ABSTRACT OF THE DISCLOSURE

A process is disclosed for recovering liquid products from a gaseous mixture. The mixture is cooled initially by countercurrent heat exchange with separated constituents of preceding portions of the same mixture and a recycled refrigerant. This initial cooling condenses some of the constituents. The liquid product thus formed is separated from the gas. The gas is then expanded to a lower pressure while doing mechanical work to cool it to a still lower temperature and condense more liquid products, which are combined with the first liquid products obtained to provide a cold liquid product stream and a cold gas stream. The cold product stream and a cold liquid refrigerant stream are combined and passed in heat exchange relationship with the gaseous feed stream to provide the initial cooling described above by partially vaporizing the combined liquid product and refrigerant stream. The vaporized portion of the combined liquid stream is separated from the remaining liquid and compressed with the power from the expanding of the gaseous feed stream as described above. The compressed vapor is cooled to condense some of it at substantially ambient temperature and to remove the latent heat of vaporization it had picked up in the heat exchanger. The partially condensed stream is recycled through the heat exchanger to condense additional vapor and thereby provide a liquid stream. This liquid stream is expanded, combined with the liquid product stream and passed through the heat exchanger as a refrigerant to cool the incoming feed stream.

This application is a continuation-in-part of our prior application Ser. No. 445,113, filed Apr. 2, 1965, now abandoned.

This invention relates to improvements in processes for the recovery of liquid products from a gaseous mixture. It is particularly useful in the recovery of ethane and heavier hydrocarbons from a stream of natural gas, but is not limited to such use.

In conventional cryogenic processes for the separation of a liquid product from a gaseous mixture it is customary to pass a feed stream of the gaseous mixture through indirect heat exchange with cold products previously separated from the gaseous mixture to cool the feed stream to or near to condensation temperature of its heaviest condensible components. The feed stream, or a portion of it, is then expanded through an expansion engine to cool it further. The refrigeration generated in the expansion engine has been used to condense a portion of the expanded stream or a side stream of the unexpanded gas mixture by indirect heat exchange, and the liquid condensate obtained is often subjected to one of many fractional separation treatments for further resolution of the mixture. The separated cold constituents usually are returned through indirect heat exchange with the incoming stream of gaseous mixture and are then withdrawn as products.

Such processes require large quantities of low temperature refrigeration to absorb the latent heat of vaporization of the product. If the cold liquid product stream is used to absorb this latent heat of vaporization of the incoming feed stream, not only will all of the lighter and more volatile hydrocarbons in the product stream be vaporized, but most of the heavier hydrocarbons as well.

It is an object of this invention to provide a process for the recovery of a liquid product from a gaseous mixture in which very little vaporization of the liquid product occurs as it passes in heat exchange relationship with the incoming gas stream.

It is another object of this invention to provide a process for the recovery of a liquid product from a gaseous mixture in which a refrigerant comprised of the lighter hydrocarbons in accumulated in the system and recycled with the cold liquid product obtained from the feed stream to vaporize and absorb the latent heat of vaporization of the condensable constituents of the feed stream.

Another object is to provide a process of the above type in which expansion of the gas furnishes all the power for the low temperature refrigeration required.

Another object is to provide a process of the above type wherein a large part of the vaporization latent heat of the product is removed at substantially ambient atmospheric or similar readily maintainable temperature.

Another object is to provide a process of the above type which results in a liquid product low in gaseous constituents.

Another object is to provide a process of the above type in which the apparatus used may be simple in construction.

Highest efficiency in operation of a process involving a heat exchanger through which streams of gases and liquids are passed countercurrently is known to require that the temperature differential between countercurrent streams be small; and if the heat exchanger is not to be excessively large, this temperature difference should be substantially uniform at all locations along their countercurrent paths in the heat exchanger, that is, the temperatures of the streams should not converge or diverge as they approach the warm or cold end of the heat exchanger. Lack of reasonably uniform temperature differential has increased the operating cost of prior processes for recovery of liquid from gaseous mixtures.

Therefore, another object is to provide a process of the above type in which a main heat exchanger may be used for cooling an incoming feed stream of a gaseous mixture by product streams in which substantially uniform temperature differential between the streams at consecutive locations in the heat exchanger is easily maintained.

Another object is to provide a process of the above type in which temperature differences between the feed stream of gaseous mixture and product streams are controllable to prevent the temperatures of the streams from converging or diverging.

Another object is to provide such process in which a single expander and a single compressor may be used, and in which the flash gas is returned to the process.

Still another object is to provide such process which is inexpensive in operation, and which can be made substantially automatic.

Another object is to provide a process utilizing an expansion engine loaded with a compressor in which a portion of the compression energy acts to operate a refrigeration system based on liquefiable components in the compressed steam to augment the refrigeration for carrying out the process.

Other objects and advantages will become apparent.
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3. to those skilled in the art from consideration of the following detailed description and the attached drawings.

In the attached drawing the single figure is a schematic flow diagram of a process involving the principles of my invention.

The following detailed description is directed to a process for recovering light and higher hydrocarbons from a feed stream of natural gas containing the same, but it is to be understood that it is applicable to a great many gaseous mixtures which contain components that are liquefied on cooling and application of pressure.

Referring to the drawing, an incoming feed stream of a gaseous mixture containing separable components, as, for example, a stream of natural gas containing ethane and/or higher hydrocarbons, such as propane and/or butane, enters under pressure through line 1 at a rate of flow controlled by valve 2 and passes through pass b in main heat exchanger 3 where it is cooled to a lower temperature, and is preferably partially condensed to produce a large portion of crude liquid products. When the stream is natural gas containing ethane and higher hydrocarbons, it may enter exchanger 3 at substantially well pressure, if desired.

The partially condensed feed stream is withdrawn from heat exchanger 3 by line 4 and is discharged into separator 5 where the liquid condensed in the heat exchanger is disengaged from residual gas. A portion of the disengaged gas may be returned through line 6 and pass e through the heat exchanger 3 and discharged as a dry gaseous product through line 7. If there is more gas being supplied to the system than is required to supply refrigeration and power to the system by expansion as described below. Frequently, the return of gas through line 6 and heat exchanger pass e is not necessary.

The remainder of the gas separated in a separator 5 is passed through a turboexpander 9 illustrated as turboexpander 9. The gaseous stream is further cooled, additional liquid is condensed therefrom, and power is generated by expanding the gaseous stream to a lower pressure through the turboexpander. The liquid condensed in pass b of heat exchanger 3 and separated from the gas in separator 5, is withdrawn from the separator and introduced into a cold gas stream coming from a later described separator 16. This cold gas stream is rich in constituents to be recovered as liquid.

The resulting combined two phase stream flows through conduit 17 and heat exchanger 18 where it is cooled still further, by which it is introduced into the low pressure and very cold effluent from the turboexpander 9 flowing through line 12 to separator 13, where the mixed liquid is disengaged from residual gas. The liquid recovered in separator 13 is the crude liquid product and is at a very low temperature and at approximately the low pressure of the expanded effluent from the turboexpander 9.

The residual gas withdrawn from separator 13 is the combined residue gas stream and is likewise at the very low temperature and low pressure of the effluent from said turboexpander. This stream flows out through line 14 which returns it to pass e of heat exchanger 3 where its refrigeration content is recovered by cooling the incoming stream of feed gas. The residual gas stream is withdrawn as a dry gas product through line 15.

The crude liquid product in separator 13 is withdrawn by line and is passed through heat exchanger 18 where its temperature is raised somewhat. This results in vaporization of some of the more volatile components of the mixture, as for example methane and part of the ethane from a natural gas mixture, and at least partially strips the liquid of such materials. The resulting two phase mixture passes through separator 21 where the liquid and vapor are separated.

The liquid product that has been recovered from the gas flowing into the system is combined with a liquid stream of refrigerant flowing through line 16 from separa-

rator 16 and the combined stream of liquid products and refrigerant flows through pass c of heat exchanger 3. As will be further explained below, the refrigerant employed comprises mostly lighter hydrocarbons that have been recovered and accumulated from the liquid products initially produced by the process and thus these lighter ends will carry the liquid product, which contains a much higher percentage of heavier hydrocarbons. Thus, as the combined streams of refrigerant and liquid product pass through heat exchanger 3, substantially all the refrigerant will vaporize as it absorbs heat from the incoming feed stream of gas. Nevertheless, the lighter ends of the liquid product may vaporize also, but this is held to a minimum. As the combined streams leave the heat exchanger and pass into separator 28 through line 27, the liquid product will still be substantially all liquid. The liquid product is separated from the vaporized refrigerant in separator 28 and collected in liquid product tank 32 through line 41.

The refrigerant, which was substantially all vaporized as it passed through the heat exchanger, will leave separator 28 as a gas through line 29, join the vapors separated from the liquid product in separator 21, and travel through line 33 to the inlet of compressor 34. The way this stream will also be joined by the vapors coming off storage tank 32. Pressure regulator 25 in line 33 maintains a given pressure on separator 28 and the liquid product and refrigerant as they pass through the heat exchanger. By controlling the pressure in the heat exchanger, the amount of liquid that is vaporized therein can be controlled and thus the pressure regulator acts as a control of the temperature in the heat exchanger.

Vapors flowing through line 33 are compressed by compressor 23, which is powered by turboexpander 9. After being compressed to a relatively high pressure, the vapors are cooled in condenser 34 which may be cooled in any convenient fashion, as by water from a cooling tower which is introduced to the condenser through line 36 and withdrawn through line 37. Compressor 23 raises the pressure of the vapors sufficiently that a substantial portion of the vapors will condense in condenser 35 at substantially ambient temperature. Thus, the heat of vaporization absorbed by the refrigerant, as it is vaporized in heat exchanger 3, is removed from the process by a conventional cooling tower operating at or around ambient temperatures. This is a very economical way to remove this heat from the incoming gas stream.

The combined liquid and vapor coming out of condenser 35 passes through separator 35a, where the liquid and vapors are separated. The vapor and liquid are still at relatively high pressures. The vapor separated from the liquid in separator 35a passes through line 35b, through back pressure valve 38, which maintains a given pressure on separator 35a, and then into line 39. The liquid in separator 35a travels through line 42, through back pressure regulator valve 43, and rejoins the vapor in line 39 before passing back through heat exchanger 3 through pass d of heat exchanger 3. Line 35c is also connected to separator 35a and permits a portion of the liquid separated in separator 35a to be drawn off through line 35c. This liquid is introduced into the liquid leg of separator 28 and returned to liquid product storage tank 31. This permits a control on the amount of refrigerant being circulated in the system and there may be periods when no liquid is being withdrawn from the separator 35a, particularly in the beginning when an inventory of refrigerant is being accumulated.

The refrigerant then flows through pass d of heat exchanger 3, is cooled by the refrigerant and liquid product stream flowing through pass e and the cold gas flowing through pass d. This condensate may be the case that when the refrigerant stream reaches separator 16, it may well be substantially all liquid. What remains as vapor is separated from the refrigerant at this point and passed back into the process through line 16b, where it
joins the liquid coming out of separator 5, is condensed subsequently by the cold effluent of turboexpander 9, and thus rejoins the liquid product stream. The liquid refrigerant is vaporized through the lower pressure of line 32, which further cools the liquid refrigerant before it joins the liquid product and returns through pass c of the heat exchanger.

Since the lighter hydrocarbons will more readily vaporize than the heavier, when the process is initially started, substantially all of the product will be vaporized in pass e and recycled back through the compressor, heat exchanger, separator 16, etc., and used as a refrigerant. As the process continues, an inventory of refrigerant will be accumulated until the system is balanced, with sufficient refrigerant being recycled to provide the cooling required. Further, during the accumulation of this refrigerant, less and less of the heavier hydrocarbons in the liquid product stream coming out of separator 21 will be vaporized in the heat exchanger. This results because the lighter hydrocarbon content of the refrigerant will increase until it consists of substantially all lighter hydrocarbons. Therefore, the refrigerant then will more readily vaporize and absorb the latent heat of vaporization that is given up by the incoming feed stream.

It is estimated that one half pound of refrigerant will be required for every two pounds of feed gas that is introduced into the system. Every two pounds of gas, however, will only produce about 5% of a pound of liquid product. From this it can be seen that the liquid product stream is relatively small compared to the refrigerant that is circulated through pass c. But with applicant's process of recombining the refrigerant and the liquid product upstream prior to being brought back to pass e of the heat exchanger, most of the liquid product is collected as liquid, in liquid product tank 32, which is what is desired.

Heat exchanger 35 is not restricted to cooling with cooling water or air. It might be a coil in the warm end of heat exchanger 3, 5, line 21, or elsewhere. It would still be advantageous to carry out the process if heat exchanger 35 is at a higher temperature than that at which the liquid is being evaporated, even though considerably lower than ambient atmospheric.

Another important feature of the process described above lies in its capability of being controlled in a way to adjust temperature differences in heat exchanger 3 to correct for either divergence or convergence of temperature difference. For example the vapor in the refrigerant stream entering pass d of the heat exchanger may condense at a temperature higher than that necessary to maintain a uniform temperature difference between the streams in the heat exchanger. This can be prevented by admitting a small amount of the incoming feed stream of gaseous mixture from line 1 through line 40 and valve 40a. The presence of a minor proportion of the gaseous mixture withdrawn from the feed stream in line 39 and pass d of the heat exchanger reduces the partial pressure of the condensing constituents in pass d so that they condense at a lower temperature, thereby liberating latent heat in pass d at a lower temperature and nearer the cold end of the heat exchanger, and adjusting the temperature difference divergence.

Under certain circumstances of feed gas composition the incoming feed stream of gaseous mixture may require further cooling after it passes through pass 6 of heat exchanger 3 and before going to the turboexpander 9. Under these circumstances it is preferred that the feed stream be passed through an auxiliary heat exchanger 46 in counter current heat exchange with cold gas from separator 13 flowing through line 14.

From the foregoing, it will be seen that this invention is one well adapted to attain all of the ends and objects hereinbefore set forth, together with other advantages which are obvious and which are inherent to the process and method.

It will be understood that certain features and sub-combinations are of utility and may be employed without reference to other features and sub-combinations. This is contemplated by and is within the scope of the claims.

As many possible embodiments may be made of the invention without departing from the scope thereof, it is to be understood that all matter herein set forth or shown in the accompanying drawing is to be interpreted as illustrative and not in a limiting sense.

The invention having been described, what is claimed is:

1. A process for separating a liquid product from a gaseous mixture at a relatively high pressure comprising the steps of separating a product stream from the gaseous mixture as a cold liquid leaving a gaseous residue, reducing the pressure of the product stream and combining the cold product stream with a cold stream of recycled liquid refrigerant, passing the combined liquid product and refrigerant stream through a heat exchanger to cool the incoming gaseous product stream by vaporizing a substantial amount of the refrigerant, and a portion of the product stream, separating the vapor so produced from the remaining liquid product, compressing the vapor, cooling the compressed vapor to condense a portion thereof, returning the vapor and condensate through the heat exchanger together to condense more of the vapor, separating the condensate and returning the pressure of the condensate to provide the cool stream of liquid refrigerant for combining with the product stream prior to passing the product stream through the heat exchanger.

2. The process of claim 1 further provided with the step of periodically removing a portion of the condensate produced when the vapor is initially cooled following compression to keep the amount of refrigerant being recycled substantially constant.

3. The process of claim 1 in which the cold product stream is obtained by initially cooling the gaseous stream by the cold product and refrigerant stream passing through the heat exchanger, separating the condensate that formed from the gaseous stream, work expanding the gaseous stream to further cool the gaseous stream to condense additional liquid therefrom and to generate power for compressing the vapor from the refrigerant mixing the liquid first condensed and separated from the gaseous stream with the liquid condensed from the work expansion and the expanded gas at the lower pressure, separating the gas from the liquid to provide a cold liquid product stream and a cold gas stream for passing through the heat exchanger.

4. The process of claim 3 in which the vapor remaining in the recycled refrigerant is separated therefrom and combined with the first condensed liquid before the liquid refrigerant is combined with the cold liquid product stream.

5. A process for recovery of a liquid product from a gaseous mixture containing a material which is liquefied on cooling comprising the steps of passing a feed stream of said gaseous mixture under a relatively high pressure through indirect heat exchange with separate streams of cold gas and liquid previously recovered from advance portions of said gaseous mixture; thereby cooling the feed stream sufficiently to condense liquid therefrom; separating condensed liquid from the gaseous stream; further cooling the separated gaseous stream to condense additional liquid therefrom and to generate power by expanding the gaseous stream to a lower pressure through an expansion engine; mixing the liquid first condensed, additional liquid condensed, and expanded gas at said lower pressure; separating the expanded gas and the mixed condensed liquid; returning streams of the cold expanded gas and mixed liquid separately in indirect heat exchange with later portions of said feed stream of gaseous mixture; maintaining the stream of mixed liquid at said lower pressure during said heat exchange; vaporizing at least a part
of the mixed liquid by absorbing heat from the feed stream of gaseous mixture; withdrawing the resulting vapor from heat exchange; compressing the vapor to a pressure higher than that at which it was evaporated with power generated by expansion of said gaseous stream in said expansion engine; recondensing at least a part of the compressed vapor to liquid at approximately ambient atmospheric temperature; withdrawing a large part of the resulting condensate as a liquid product and recycling the remainder into said stream of mixed liquid returning to said indirect heat exchange to act as a refrigerant in maintaining the temperature range of said indirect heat exchange at a desired value.

6. The process of claim 5 wherein the feed stream is a stream of natural gas containing hydrocarbons in the range of ethane, propane, butane and heavier hydrocarbons.

7. The process of claim 6 wherein the mixed liquid is warmed and partially vaporized by heat exchange with the liquid first condensed from the feed stream, the liquid product is held in a container, vapor is evolved from said liquid product, and prior to the compression step said last-mentioned evolved vapor is mixed with vapor of mixed liquid from heat exchange with the feed stream and with vapor from said partial vaporization of mixed liquid.

8. A process for recovery of a liquid product from a gaseous mixture containing a material which is liquefied on cooling comprising the steps of passing a feed stream of said gaseous mixture under a relatively high pressure through indirect heat exchange with separate streams of cold gas and liquid previously recovered from advance portions of said gaseous mixture; thereby cooling the feed stream sufficiently to condense liquid therefrom; separating condensate from the gaseous stream; further cooling the separated gaseous stream, condensing additional liquid therefrom and generating power by expanding the gaseous stream to a lower pressure through an expansion engine; mixing the liquid first condensed, additional liquid condensed, and expanded gas at said lower pressure; separating the expanded gas from the mixed condensed liquid; returning the stream of cold expanded gas separately to indirect heat exchange with later portions of said feed stream of gaseous mixture; passing the liquid first condensed through indirect heat exchange with said mixed condensed liquid prior to mixing the liquid first condensed with said additional liquid condensed and expanded gas and thereby partially vaporizing the stream of mixed liquid by such heat exchange, separating the resulting vapor from the mixed liquid, returning the resulting mixed liquid residue to indirect heat exchange with the feed stream of gaseous mixture; maintaining the stream of mixed liquid at said lower pressure during said heat exchange; vaporizing at least a part of the mixed liquid by absorbing heat from the feed stream of gaseous mixture in such heat exchange; withdrawing the resulting vapor from said heat exchange and mixing it with the vapor previously separated from said mixed liquids; compressing the vapor mixture so produced to a pressure higher than that at which it was evaporated with power generated by expansion of said gaseous stream in said expansion engine; recondensing at least a part of the compressed vapor to liquid at approximately ambient atmospheric temperature; and withdrawing a large part of the resulting condensate as a liquid product.

9. A process for separating a liquid product from a gaseous mixture at a relatively high pressure comprising the steps of separating a product stream from the gaseous mixture as a cold liquid leaving a gaseous residue; reducing the pressure and evaporating a portion of the cold liquid residue as it passes through a heat exchanger, thereby recovering its refrigeration content at low temperature; expanding the gaseous residue to a relatively low pressure through an expansion engine to generate power and to provide additional liquid product for the liquid product stream; compressing the portion of the cold liquid product stream that is evaporated as it passes through the heat exchanger to a pressure higher than said reduced lower pressure with the power generated by the expansion engine, thereby raising its condensing temperature; condensing part of the compressed portion to liquid at a temperature higher than that at which it was evaporated; separating said condensed liquid from said portion to provide a vapor recycle stream from said portion; passing said vapor recycle stream through said heat exchanger to be cooled by the liquid product stream to condense a portion of the vapor recycle stream; separating the condensed liquid from said vapor recycle stream after it passes through the heat exchanger, and combining said separated liquid with said cold liquid product stream before the cold liquid product stream enters the heat exchanger.

10. The process of claim 9 in which the said higher temperature at which the product is condensed is approximately ambient atmospheric temperature.

11. The process for recovery of a liquid product from a gaseous mixture containing a material which is liquefied on cooling comprising the steps of passing a feed stream of said gaseous mixture under a relatively high pressure through heat exchange with separate streams of cold gas and liquid previously recovered from treatment of advance portions of said gaseous mixture; thereby cooling the feed stream sufficiently to condense liquid therefrom at such relatively high pressure; separating condensed liquid from the gaseous stream, separating the liquid product stream of cold gas and liquid streams; condensing additional liquid for the cold liquid stream and generating power by expanding the gaseous stream from said relatively high pressure to a lower pressure through an expansion engine; returning the condensed liquid stream and expanded gaseous stream at such lower pressure to indirect heat exchange with later portions of the feed stream to produce said initial cooling of the feed stream; vaporizing a portion of the liquid stream at low temperature by said indirect heat exchange; compressing said vapor to a pressure higher than said lower pressure by power derived from expansion of said gaseous stream; condensing at least a part of the compressed vapor at approximately ambient atmospheric temperature; separating the condensate formed by said compression to provide a vapor stream; recycling said vapor stream in indirect heat exchange with said separate streams of cold liquid and gas initially obtained from the gaseous mixture to re-cool the vapor and condense a portion thereof thereby reclaiming at least a portion of the product previously vaporized when used as a refrigerant to cool the incoming feed stream, separating the condensed liquid product from said vapor recycle stream and returning said liquid from the recycled vapor to said stream of cold liquid to act again as a refrigerant to cool said incoming feed stream.

12. The process of claim 11 wherein the feed stream is a stream of natural gas containing hydrocarbons in the range of ethane, propane, butane and heavier hydrocarbons.

13. In a process for the recovery of a liquid product from a gaseous mixture containing a material liquefied by a cooling medium pressure in which a feed stream of the gaseous mixture is passed through simultaneous countercurrent indirect heat exchange with at least two cold product streams, at least one of said cold product streams being substantially liquid, with a portion of said liquid product stream being vaporized during said heat exchange to increase the heat absorbed from said feed stream by the latent heat of vaporization, and separating said portion of the liquid product stream that is vaporized, that improvement which comprises maintaining a more uniform temperature difference between the feed stream and the cold product streams as they flow countercurrently through the heat exchange by the steps consisting of separating a vapor recycle stream from the vaporized
portion of the liquid product stream downstream of said heat exchange; passing said vapor recycle stream concurrently with said feed stream through said heat exchange to cool said vapor recycle stream sufficiently to condense a portion thereof to thereby reduce the temperature difference between the cold product streams and the feed stream; separating the liquid from said cooled recycle stream, and returning said separated liquid to said liquid product stream upstream of the heat exchange.

14. The process of claim 13 including the steps of compressing said vaporized portion of the liquid product stream to a pressure wherein constituents therein are easily condensed at a temperature higher than the temperature in said heat exchange, cooling said compressed vapor stream to condense said constituents, and separating the condensate formed from said vapor stream to form said vapor recycle stream.

15. The process of claim 14 further including the step of re-combining a portion of the liquid condensed from said compressed vaporized portion with the vapor recycle stream to cause the condensation of the vapor to occur earlier in said heat exchange and closer to the inlet of the feed stream.

16. The process of claim 14 further including the step of combining a portion of the feed stream with said vapor recycle stream to cause the condensation of the vapor to occur later in said heat exchange and closer to the inlet of the cold product streams.

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