purity of the hydrocarbon-rich product to be left unchanged). This limiting case would require additional cooling of the feed gas mixture such that the temperature of the combined streams from lines 29 and 30 entering the first separation zone would be about  $-289^{\circ}\text{F}$ , some  $10^{\circ}\text{F}$  colder than the temperature under the conditions shown, and the methane content of the nitrogen-rich vapor product from the first stage of separation would be about 4 percent.

It can be seen that as long as the foregoing limit is not too closely approached it is possible to control the hydrocarbon fraction discharged along with the nitrogen-rich stream nearly independently of the composition of the methane-rich product by adjusting the capacity of the heat exchanger and subcooler, and enlarging the equipment associated with the first and second interstage streams in order to handle the greater interstage flow rates. These changes can be incorporated easily in applications where greater efficiency of separation warrants the increased equipment capacity, still within the scope of the present invention. It can be seen further, that this nearly independent control of the composition of the methane-rich product and the nitrogen-rich product leads to the result that the separation efficiency is relatively insensitive to the separation pressure as long as the phase separation behavior exhibits fairly sharp separation such as that shown in FIG. 2. In practice, this permits production of a purified hydrocarbon-rich product containing only a few percent of contaminant while the separated nitrogen-rich stream contains a comparably low percentage of hydrocarbon at separation pressures in the range of up to several atmospheres, absolute. This result is in contrast to the single stage process described in our referenced patent in which separation efficiency is very sensitive to pressure and in practice the separation is performed as near atmospheric pressure as possible.

In the illustrative example, shown in Table I, both separation vessels as shown are at a pressure of about 50 p.s.i.a., with the second stage vessel at slightly higher pressure. This may be accomplished by employing a static head of liquid flowing down through line 25, subcooler shell 19 and line 26 to maintain a slight positive pressure in the lower vessel 27. Alternatively, a liquid pump in line 25 may be used for the same purpose, or else a gas pump can be used to force gas through line 28, intercooler 23, and line 29 to the first stage vessel 24. In this latter case, the second stage vessel is at a lower pressure than the first stage vessel. Variations such as the foregoing may be employed in various embodiments without departing from the invention.

Similarly, the first stage separation vessel 24, the intercooler 23, the second stage separation vessel 27, and the subcooler 19 are each shown as separate components, whereas the heat exchanger 17 is shown as a single unit. Other arrangements of these functional elements are possible within the scope of the invention in which, for example, the subcooler and second stage vessel may be advantageously combined in a single unit; the intercooler and the first stage vessel likewise

may be integrated into a single unit; whereas the heat exchanger may be comprised of separate primary and secondary heat exchangers in which two streams of feed gas mixture are separately cooled by the hydrocarbon-rich first product stream and nitrogen-rich second product stream, respectively. Each of these foregoing illustrative variations is depicted in the embodiment of FIG. 3 and described in more detail in relation thereto.

With reference to FIG. 3, a preferred embodiment of the invention is depicted showing some detail of construction of the various elements as well as additional valves and lines for effecting control of the process.

The system of FIG. 3 shows a feed conditioning system similar to that of FIG. 1. Thus, as previously described in reference to FIG. 1, feed gas mixture produced at the wellhead 59 is passed successively through separator 61, regulator 62, chiller 63, filter-separator 64, and dryer system 65 comprising feed conditioning system 60. Thereafter, conditioned feed gas mixture is introduced into the low temperature portions of the process system. It should be pointed out that although the feed conditioning steps are not a part of the invention, the design, arrangement, and practical operation of the process depends upon the manner and success with which the conditioning is performed. The feed conditioning steps shown in the Figures are not to be thought of as the only possible system; indeed the presence of the other contaminants such as hydrogen sulfide; carbon dioxide, and natural gasoline require additional special equipment in the conditioning system for their removal. The conditioning steps shown are only typical of some of the more common, simple applications.

In FIG. 3, liquids accumulated in separator 61 are removed through drain valve 93; provision is made for cleaning filter-separator 64 by closing valves 95 and 96 while bypassing feed through valve 97. Liquids such as water and light oil accumulated in filter-separator 64 may be drained periodically through valve 98.

The dryer system 65 consists typically of a set of two or more desiccant or molecular sieve-type dryer vessels shown at 103 and 104, associated valves 99, 100, 101, 102, 105, 106, 107 and 108, and a heater 109. The valves are operated such that while one dryer dehydrates the feed gas flowing through the system, the other dryer is regenerated by a reverse flow of hot, dry nitrogen-rich gas from the process. Powdered material entrained in the gas stream as it flows through the dryer bed is removed from the conditioned feed gas by means of a particle filter 110 which can be cleaned periodically by closing valves 111 and 112 and bypassing the feed gas through valve 113.

Conditioned feed gas enters the process system proper through line 66 and shutoff valve 114. The feed gas stream is cooled in a pair of heat exchangers 67, consisting of primary heat exchanger 115 and secondary heat exchanger 116. The division of feed gas mixture between primary and secondary heat exchanger is controlled by balancing valves 117 and 118 in the feed mixture lines at the inlet of each heat exchanger. The heat exchangers are typically coiled tube-and-shell type construction, commonly referred to as Giauque-Hampson type. The process is amenable also to other forms of heat exchangers.

For the case illustrated in Table I, approximately 56 percent of the feed mixture is cooled in the primary heat exchanger and the balance in the secondary. This



proportion affords optimal utilization of the two product streams used as the cooling media in each heat exchanger, the two cold feed streams leaving each exchanger at about equal temperatures. A bypass valve 119 is provided to permit control of the final temperature of the cold feed mixture in line 68.

The cold feed mixture is further cooled in subcooler 69, installed within the second stage separation vessel 77. The subcooler comprises high pressure passages, which are typically coils of tubing indicated at 150, arranged for counterflow heat exchange with upflowing boiling liquid in a low pressure shroud 151. The shroud has an inlet pipe 152 for liquid from the first stage and is open at the top to permit escape of warmed liquid 76 and vapor boil-off into the second stage separation vessel 77. A spray baffle 149 minimizes entrainment of liquid in the vapor phase.

Bypass valve 120 in conjunction with valve 121 permits controlled bypassing of the cooled feed mixture around the subcooler for particular purposes such as starting or control of the subcooled feed temperature.

The subcooled feed passes through line 70 to expander 71, in this case a Joule-Thompson expansion valve. The expanded feed then passes through line 72 to intercooler 73 and thence through pipe 126 to first stage separation vessel 74. In the embodiment of FIG. 3, intercooler 73 consists of a mixing tube. The high velocity of the expanded feed mixture in line 72 is employed by means of a nozzle 122 directed concentrically toward the throat of a venturi 124. The second interstage stream, consisting of gas withdrawn from the second stage separation vessel through line 78, is entrained from a chamber 123 by the high velocity feed stream issuing from the nozzle and, as the two streams mix in the venturi, the entrained second interstage stream gains momentum. This momentum is converted into additional pressure of the combined stream in a diffuser 125. This device is referred to as an eductor pump and has the effect, as explained above, of pressurizing the second interstage stream before passing into the first separation vessel. Consequently, the second separation vessel is maintained at a lower pressure than the first. It should be noted that the slightly lower pressure of the second separation vessel is opposite to the system of FIG. 1 in which a liquid head maintains the second separation vessel at slightly higher pressure than the first separation vessel.

The heat exchange between the second interstage stream and the expanded feed mixture takes place by direct mixing in the diffuser of the eductor pump and in the downstream mixing tube (intercooler) 126.

The mixed two-phase stream is introduced into the first stage separation vessel 74 by a distributor 128. The mixture separates into a single hydrocarbon-rich liquid phase 129 and a single nitrogen-rich gaseous phase occupying the space above the liquid 127. The composition of these two phases is governed by the temperature of the separation and, accordingly, by the cooling of the feed stream in the heat exchangers and subcooler.

The embodiment of FIG. 3 depicts a particular arrangement of the distributor 128 designed to facilitate the separation of gaseous and liquid phases entering the vessel. The mixed two-phase stream from the mixing tube 126 enters the annular space between the distributor shroud 154 and the vapor tube 155 below the inlet 156 of the vapor tube. The vapor phase rises to enter the vapor tube while the liquid falls to the bottom of the

annular space. Separation may be improved by introducing the mixture eccentrically with respect to the distributor shroud in order to cause swirling flow. Liquid flows from the bottom of the annular space into the lower portion of the vessel 74 through drain tubes 157, while vapor entering the vessel from the bottom of the vapor tube is deflected horizontally so as not to impinge directly on the liquid surface by a deflector plate 158, through which the drain tubes pass.

It is important that the amount of liquid phase in the first stage vessel which may be entrained with the gas phase withdrawn from the vessel be held to a minimum. For this reason, a mist-extractor section 130 employing a suitable packing material 136 is contained between screens 137 and 138 near the top of the first vessel such that a mixture removal of entrained liquid from the nitrogen-rich gas stream is effected. The nitrogen-rich second product is withdrawn from the first stage vessel through the dome 159 containing the distributor and through line 85, warmed in the secondary heat exchanger 116 and discharged through line 86, back pressure regulator 87 and line 88 to atmosphere or other use as a by-product. Nitrogen-rich gas is available for purging the dryers through valve 131, line 89, heater 109, and valves 107 or 108 with exhaust through valves 101 or 102. Also, nitrogen-rich gas at a temperature below that of the feed mixture is available for cooling feed mixture in the chiller 63 by flowing through valves 132 and 133 in lines 91 and 92, valve 90 being closed for this purpose.

The hydrocarbon-rich liquid phase 129 in the first stage separation zone is withdrawn as the first interstage stream through line 75, at a rate controlled by valve 79, and into shroud 151 enclosing the subcooler coils 150. The liquid stream passing over the coils is partially boiled by heat exchange with the relatively warmer feed mixture in the coils and finally leaves the subcooler at 26 through an opening in the top of the shroud. Splash guard 149 prevents excessive entrainment of liquid in the gaseous phase separated from the warm liquid in the vessel and withdrawn as the second interstage stream through line 78, as described previously.

The operation of the process can be controlled to vary the product purity within limited ranges by changing the temperature at which the second stage separation is effected at a given pressure, a relatively small increase in temperature serving to increase the purity of the liquid product as can be seen by studying the curves of FIG. 2. To increase product purity above the nominal level requires more heating of the liquid from the first stage as it passes through the subcooler. One practical method of providing more heat in the subcooler is to increase the temperature of the cooled feed mixture entering the subcooler, either by bypassing a portion of one or both of the product streams around the heat exchangers. One method of controlling the temperature of the cold feed mixture is to bypass a portion of the nitrogen-rich second product stream through valve 135 to the discharge line 86.

The hydrocarbon-rich first product stream derived from the liquid phase 80 in the second stage separation vessel is withdrawn through line 81 to product pump 82. Alternately, for special purposes such as starting the process, liquid may be drawn directly from the first stage vessel through line 75 and valve 139 into the



product pump. If desired to retain liquid in the second stage vessel, valve 140 may also be closed.

If the product pump produces a pulsating output such as is the case with a single or multicylinder reciprocating unit, an accumulator 141 is provided to dampen the pulsations. The stream of first product liquid, pressurized in excess of the required delivery pressure by the pump, is passed through line 83, warmed and vaporized in the primary heat exchanger 115, and discharged through product valve 143 to the delivery line at 84 or a back pressure regulator 142 through to flare stack 94 in the event of over-pressure or in the event that closure of discharge valve 43 becomes necessary.

A portion of the product pump discharge may be bypassed through valve 145 and line 146 back into the second stage separation vessel to afford one method of controlling the net flow of liquid pumped from the process system. Another equivalent method is to controllably vary the pump speed.

A small sample of the first product can be periodically or continuously withdrawn at 147 for analysis in composition analyzer 148. The latter may be a comparative infrared absorption device which continuously monitors the composition of the hydrocarbon-rich product and provides an output signal which can be used for control purposes. Alternate analysis methods such as flame pyrometer-based calorimeters may be used to directly monitor product heating valve.

Under most conditions of operation, excess refrigeration is available and, in order to control the hydrocarbon-rich product purity, cold nitrogen-rich vapor is bypassed around secondary heat exchanger 116 through valve 135, as explained previously. When a requirement exists for liquefied natural gas, this excess cooling permits withdrawal of liquid from the second stage separation vessel through valve 152 and line 153. The rate of liquid withdrawal can be used to control product purity by analogy to the method of bypassing first stage vapor as described, or else liquid may be withdrawn as required while control of purity continues to be effected by means of bypassing cold first stage vapor.

Another important feature of the process necessary to achieve the substantial condensation of the feed stream is the manner of using the fluid product streams as heat exchange media for the feed stream. To this end, it is important to take advantage of the greatest possible cooling capability of both product streams. This result is achieved in one embodiment of the invention by use of parallel countercurrent heat exchangers.

It will be appreciated that instruments, automatic controls, and other accessory equipment for maintenance may be incorporated in the system.

The object of this and present conventional processes is to make available in usable form the latent fuel energy of the hydrocarbon. The measure of the effectiveness of such processes is the energy made available over and above that consumed in the operation of the process, i.e., the net energy production. We have discovered that by carrying out our process with the disciplines as set forth, the net energy production is substantially equal to that of much more complicated fractionation processes entailing much greater capital expenditures.

The specific features of embodiments of the process of the present invention have been set forth merely by way of example to illustrate forms in which the invention may be practiced.

What is claimed is:

1. A process for separating a high pressure normally gaseous natural gas mixture comprising predominantly nitrogen and methane into a first product stream containing a substantially higher proportion of methane than the mixture and a second product stream containing a substantially higher proportion of nitrogen than the mixture, the process comprising:

- a. cooling the natural gas mixture by passing the mixture in heat exchange relation to the first and second product streams in heat exchanger means,
  - b. subcooling and at least partially liquefying the cooled natural gas mixture by passing at least a portion of the cooled mixture in heat exchange relation to a first interstage stream in subcooler means,
  - c. expanding the subcooled natural gas mixture to a first reduced pressure,
  - d. utilizing a portion of the cooling capacity of the expanded subcooled natural gas mixture to cool a second interstage stream by passing at least a portion of the expanded subcooled natural gas mixture in heat exchange relation to a second interstage stream in intercooler means,
  - e. introducing the resultant partially warmed natural gas mixture and the cooled second interstage stream into a first stage separation zone to produce in the separation zone a single methane-rich liquid phase and a single nitrogen-rich gaseous phase in approximate thermal equilibrium at substantially said first reduced pressure,
  - f. continuously and separately withdrawing from the first stage separation zone separate streams of the methane-rich liquid phase as the first interstage stream and the nitrogen-rich gaseous phase as the second product stream,
  - g. partially vaporizing the first interstage stream by passing at least a portion of the stream in heat exchange relation to the cooled natural gas mixture in the subcooler means as recited in step (b),
  - h. introducing the partially vaporized first interstage stream into a second stage separation zone to produce in the zone in a single methane-rich liquid phase, richer in methane than the first interstage stream, and a single nitrogen-rich gaseous phase in approximate thermal equilibrium at a second reduced pressure,
  - i. continuously and separately withdrawing from the second stage separation zone separate streams of the methane-rich liquid phase as the first product stream and the nitrogen-rich gaseous phase as the second interstage stream,
  - j. partially condensing the second interstage stream by passing at least a portion of the stream in heat exchange relation to the expanded mixture in intercooler means as recited in step (d), the partially condensed second interstage stream being introduced into the first stage separation zone as recited in step (e),
  - k. separately passing at least a portion of the second product stream withdrawn from the first stage separation zone as recited in step (f) and at least a portion of the first product stream withdrawn from the second stage separation zone as recited in step (i) in heat exchange relation to the natural gas mixture in heat exchanger means as recited in step (a).
2. A process in accordance with claim 1 in which the portion of the first product stream which is passed in



heat exchange relation to the natural gas mixture as recited in step (k) of claim 1 is repressurized to a relatively higher pressure in the liquid phase prior to passage through the heat exchanger means.

3. A process in accordance with claim 1 in which the partial condensation of the second interstage stream as recited in step (j) of said claim 1 is accomplished by direct mixing of at least a portion of said second interstage stream with the expanded subcooled natural gas mixture referred to in step (d) of said claim 1.

4. A process in accordance with claim 1 in which step (a) thereof is carried out by passing separate portions of the natural gas mixture through parallel heat exchanger means in separate heat exchange relation with at least a portion of the first and second product streams passed separately through said parallel heat exchanger means.

5. A process in accordance with claim 4 in which that portion of the first product stream passed through the heat exchanger means in heat exchange relation to a portion of the natural gas mixture is repressurized to a relatively higher pressure in the liquid phase prior to its passage through said heat exchanger means.

6. A process in accordance with claim 5 in which the partial condensation of the second interstage stream as recited in step (j) in claim 1 is accomplished by direct mixing of at least a portion of said second interstage stream with the expanded subcooled natural gas mixture referred to in step (d) of claim 1.

7. A process in accordance with claim 1 in which the temperature in the first stage separation zone is maintained at a predetermined level so as to achieve a controlled predetermined composition of the first product stream.

8. A process in accordance with claim 1 in which the temperature in the second stage separation zone is maintained at a predetermined level so as to achieve a controlled predetermined composition of the second product stream.

9. A process in accordance with claim 1 in which the partially warmed natural gas mixture and the cooled second interstage stream referred to in step (e) of said claim 1 are subjected to a partial separation of liquid and gaseous phases thereof prior to discharge into said first stage separation zone.

10. A process in accordance with claim 1 in which the expansion of the subcooled natural gas mixture referred to in step (c) of said claim 1 is carried out so as to produce external work.

11. A process in accordance with claim 1 in which a portion of the liquid phase from the second stage separation zone is removed as a liquid product and the balance of the liquid phase removed from the second stage separation zone is used for the cooling step referred to in step (k) of said claim 1.

12. A process in accordance with claim 1 in which the expansion of the subcooled natural gas mixture recited in step (c) of said claim 1 is carried out with the production of external work and a portion of the liquid phase in the second stage separation zone is removed as a liquid product and the balance of the liquid phase removed from the second stage separation zone is used in the heat exchange step referred to in step (k) of said claim 1.

13. Apparatus for separating a high pressure normally gaseous natural gas mixture comprising predominantly nitrogen and methane into a first product stream containing a substantially higher proportion of methane than the mixture and a second product stream containing a substantially higher proportion of nitrogen than the mixture; the apparatus comprising:

- a. heat exchanger means,
- b. means for passing the natural gas mixture through the heat exchanger means in heat exchange relation to a first and second product stream,
- c. subcooler means and means for passing at least a portion of the cooled mixture in heat exchange relation to a first interstage stream in the subcooler means so as to subcool and at least partially liquefy the cool natural gas mixture,
- d. means for expanding the subcooled natural gas mixture to a first reduced pressure,
- e. intercooler means and means for passing at least a portion of the expanded subcooled natural gas mixture in heat exchange relation to a second interstage stream in the intercooler means,
- f. first stage separation vessel means and means for introducing the partially warmed natural gas mixture from the intercooler means and the cooled second interstage stream into the first stage separation vessel means to produce therein a single methane-rich liquid phase and a single nitrogen-rich gaseous phase in approximate thermal equilibrium at substantially said first reduced pressure,
- g. means for withdrawing from the first stage separation vessel means separate streams of the methane liquid phase as the first interstage stream and the nitrogen-rich gaseous phase as the second product stream and for introducing said first interstage stream into the subcooler means recited in (c) in heat exchange relation to the cooled natural gas mixture,
- h. second stage separation vessel means and means for introducing the partially vaporized first interstage stream into the second stage separation vessel means to produce therein a single methane-rich liquid phase richer in methane than the first interstage stream and a single nitrogen-rich gaseous phase in approximate thermal equilibrium at a second reduced pressure,
- i. means for continuously and separately withdrawing from the second stage separation vessel means separate streams of the methane-rich liquid phase as the first product stream and the nitrogen-rich gaseous phase as the second interstage stream,
- j. means for passing at least a portion of the second interstage stream in heat exchange relation to the expanded mixture in the intercooler means recited in (e),
- k. means for introducing the partially condensed second interstage stream into the first stage separation vessel means as recited in (f), and
- l. means for passing at least a portion of the second product stream and at least a portion of the first product stream in heat exchange relation to the natural gas mixture in the heat exchanger means recited in (a).

\* \* \* \* \*



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UNITED STATES PATENT OFFICE  
CERTIFICATE OF CORRECTION

Patent No. 3,791,157 Dated February 12, 1974

Inventor(s) Richard R. Tracy, Reginald G. Spear

It is certified that error appears in the above-identified patent and that said Letters Patent are hereby corrected as shown below:

Column 10, line 39 "26" should be -- 78 --

Sheets 2 and 3 of the Drawings should appear as attached hereto.

Signed and Sealed this  
twenty-fourth Day of February 1976

[SEAL]

Attest:

RUTH C. MASON  
Attesting Officer

C. MARSHALL DANN  
Commissioner of Patents and Trademarks



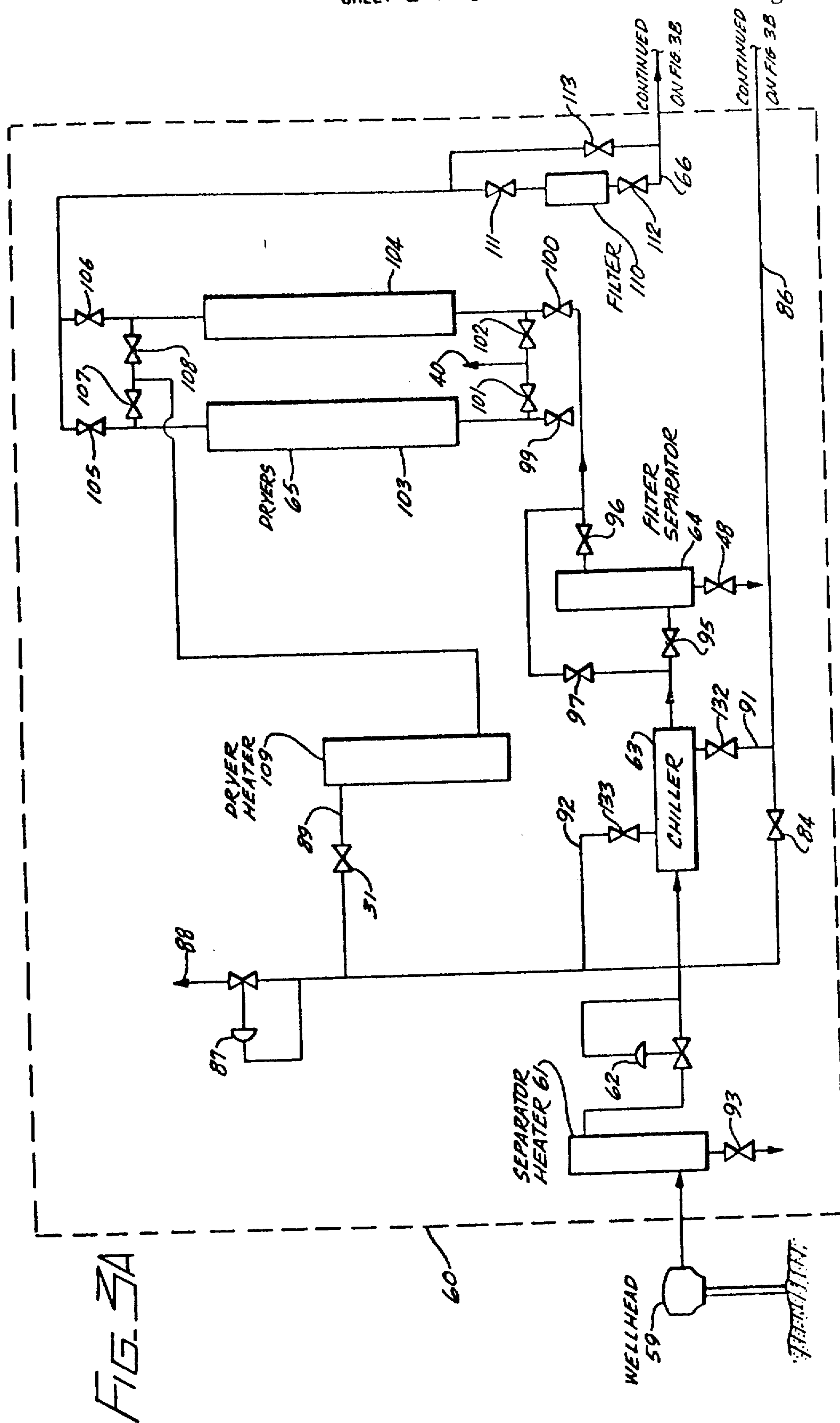


FIG. 3A



FIG. 3B

