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(54) METHOD FOR ETHANE LIQUEFACTION

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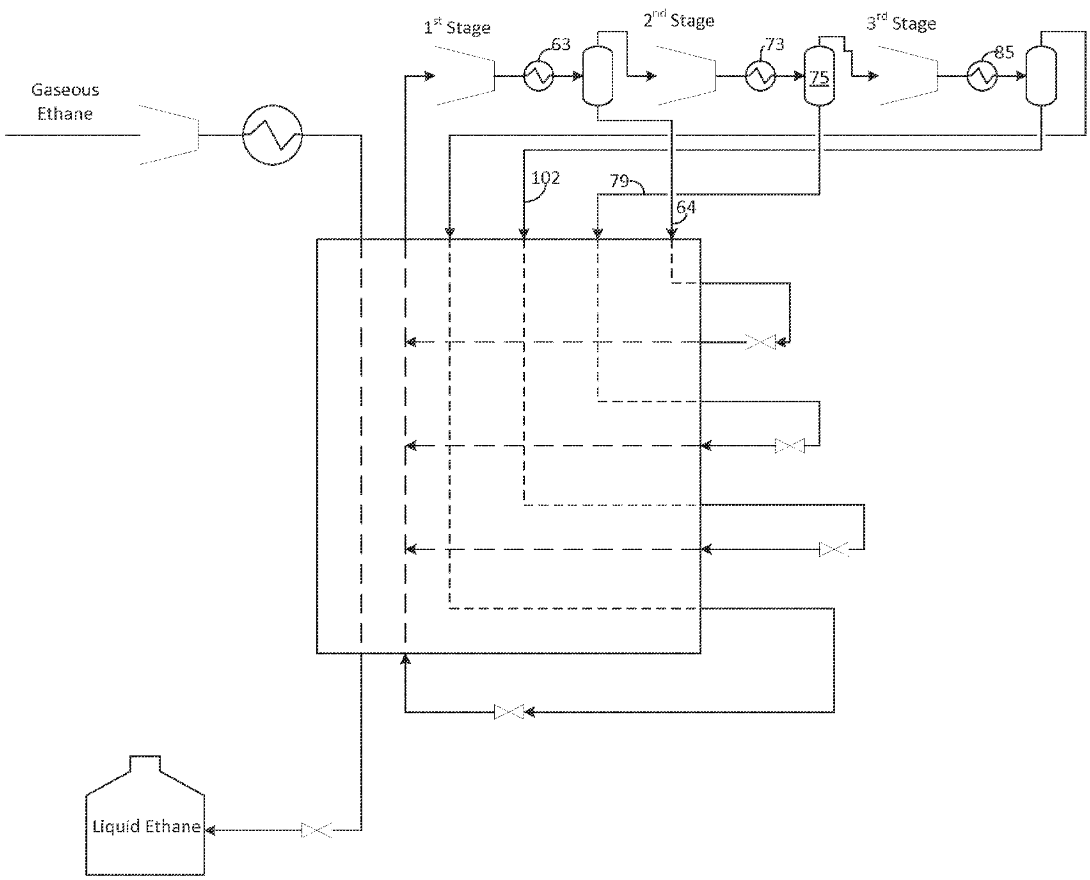
(57) **ABSTRACT**

The process for ethane liquefaction can include a mixed refrigerant containing heavy hydrocarbons (e.g., butane and/or pentane) using compression without interstage cooling, or cooling only after the first compression stages where there is no liquid formation yet, such that the number of liquid recycle loops are reduced. The lack of cooling in the compressor reduces the compressor's mechanical efficiency; however, this is offset by having a more thermodynamically efficient process cycle because the cycle can operate as a mixed refrigerant rather than cascade.



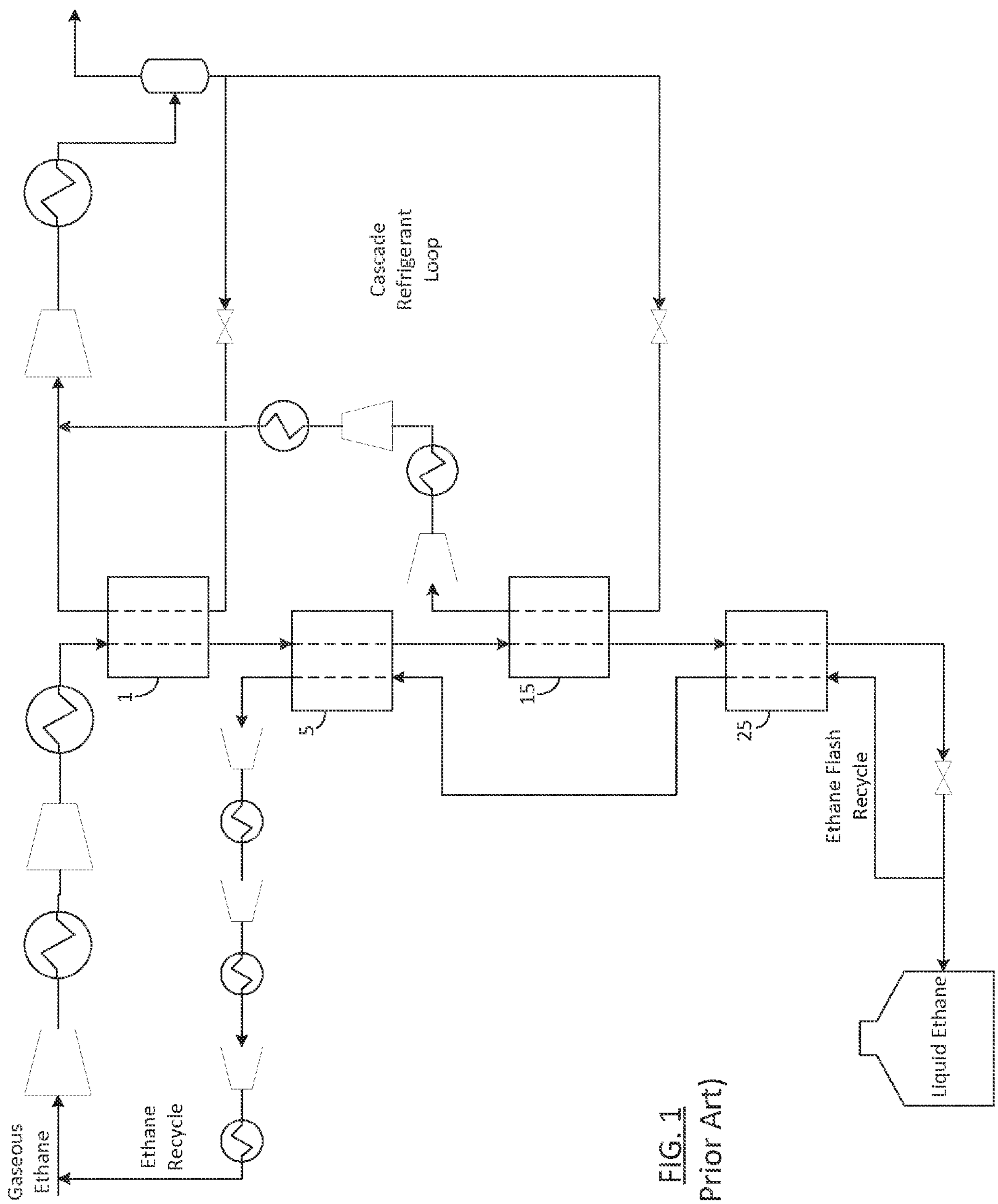
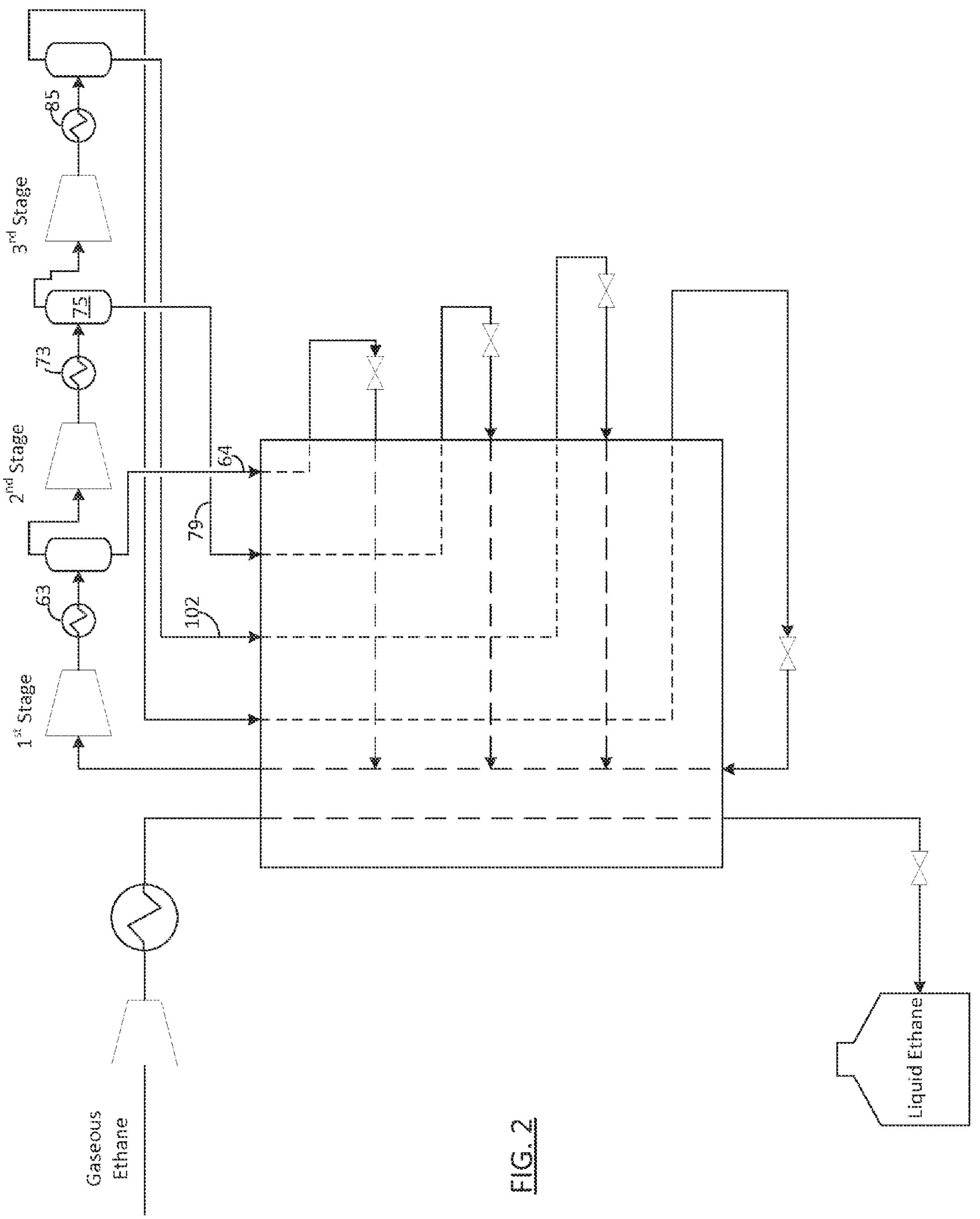


FIG. 1
(Prior Art)



1st Stage
2nd Stage
3rd Stage
63
73
75
85
64
79
102
Gaseous Ethane
Liquid Ethane

FIG. 2

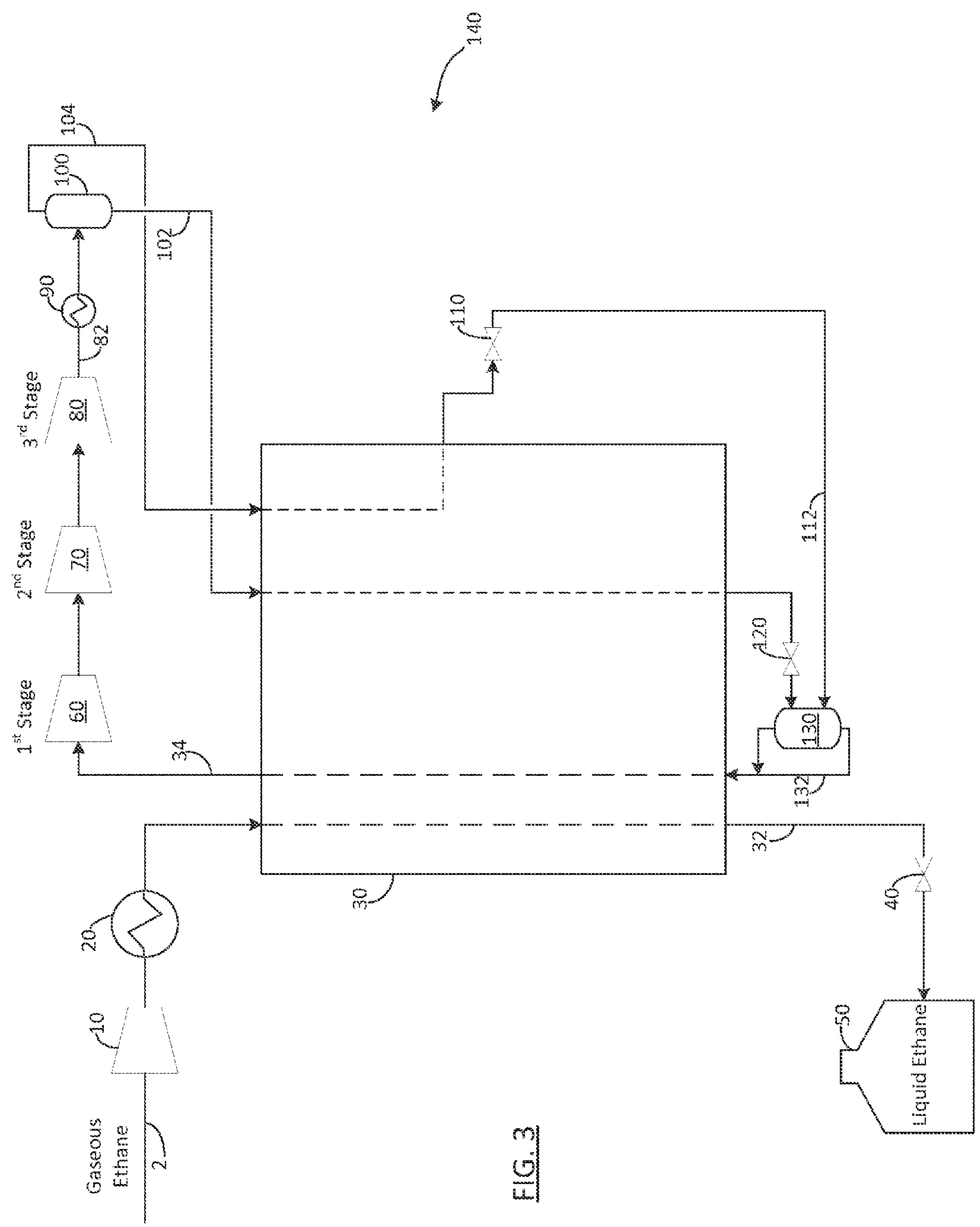

140
104
100
102
90
82
3rd Stage
80
2nd Stage
70
1st Stage
60
110
112
120
130
132
34
32
30
20
10
40
50
Liquid Ethane
Gaseous Ethane
2

FIG. 3

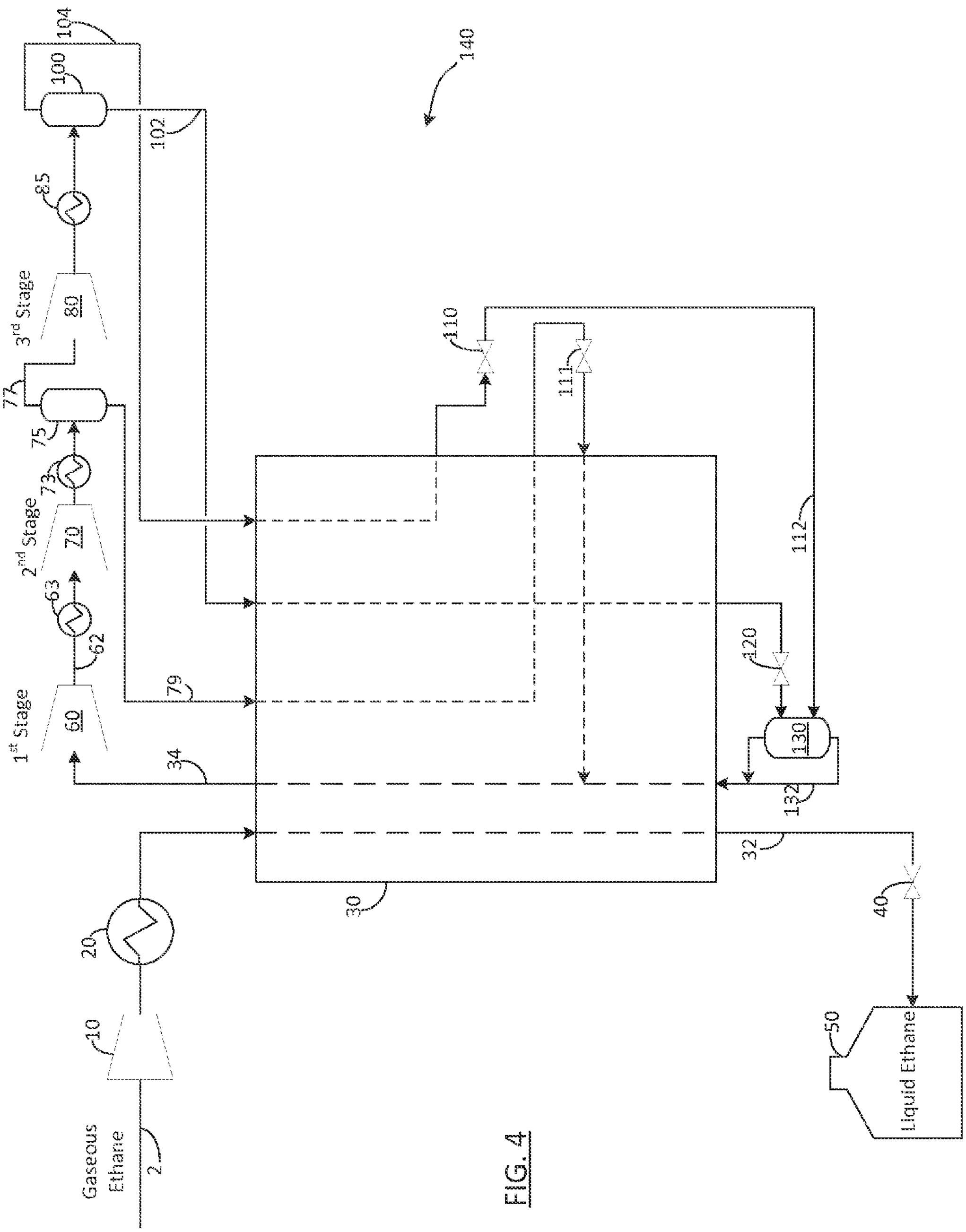

Gaseous Ethane
2
10
20
30
32
34
1st Stage
60
62
63
2nd Stage
70
73
75
77
3rd Stage
80
85
100
102
104
79
110
111
112
120
130
132
40
50
Liquid Ethane
140

FIG. 4

METHOD FOR ETHANE LIQUEFACTION

CROSS-REFERENCE TO RELATED APPLICATIONS

[0001] This application is a non-provisional application of U.S. Provisional Applicant No. 62/060,609, filed Oct. 7, 2014, which is herein incorporated by reference in its entirety.

TECHNICAL FIELD OF THE INVENTION

[0002] The present invention relates to an apparatus and method for liquefaction of ethane. More specifically, embodiments of the present invention are related to liquefying a gaseous stream comprised predominantly of ethane by using a mixed refrigerant loop incorporating heavy hydrocarbons (butane, and/or pentane) compression without providing cooling between stages or with limited cooling between stages.

BACKGROUND OF THE INVENTION

[0003] Liquefaction of methane (LNG) is well established, dating back to over 50 years. In certain cases, liquid ethane can also be produced directly from these LNG plants along with other higher hydrocarbon chain components and are called natural gas liquids (NGLs). However, many applications require the independent liquefaction of a gaseous ethane stream from a pipeline.

[0004] FIG. **1** shows a prior art ethane liquefaction process using a cascade refrigerant loop with a pure component such as propane to provide refrigeration at intermediate heat exchange section **15** and warm section **1**, while an ethane flash gas recycle provides cold end **25** and another intermediate level **5** cooling. However, because this process employs large temperature differences between the hot and cold fluids, brazed aluminum heat exchangers cannot be used as the exchangers would be subjected to very high thermal stresses. As such, the process known heretofore suffers several drawbacks, including using multiple, independent shell and tube type exchangers (or the like), suffering high irreversible losses, and having increased power and capital costs.

[0005] Therefore, it would be desirable to have an improved process for liquefaction of a gaseous stream comprised predominantly of ethane that was simple and efficient.

SUMMARY OF THE INVENTION

[0006] The present invention is directed to a process that satisfies at least one of these needs. In one embodiment, the process for ethane liquefaction can include using inline compression without interstage cooling (or cooling only after the first compression stages where there is no liquid formation yet) such that the number of liquid recycle loops are reduced or eliminated. The lack of cooling in the compressor slightly reduces the compressor efficiency; however, this is offset by having a more thermodynamically efficient process because the cycles can operate as a mixed refrigerant rather than cascade.

[0007] In one embodiment of the invention, a method is provided for the liquefaction of ethane. The method can include the steps of: (a) providing a stream of gaseous ethane under pressure, wherein the stream of gaseous ethane comprises at least 90% ethane; and (b) condensing the stream of gaseous ethane to produce liquid ethane by exchanging heat with a mixed refrigerant within a heat exchanger, wherein the mixed refrigerant is subjected to a mixed refrigerant refrigeration cycle.

[0008] In one embodiment, the mixed refrigerant refrigeration cycle includes the steps of: compressing the mixed refrigerant in a first compression section to produce a first compressed stream; compressing the first compressed stream in a second compression section to produce a second compressed stream; compressing the second compressed stream in a third compression section to produce a third compressed stream; cooling the third compressed stream to approximately ambient temperature to form a cooled third compressed stream, wherein the cooled third compressed stream is a two phase fluid; separating the cooled third compressed stream into a liquid refrigerant and a gas refrigerant; expanding the gas refrigerant to produce a cooled refrigerant; and introducing the cooled refrigerant to the heat exchanger under conditions effective for absorbing heat from the gaseous ethane such that the gaseous ethane condenses to form the liquid ethane.

[0009] In optional embodiments of the method for liquefaction of ethane:

- **[0010]** the mixed refrigerant comprises a heavy hydrocarbon selected from the group consisting of butane, pentane, and combinations thereof;
- **[0011]** the mixed refrigerant further comprises a hydrocarbon selected from the group consisting of methane, ethane, ethylene, propane, and combinations thereof;
- **[0012]** the gaseous ethane is at a pressure of at least 15 bara;
- **[0013]** the method can also include an absence of a cooling step between each of the three compressing steps;
- **[0014]** the method can also include an absence of a cooling step between the second and third compressing steps;
- **[0015]** the method can also include the step of cooling the first compressed stream prior to compressing said first compressed stream in the second compression section;
- **[0016]** during the step of cooling the first compressed stream, the first compressed stream is cooled to a temperature sufficiently warm enough to prevent formation of a liquid phase; and/or
- **[0017]** the method can also include the step of expanding the liquid refrigerant and combining the expanded liquid refrigerant with the cooled refrigerant prior to the step of introducing the cooled refrigerant to the heat exchanger.

[0018] In another embodiment of the invention, a method is provided for the liquefaction of ethane. The method can include the steps of: (a) providing a stream of gaseous ethane under pressure; and (b) condensing the stream of gaseous ethane to produce liquid ethane by exchanging heat with a mixed refrigerant within a heat exchanger, wherein the mixed refrigerant is subjected to a mixed refrigerant refrigeration cycle.

[0019] In one embodiment, the mixed refrigerant refrigeration cycle can include the steps of: compressing the mixed refrigerant to produce a first compressed stream; compressing the first compressed stream to produce a second compressed stream; compressing the second compressed stream to produce a third compressed stream; cooling the third compressed stream to approximately ambient temperature to form a cooled third compressed stream, wherein the cooled third compressed stream is a two phase fluid; separating the cooled third compressed stream into a liquid refrigerant and a gas refrigerant; expanding the gas refrigerant to produce a cooled refrigerant; and introducing the cooled refrigerant to the heat exchanger under conditions effective for absorbing heat from the gaseous ethane.

[0020] In one embodiment, the mixed refrigerant refrigeration cycle further includes an absence of a formation of a liquid phase of the mixed refrigerant at a point that is located both downstream the first compression step and upstream the final compression step. In another embodiment, there is an absence of a liquid/gas separation subsequent the first compression step and prior to the last compression step.

[0021] In another embodiment of the invention, an apparatus is provided for the liquefaction of ethane. In this embodiment, the apparatus can include: (a) a gaseous ethane source; (b) a heat exchanger configured to condense gaseous ethane received from the gaseous ethane source; (c) a mixed refrigerant refrigeration cycle configured to provide sufficient refrigeration to condense the gaseous ethane in the heat exchanger. In one embodiment, the mixed refrigerant refrigeration cycle further includes: at least two compression stages configured to compress the mixed refrigerant received from a warm end of the heat exchanger; a final cooler in fluid communication with the final compression stage, wherein the final cooler is configured to cool the compressed mixed refrigerant received from the final compression stage to a temperature that is sufficiently low to produce a two phase fluid; a liquid/gas separator in fluid communication with the cooler, wherein the liquid/gas separator is configured to receive the two phase fluid and separate the two phase fluid into a liquid refrigerant and a gas refrigerant; and means for expanding the gas refrigerant and the liquid refrigerant to form a cooled gas refrigerant and a cooled liquid refrigerant; wherein the heat exchanger is configured to receive the cooled gas refrigerant and the cooled liquid refrigerant such that heat from the gaseous ethane is absorbed by the cooled gas refrigerant and the cooled liquid refrigerant within the heat exchanger.

[0022] In optional embodiments of the apparatus for liquefaction of ethane:

- **[0023]** the apparatus can also include an absence of a liquid/gas separator subsequent the first compression step and prior to the last compression step;
- **[0024]** the apparatus can also include an absence of a cooler configured to condense a portion of the mixed refrigerant disposed between the at least two compression stages;
- **[0025]** the apparatus can also include a first cooler disposed between the at least two compression stages, wherein the first cooler is configured to cool the mixed refrigerant to a temperature that is sufficiently warm enough to prevent a portion of the mixed refrigerant to condense; and/or
- **[0026]** the mixed refrigerant refrigeration cycle can include an absence of a cascade cycle.

BRIEF DESCRIPTION OF THE DRAWINGS

[0027] These and other features, aspects, and advantages of the present invention will become better understood with regard to the following description, claims, and accompanying drawings. It is to be noted, however, that the drawings illustrate only several embodiments of the invention and are therefore not to be considered limiting of the invention's scope as it can admit to other equally effective embodiments.

[0028] FIG. **1** shows the prior art.

[0029] FIG. **2** shows an embodiment of the present invention.

[0030] FIG. **3** shows an embodiment of the present invention without any intercooling.

[0031] FIG. **4** shows an embodiment of the present invention with one intercooling step.

DETAILED DESCRIPTION

[0032] While the invention will be described in connection with several embodiments, it will be understood that it is not intended to limit the invention to those embodiments. On the contrary, it is intended to cover all the alternatives, modifications and equivalence as may be included within the spirit and scope of the invention defined by the appended claims.

[0033] To overcome the problems associated with the cascading refrigeration cycle of FIG. **1**, the natural progression of development for one skilled in the art would be to propose using a mixed refrigerant process similar to those used in LNG processes. However, because ethane liquefies at a warmer temperature than methane (i.e., LNG), large quantities of heavier components (e.g., butane and/or pentane) would be the natural choice of the refrigerant. For example, a typical mixed refrigerant composition for LNG production is 30% methane, 38% ethane, 11% propane, 6% butane, 15% pentane

[0034] FIG. **2** provides an example of a thermodynamically optimal solution using a mixed refrigerant refrigeration cycle. However, as the butane and pentane composition increases in the refrigerant mix, which is needed for ethane liquefaction compared to methane liquefaction, the refrigerant stream is partially liquefied by the interstage cooling **63**, **73**, **85**. This occurs because the heavier components liquefy at warmer temperatures than lighter components. The liquid from each stage **64**, **79**, **102** must be removed prior to the next compression stage to prevent mechanical damage to the compressor. This liquid is cooled and flashed to recover refrigeration as shown in FIG. **2**. While the method shown in FIG. **2** yields a thermodynamically optimal solution, it comes at the expense of having to employ a very complex exchanger, and a process that is extremely difficult to control since the quantity of liquid formed at each intercooler is very sensitive to its pressure and temperature at the intercoolers. Consequently, the temperatures at the intercoolers must be precisely controlled to compensate for fluctuations in the cooling medium, which often is related to ambient conditions. If the interstage liquid quantities cannot be precisely controlled, the liquefaction in main exchanger will either not work or be significantly penalized. This is because the design of the main heat exchange is sensitive to controlling these flow rates.

[0035] In FIG. **3**, an ethane feed **2** having a typical composition of 2% methane, 95.5% ethane, 0.5% ethylene, and 2% propane, is compressed in compressor **10** and cooled in aftercooler **20** to near ambient conditions before being further cooled and condensed in the heat exchanger **30** to form the liquid ethane product **32**. The refrigeration for the process is provided by a mixed refrigerant system **140**. In this embodiment, the mixed refrigerant **34** is compressed in a first **60**, second **70** and third stage **80** of a compressor (or in three separate compressors), without any cooling between the various compression steps to avoid liquid formation. The compressed stream **82** is cooled in aftercooler **90** and sent to a liquid/gas separator **100**, wherein the liquid **102** is cooled within the heat exchanger **30**. The gas **104** is partially cooled in the heat exchanger **30**, and then expanded in valve **110**. Following cooling, the liquid **102** and expanded gas **112** can be introduced to a second phase separator **130**, and these streams **132** are used to provide the refrigeration for the system. Those of ordinary skill in the art will recognize that the top gas of second phase separator **130** can be sent to heat exchanger **30** as a separate stream depending on the needs of the system.

[0036] FIG. **4** provides an alternate embodiment to FIG. **3**. While FIG. **3** shows no interstitial cooling, FIG. **4** provides at least one interstitial cooling stage **73** and an optional interstitial cooling stage **63**; however, the embodiment of FIG. **4** is less complex than that shown in FIG. **2**, and therefore, can provide an advantage in terms capital expenditures and ease of operation. As described above, heat exchanger **30** will only operate (or operate with a reasonable efficiency near its theoretical) if the flow rates to it are stable. Therefore, the process of FIG. **2** must precisely control three intercooler temperatures; however, the embodiment shown in FIG. **4** only needs to control two temperatures (e.g., at separators **75** and **100**) in order to operate at its highest efficiency.

[0037] In the embodiment shown in FIG. **4**, compressed refrigerant **62** is optionally cooled in interstitial cooling exchanger **63** and then fed to second compression section **70**. This compressed stream is then cooled and partially condensed in cooler **73** and fed to liquid/gas separator **75**. In the embodiment shown in FIG. **4**, top gas **77** is withdrawn and sent to third compression section **80** for further compression, and liquid refrigerant **79** is withdrawn from the bottom of liquid/gas separator **75** and introduced to heat exchanger **30** for partial cooling, before being expanded in valve **111** and combined with stream **132**.

[0038] In the optional embodiment of FIG. **4** using interstitial cooling stage, compressed gas **62** is cooled; however, it is only cooled to a temperature that is sufficiently warm enough to prevent formation of a liquid phase (i.e., it is cooled to a temperature that is still just above the boiling point of the compressed fluid). Depending on the mechanical design of second compression section **70**, justification for this partial cooling without condensation upstream of second stage **70** is based on the efficiency gain of second compression section **70** due to the cooler temperature.

[0039] Table I below provides efficiency data for the various embodiments shown in the Figures.

TABLE I

Efficiency Data for Various Embodiments

OPEX		A FIG. 1	B FIG. 2	C FIG. 3	D FIG. 4
Liquid Ethane Production	(mt/d)	5412	5412	5412	5412
Total Power	(kW)	80830	67352	79300	69251
Specific Power	(kW-h/mt)	358.4	298.7	351.7	307.1
		100.0%	116.7%	101.9%	114.3%

[0040] Table II below provides capital expenditure data, along with the proposed mixed refrigerant composition for each Figure. It is understood that capital expenditures increase with an increase in compression stages, coolers, and refrigeration loops.

TABLE II

CAPEX Data for Various Embodiments

CAPEX		A FIG. 1	B FIG. 2	C FIG. 3	D FIG. 4
Heat Exchangers	(#)	4	1	1	1
compression stages	(#)	8	3	3	3
coolers	(#)	8	3	1	2
refrigeration loops	(#)	3	4	2	3
Components in mixed refrigerant	(#)	—	4	4	4
MR composition	methane	—	15%	23%	12%
	ethane	—	33%	18%	38%
	propane	—	10%	15%	10%
	butane	—	42%	44%	39%

[0041] It is important to note that efficiency indicated for the process of FIG. **2** (298.7 kW-h/mt) is only theoretical and assumes precise control of the three compressor coolers, which is unlikely to occur during operation.

[0042] The efficiency of cycle in FIG. **3** is only slightly better than prior art of FIG. **1** (Case C efficiency is 1.9% more efficient than Case A). However, as indicated in Table II above, the cycle of FIG. **3** is much simpler (fewer refrigeration loops), and less capital expenditures (fewer compressors, compression stages and coolers), and consequently, provides a significant cost advantage over FIG. **1** and FIG. **2**.

[0043] The cycle of FIG. **4** is slightly more complex than FIG. **3** due to the one additional refrigeration loop (i.e., stream **79** and valve **111**), which is more difficult to control; however, the additional temperature control of one additional cooler is offset by a significant efficiency gain (1.9% vs 14.3%).

[0044] The number of components in the refrigerant cycle is also a degree of freedom in the balance between complexity (operability) and efficiency. Simulations found that going from four components to five components for FIG. **2**, can increase efficiency by approximately 2.6%

[0045] One side effect of reducing or removing the first and/or second stage cooling steps is a slightly reduced compressor performance. This is due to the warmer temperatures entering the second and third stages. However, this effect is only in the range of 2 to 3% and is more than compensated by the thermal efficiency gain of the main exchanger. Also, if the coolers are removed, the mechanical technology of the compressor can be adjusted to inline type rather than a bull gear type.

[0046] While the invention has been described in conjunction with specific embodiments thereof, it is evident that many alternatives, modifications, and variations will be apparent to those skilled in the art in light of the foregoing description. Accordingly, it is intended to embrace all such alternatives, modifications, and variations that fall within the spirit and broad scope of the appended claims. The present invention may suitably comprise, consist or consist essentially of the elements disclosed and may be practiced in the absence of an element not disclosed. Furthermore, language referring to order, such as first and second, should be understood in an exemplary sense and not in a limiting sense. For example, it can be recognized by those skilled in the art that certain steps or devices can be combined into a single step/device.

[0047] The singular forms "a", "an", and "the" include plural referents, unless the context clearly dictates otherwise.

[0048] Optional or optionally means that the subsequently described event or circumstances may or may not occur. The description includes instances where the event or circumstance occurs and instances where it does not occur.

[0049] Ranges may be expressed herein as from about one particular value, and/or to about another particular value. When such a range is expressed, it is to be understood that another embodiment is from the one particular value and/or to the other particular value, along with all combinations within said range.

I claim:

1. A method for the liquefaction of ethane, the method comprising the steps of:

(a) providing a stream of gaseous ethane under pressure, wherein the stream of gaseous ethane comprises at least 90% ethane; and

(b) condensing the stream of gaseous ethane to produce liquid ethane by exchanging heat with a mixed refrigerant within a heat exchanger, wherein the mixed refrigerant is subjected to a mixed refrigerant refrigeration cycle,

wherein the mixed refrigerant refrigeration cycle comprises the steps of:

compressing the mixed refrigerant in a first compression section to produce a first compressed stream;

compressing the first compressed stream in a second compression section to produce a second compressed stream;

compressing the second compressed stream in a third compression section to produce a third compressed stream;

cooling the third compressed stream to approximately ambient temperature to form a cooled third compressed stream, wherein the cooled third compressed stream is a two phase fluid;

separating the cooled third compressed stream into a liquid refrigerant and a gas refrigerant;

expanding the gas refrigerant to produce a cooled refrigerant; and

introducing the cooled refrigerant to the heat exchanger under conditions effective for absorbing heat from the gaseous ethane such that the gaseous ethane condenses to form the liquid ethane.

2. The method as claimed in claim **1**, wherein the mixed refrigerant comprises a heavy hydrocarbon selected from the group consisting of butane, pentane, and combinations thereof.

3. The method as claimed in claim **2**, wherein the mixed refrigerant further comprises a hydrocarbon selected from the group consisting of methane, ethane, ethylene, propane, and combinations thereof.

4. The method as claimed in claim **1**, wherein the gaseous ethane is at a pressure of at least 15 bara.

5. The method as claimed in claim **1**, further comprising an absence of a cooling step between each of the three compressing steps.

6. The method as claimed in claim **1**, further comprising an absence of a cooling step between the second and third compressing steps.

7. The method as claimed in claim **1**, further comprising the step of cooling the first compressed stream prior to compressing said first compressed stream in the second compression section.

8. The method as claimed in claim **7**, wherein, during the step of cooling the first compressed stream, the first compressed stream is cooled to a temperature sufficiently warm enough to prevent formation of a liquid phase.

9. The method as claimed in claim **1**, further comprising the step of expanding the liquid refrigerant and combining the expanded liquid refrigerant with the cooled refrigerant prior to the step of introducing the cooled refrigerant to the heat exchanger.

10. A method for the liquefaction of ethane, the method comprising the steps of:

(a) providing a stream of gaseous ethane under pressure; and

(b) condensing the stream of gaseous ethane to produce liquid ethane by exchanging heat with a mixed refrigerant within a heat exchanger, wherein the mixed refrigerant is subjected to a mixed refrigerant refrigeration cycle,

wherein the mixed refrigerant refrigeration cycle comprises the steps of:

compressing the mixed refrigerant to produce a first compressed stream;

compressing the first compressed stream to produce a second compressed stream;

compressing the second compressed stream to produce a third compressed stream;

cooling the third compressed stream to approximately ambient temperature to form a cooled third compressed stream, wherein the cooled third compressed stream is a two phase fluid;

separating the cooled third compressed stream into a liquid refrigerant and a gas refrigerant;

expanding the gas refrigerant to produce a cooled refrigerant; and

introducing the cooled refrigerant to the heat exchanger under conditions effective for absorbing heat from the gaseous ethane,

wherein the mixed refrigerant refrigeration cycle further comprises an absence of a formation of a liquid phase of the mixed refrigerant at a point that is located both downstream the first compression step and upstream the final compression step.

11. The method as claimed in claim **10**, wherein there is an absence of a liquid/gas separation subsequent the first compression step and prior to the last compression step.

* * * * *