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(54) **SYSTEMS AND METHODS FOR DELIVERING HYDROGEN AND SEPARATION OF HYDROGEN FROM A CARRIER MEDIUM**

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(57) **ABSTRACT**

Methods and systems are provided for the transportation of hydrogen including the use of a carrier medium and the subsequent separation of the hydrogen from the carrier medium upon delivery of the mixed gas stream to a desired location. In one example, up to approximately 20% hydrogen (molar fraction) may be mixed with natural gas and transported through a natural gas pipeline. The hydrogen may be separated from the natural gas by liquefying the natural gas and separating the liquid and vapor components in a gas-liquid separator to form a natural gas stream and a hydrogen stream. One or both of the separated streams may be used in cooling of the mixed stream. Precompression and expansion pressure of the various streams may be used to influence the purity level of the hydrogen. Other constituents, or impurities, present in the mixed gas stream may also be removed as may be desired.

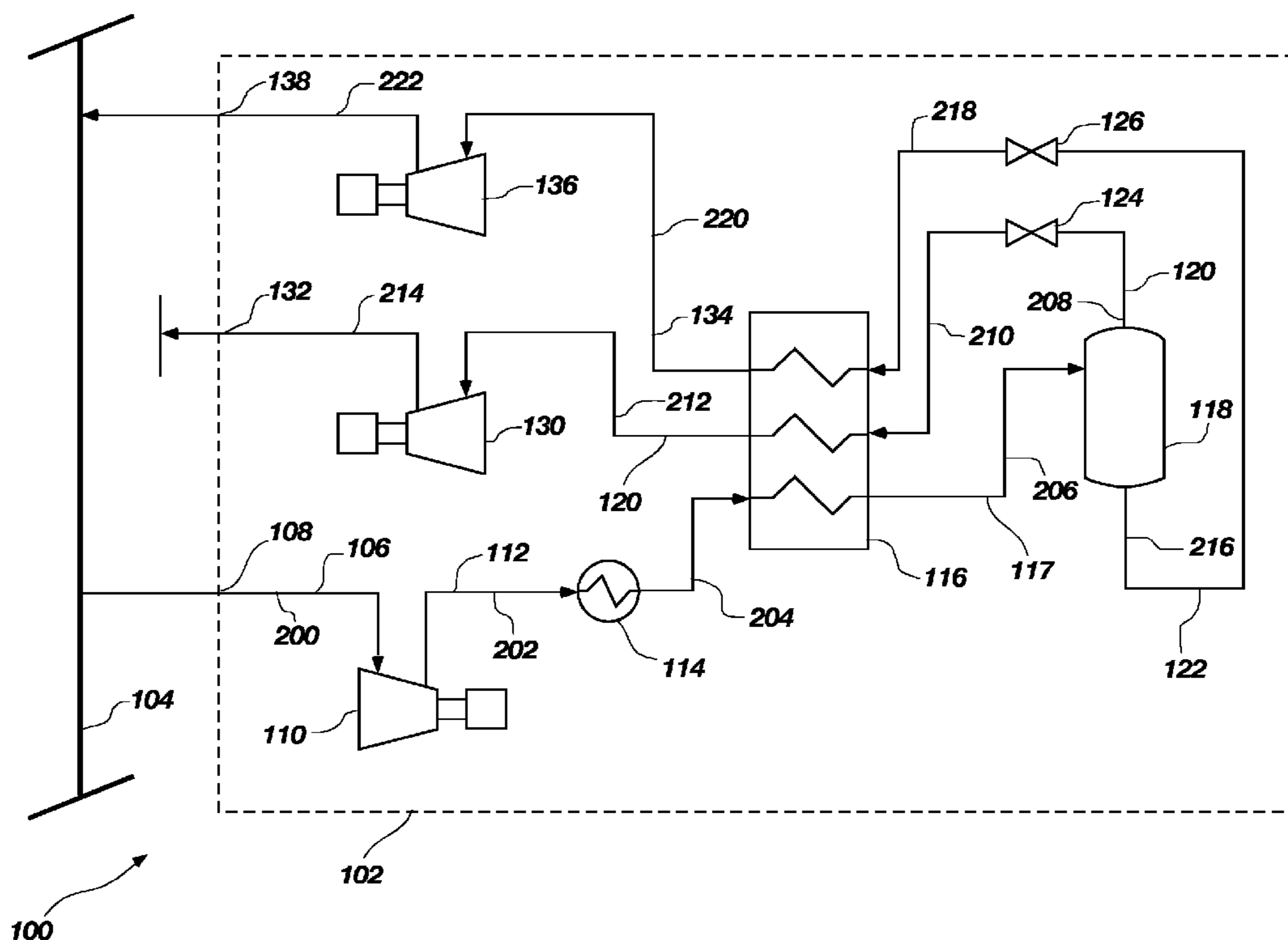
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Related U.S. Application Data

(60) Continuation-in-part of application No. 11/124,589, filed on May 5, 2005, which is a continuation of application No. 10/414,991, filed on Apr. 14, 2003, now Pat. No. 6,962,061, which is a division of application No. 10/086,066, filed on Feb. 27, 2002, now Pat. No. 6,581,409.



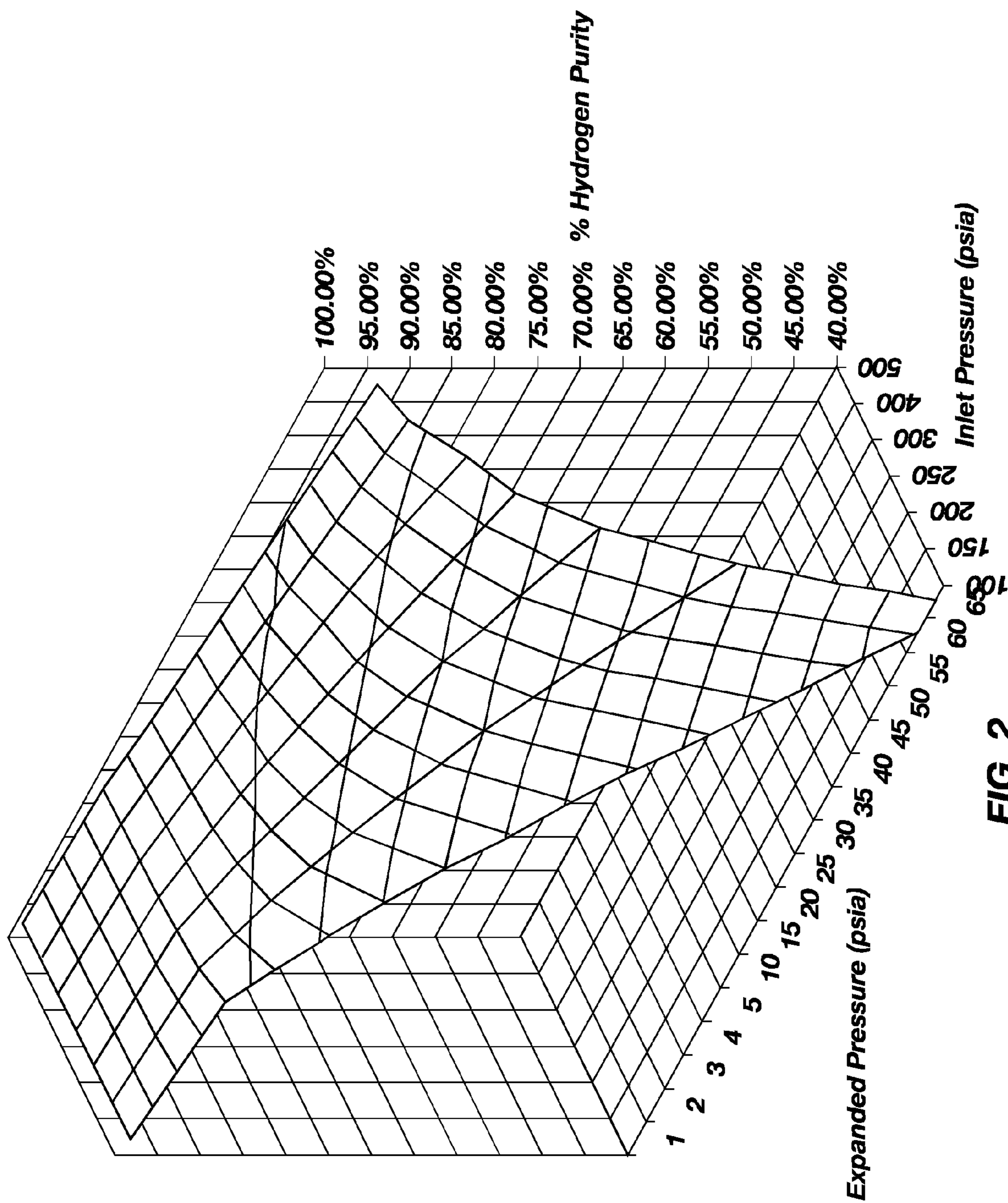


FIG. 2

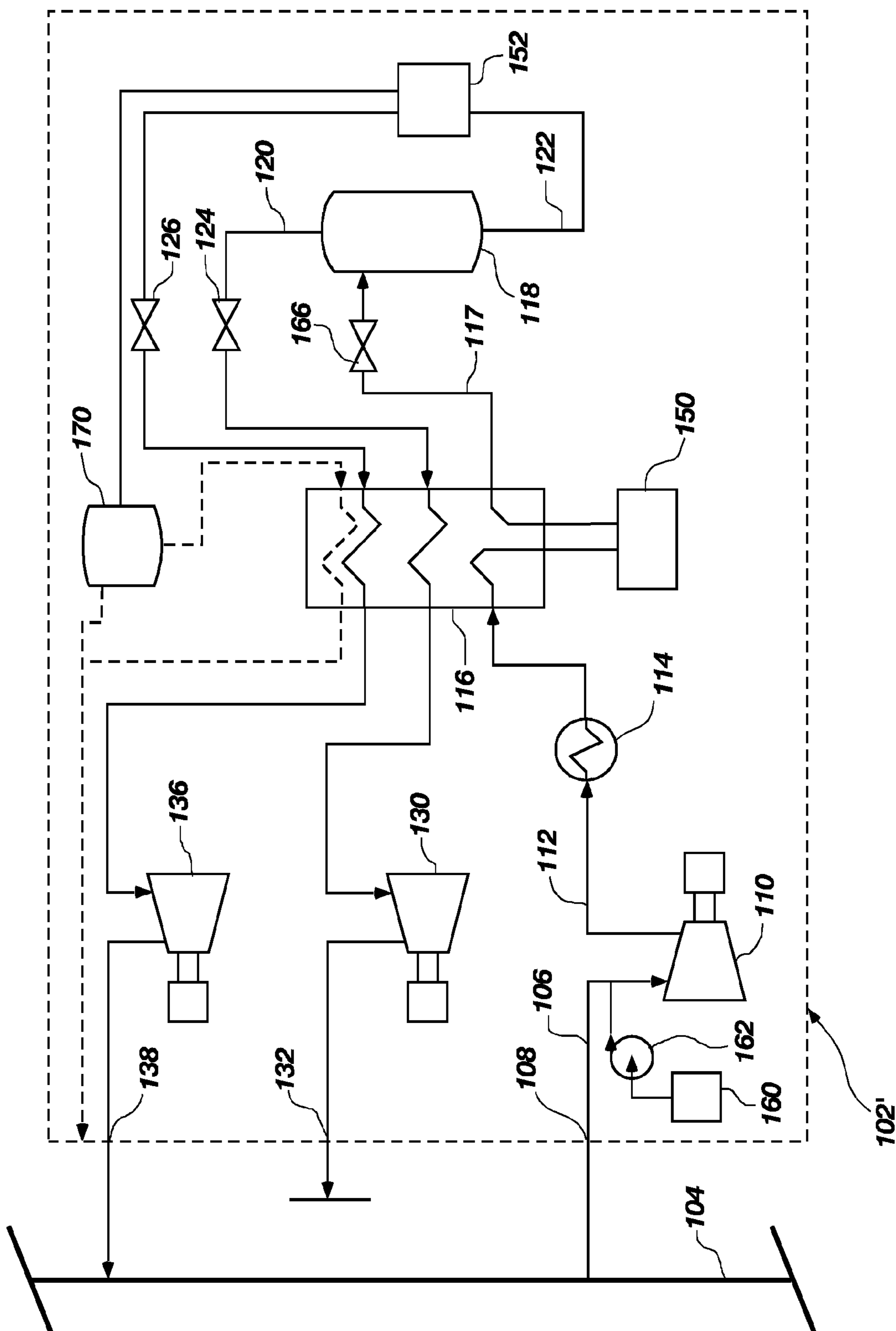


FIG. 3

| | State Point 200 | State Point 202 | State Point 204 | State Point 206 | State Point 208 | State Point 210 | State Point 212 | State Point 214 | State Point 216 | State Point 218 | State Point 220 | State Point 222 |
|----------------------------------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| Vapor Fraction | 1 | 1 | 1 | 0.1972 | 1 | 1 | 1 | 1 | 0 | 0.0528 | 1 | 1 |
| Temperature (K) | 288.7 | 338.2 | 288.7 | 108.1 | 108.1 | 102.2 | 264.7 | 453.2 | 108.1 | 104.9 | 278 | 418.5 |
| Pressure (N/m ²) | 2.07E+06 | 3.45E+06 | 3.45E+06 | 3.45E+06 | 3.45E+06 | 1.01E+05 | 1.01E+05 | 4.48E+05 | 3.45E+06 | 1.01E+05 | 1.01E+05 | 4.48E+05 |
| Molar Flow (gmole/s) | 90.79 | 90.79 | 90.79 | 90.79 | 17.9 | 17.9 | 17.9 | 17.9 | 72.89 | 72.89 | 72.89 | 72.89 |
| Mass Flow (kg/h) | 4536 | 4536 | 4536 | 4536 | 187.7 | 187.7 | 187.7 | 187.7 | 4348 | 4348 | 4348 | 4348 |
| Liquid Volume Flow (m ³ /h) | 16.05 | 16.05 | 16.05 | 16.05 | 1.92 | 1.92 | 1.92 | 1.92 | 14.13 | 14.13 | 14.13 | 14.13 |
| Heat Flow (kW) | -5422 | -5269 | -5439 | -6549 | -145.5 | -145.5 | -62.88 | 34.81 | -6403 | -6403 | -5376 | -4980 |
| Molar Enthalpy (J/gmole) | -5.97E+04 | -5.80E+04 | -5.99E+04 | -7.21E+04 | -8.13E+03 | -8.13E+03 | -3.51E+03 | 1.94E+03 | -8.79E+04 | -8.79E+04 | -7.38E+04 | -6.83E+04 |

FIG. 4

| | State Point 200 | State Point 202 | State Point 204 | State Point 206 | State Point 208 | State Point 210 | State Point 212 | State Point 214 | State Point 216 | State Point 218 | State Point 220 | State Point 222 |
|----------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| Methane | 0.76 | 0.76 | 0.76 | 0.76 | 0.0341 | 0.0341 | 0.0341 | 0.0341 | 0.9383 | 0.9383 | 0.9383 | 0.9383 |
| Hydrogen | 0.2 | 0.2 | 0.2 | 0.2 | 0.09498 | 0.09498 | 0.09498 | 0.09498 | 0.0158 | 0.0158 | 0.0158 | 0.0158 |
| Nitrogen | 0.016 | 0.016 | 0.016 | 0.016 | 0.0161 | 0.0161 | 0.0161 | 0.0161 | 0.016 | 0.016 | 0.016 | 0.016 |
| Ethane | 0.016 | 0.016 | 0.016 | 0.016 | 0 | 0 | 0 | 0 | 0.0199 | 0.0199 | 0.0199 | 0.0199 |
| Propane | 0.008 | 0.008 | 0.008 | 0.008 | 0 | 0 | 0 | 0 | 0.01 | 0.01 | 0.01 | 0.01 |

FIG. 5

**SYSTEMS AND METHODS FOR DELIVERING
HYDROGEN AND SEPARATION OF HYDROGEN
FROM A CARRIER MEDIUM**

CROSS-REFERENCE TO RELATED
APPLICATIONS

[0001] This application is a continuation-in-part of U.S. patent application Ser. No. 11/124,589 filed on May 5, 2005, which is a continuation of U.S. patent application Ser. No. 10/414,991 filed on Apr. 14, 2003, now U.S. Pat. No. 6,962,061 issued on Nov. 8, 2005, which is a divisional of U.S. patent application Ser. No. 10/086,066 filed on Feb. 27, 2002, now U.S. Pat. No. 6,581,409 issued on Jun. 24, 2003 and which claims the benefit of U.S. Provisional Patent Application Ser. No. 60/288,985, filed May 4, 2001, the disclosures of which applications are each hereby incorporated by reference in the entireties.

GOVERNMENT RIGHTS

[0002] The United States Government has certain rights in this invention pursuant to Contract No. DE-AC07-05ID14517 between the United States Department of Energy and Battelle Energy Alliance, LLC.

BACKGROUND OF THE INVENTION

[0003] 1. Field of the Invention

[0004] The present invention relates generally to the transportation and delivery of gases such as hydrogen and, more particularly, the transportation and delivery a first gas inter-mixed with another gas and the subsequent separation of such gases at desired levels of purity.

[0005] 2. State of the Art

[0006] Hydrogen is considered a promising energy alternative to carbon based fuels. Various technologies have been and are still being developed regarding the production and use of hydrogen as a fuel or energy source. While many people consider hydrogen to be a desirable energy alternative to carbon based fuels, often based on the perceived lack of pollutants in using hydrogen as an energy source, various obstacles exist in creating a society which relies in substantial part on hydrogen as opposed to other forms of energy. Such obstacles generally include the ability to efficiently, safely and economically produce, transport and store hydrogen.

[0007] Hydrogen is conventionally transported and delivered by way of railroad cars, tanker trucks and tanker ships. Such means of transportation and delivery are generally considered less economically desirable as compared to transporting other fuels in the same manner. One reason for this is the energy density of hydrogen as compared to that of other fuels. For example, substituting the same volume of gasoline for hydrogen in a tanker would result in the effective delivery of three times the amount of energy by the tanker. Considering this from another standpoint, the net heat of combustion of hydrogen per unit volume has been stated to be lower than that of methane by a factor of 3.3 or greater. Thus, hydrogen would have to be compressed at least 3.3 times as much as methane in order to transport the same energy quantity as methane for a given volume. Substantial expenses in equipment and energy would be required (not to mention potential structural upgrades in

transportation equipment) to compress and transport hydrogen at such elevated pressures.

[0008] A limited amount of hydrogen is currently shipped by pipelines specifically designed and built for carrying hydrogen. For example, according to a report by the International Atomic Energy Agency (IAEA) in May of 1999, a pipeline in the Rhine region includes approximately 250 kilometers (km) of pipe which delivers 250,000,000 cubic meters per year at a system pressure of 2.2 megaPascals (MPa). According to the same report, 232 km of hydrogen pipeline is installed near Houston, Texas, delivering hydrogen at a pressure of 6 MPa. However, as already noted, dedicated hydrogen pipelines are specifically designed and constructed in accordance with special issues associated with delivering hydrogen.

[0009] For example, the hydrogen molecule is extremely small and, therefore, can escape through the lattice of metals. Additionally, the hydrogen molecule can interact with certain metals, causing the metals to become brittle. Escaping hydrogen results in inefficiencies of the delivery system. Embrittlement of the pipeline clearly poses a threat of structural failure. Thus, in building hydrogen specific pipelines, more expensive materials (such as stainless steel) must be used or, alternatively (or in addition to the use of special materials), the inner surfaces of the pipelines need to be coated so as to protect the metals and prevent embrittlement. Additionally, in transporting hydrogen by pipeline multi-stage compression is likely to be required due to higher friction losses caused by the compressibility factor previously mentioned. These factors, coupled with the likely issues regarding obtaining licenses, permits and satisfying other regulatory requirements, pose substantial hurdles to building a comprehensive pipeline dedicated to hydrogen transportation and delivery.

[0010] Another concept for transporting hydrogen through pipelines includes injecting the hydrogen into a carrier medium. For example, the above-referenced IAEA report indicates that hydrogen (H₂) may be mixed with natural gas in quantities of up to 5% (mole fraction) of hydrogen and carried in existing natural gas pipelines without requiring any modifications to the pipeline. Additionally, the same report indicates that hydrogen may be added to natural gas in a quantity of up to 20% (mole fraction) and carried in existing pipelines with relatively minimal modifications of such pipelines. However, in order for such a transportation and delivery option to be feasible, separation of the hydrogen from the natural gas, or other carrier medium, in desired levels of purity will be required. Existing facilities that might be used for separating natural gas and hydrogen are large scale with footprints measured in acres and with estimated capital costs ranging from \$50 million to \$3 billion. Smaller scale, "appliance" size plants for separating natural gas and hydrogen are not currently known to be available in the market.

[0011] With hydrogen being a focus for the energy needs of the future, the economical transportation of large volumes of hydrogen will be required. It would be desirable to provide a more efficient method and system of transporting hydrogen. It would also be desirable to provide a method and system of transporting hydrogen that utilizes existing infrastructure and, perhaps, avoids many of the regulatory issues that would be encountered in implementing new

infrastructure. Further, it would be desirable to provide systems and methods for separating hydrogen from a carrier medium at desired purity levels to take advantage of potential pipeline distribution of hydrogen.

BRIEF SUMMARY OF THE INVENTION

[0012] In accordance with one embodiment of the present invention, a method is provided for separating hydrogen from natural gas, both of which are contained in a mixed gas stream. The method includes cooling the mixed gas stream and liquefying a substantial portion of the volume of natural gas. The liquefied natural gas is substantially separated from the volume of hydrogen to provide a natural gas stream and a hydrogen stream. At least one of the natural gas stream and the hydrogen stream are used to cool the mixed gas stream. In one embodiment, both the natural gas stream and the hydrogen gas stream are used to cool the mixed gas stream. The desired purity level of the resulting hydrogen stream may be altered by compressing the incoming mixed gas stream, adjusting the pressure to which the gas stream is expanded, or by both actions.

[0013] In accordance with another embodiment of the present invention, a method is provided for transporting hydrogen. The method includes adding a volume of hydrogen to a volume of natural gas to provide a mixed gas stream. The mixed gas stream is flowed from a first location to a second location and then cooled. A substantial portion of the volume of natural gas is liquefied and the liquefied natural gas is separated from the volume of hydrogen to provide a natural gas stream and a hydrogen stream. At least one of the natural gas stream and the hydrogen stream is used to cool the mixed gas stream. In one embodiment, both the natural gas stream and the hydrogen gas stream are used to cool the mixed gas stream. The desired purity level of the resulting hydrogen stream may be altered by compressing the incoming mixed gas stream, adjusting the pressure to which the gas stream is expanded, or by both actions.

[0014] In accordance with another embodiment of the present invention, a system is provided for separating hydrogen from natural gas. The system includes a first compressor, a first heat exchanger, a second heat exchanger, a gas-liquid separator and at least one expansion device. The system further includes a first flow path that is defined and configured to deliver a mixed gas stream sequentially through the first compressor, the second compressor, and into the gas-liquid separator. The system additionally includes a second flow path that is defined and configured to deliver at least one of liquid natural gas and hydrogen gas from the gas liquid separator, through the at least one expansion device and through the second heat exchanger. In other embodiments, the system may include additional or different components.

[0015] Other methods and systems are also provided in accordance with various embodiments of the present invention as will be appreciated upon a reading of the detailed description of the various embodiments of the present invention.

BRIEF DESCRIPTION OF THE SEVERAL VIEWS OF THE DRAWINGS

[0016] The foregoing and other advantages of the invention will become apparent upon reading the following detailed description and upon reference to the drawings in which:

[0017] FIG. 1 is a process diagram of one embodiment of a system and process for separating hydrogen from a carrier medium;

[0018] FIG. 2 is a chart showing purity levels of hydrogen based on inlet and outlet pressures;

[0019] FIG. 3 is a process diagram of another embodiment of a system and process for separating hydrogen from a carrier medium;

[0020] FIG. 4 is a table listing various characteristics of fluids at certain locations within the system shown in FIG. 1 in accordance with an embodiment of the present invention; and

[0021] FIG. 5 is a table listing the gas compositions at various locations within the system shown in FIG. 1 in accordance with an embodiment of the present invention.

DETAILED DESCRIPTION OF THE INVENTION

[0022] Referring to FIG. 1, a system 100 is shown for separating a desired gas from a carrier medium. While the present embodiment is set forth in terms of separating hydrogen from natural gas, it is noted that the present invention may be practiced using various other types of gases and carrier mediums.

[0023] The system 100 includes a separating plant 102 which is coupled to a source of gas such as, for example, a pipeline 104. In one example, the gas stream may be flowing through the pipeline 104 which includes approximately 80% natural gas (e.g., approximately 95% methane, approximately 2% ethane, approximately 2% nitrogen and approximately 1% propane) and approximately 20% hydrogen (percentages in molar fractions). An incoming stream of gas 106 from the natural gas/hydrogen stream in the pipeline 104 may be drawn into the plant 102 through an inlet 108 at, for example, a pressure of approximately 100 pounds per square inch absolute (psia) to approximately 500 psia depending on the pressure of the stream in the pipeline 104. If desired, the pressure of the incoming stream 106 may be flowed through a compressor 110 to raise the pressure to a desired level. For example, the incoming stream 106 may be compressed to raise the pressure to between, for example, approximately 300 psia and approximately 500 psia. As will be discussed in further detail hereinbelow, an increase in the pressure of the gas stream at the inlet 106 of the plant 102 may result in increased purity of the hydrogen that is separated from the natural gas or other carrier medium.

[0024] If the incoming stream does pass through the compressor 110, heat energy will be added to the stream. To remove the added heat energy, or at least a portion thereof, the compressed gas stream 112 may be flowed through a heat exchanger 114. The heat exchanger 114 may use ambient conditions, such as, for example, air, water, or ground temperature, or a combination thereof, for cooling the compressed gas stream 112. In one example, an ambient heat exchanger 114 may be designed to process the compressed gas stream 112 at approximately 4500 to 4600 lbs mass per hour (lbn/hr) at a design pressure of approximately 500 psia. In one embodiment, the heat exchanger 114 may further be configured such that the inlet temperature of the gas is approximately 149° F. and the outlet temperature of the gas

is approximately 59° F. The heat exchanger 114 may be provided with a fan that is driven, for example, by a suitable electric motor.

[0025] The compressed gas stream 112 may then enter a first side, or a warm side, of a high efficiency heat exchanger 116 where, if desired, the compressed gas stream 112 may be cooled to cryogenic temperatures such that the natural gas becomes liquefied. For example, the compressed gas stream may be cooled to temperatures below approximately -200° F. In one embodiment, the high efficiency heat exchanger 116 may be configured as a countercurrent flow, plate and fin type heat exchanger. Additionally, the plates and fins may be formed of a highly thermally conductive material such as, for example, aluminum. The high efficiency heat exchanger 116 is positioned and configured to efficiently transfer as much heat as possible from the compressed gas stream 112 to the cooling streams that are discussed in further detail below. The high efficiency heat exchanger 116 may be configured, in one example, such that the inlet temperature of the gas will be approximately 59° F. and the outlet temperature of the gas will be approximately -265° F.

[0026] The cooled stream 117 flows from the high efficiency heat exchanger 116 to a gas-liquid separator 118. The separator may include a pressure vessel configured to withstand a desired pressure. For example, the separator 118 may be maintained at a pressure of approximately 500 psia. The hydrogen vapor and liquid natural gas (LNG) are separated in the gas-liquid separator 118. A hydrogen stream 120 flows from an upper portion of the separator 118 through appropriate piping. An LNG stream 122 flows from a lower portion of the separator 118 through appropriate piping. The hydrogen stream 120 and the LNG stream 122 each flow through associated expansion valves 124 and 126, respectively, causing each of the associated streams to expand to pressures of, for example, between approximately 1 psia and approximately 65 psia and cool to temperatures of, for example, between approximately -270° F. and approximately -280° F. The expansion valves 124 and 126 may include Joule-Thomson (JT) valves which operate on the principle that expansion of gas will result in an associated cooling of the gas as well, as is understood by those of ordinary skill in the art. Of course other expansion devices may be used in place of the expansion valves 124 and 126 if desired.

[0027] After flowing through their associated expansion valves 124 and 126, the hydrogen stream 120 and LNG stream 122 enter the second, or cold, side of the high efficiency heat exchanger 116 providing cooling to the compressed gas stream 112. In one example, the temperature of the hydrogen stream 120 and the LNG stream 122 may be raised to between approximately 15° F. and 40° F. as it passes through the heat exchanger 116. It is noted that the LNG stream 122 returns to a gaseous state after passing sequentially through the expansion valve 126 and the heat exchanger 116. The hydrogen stream 120 may then flow through a compressor 130, if desired, to raise its pressure to, for example approximately 65 psia at a temperature of approximately 355° F. before flowing through a plant outlet 132 to be collected for storage, further processing or use, for example, as a fuel. Similarly, the natural gas stream (now indicated by reference numeral 134 based on its state change) may flow through a compressor 136, if desired, to raise its pressure to, for example, approximately 65 psia at

a temperature of approximately 294° F. before flowing through a plant outlet 138 to be collected for storage, further processing or use, for example, as a fuel. In one example, the natural gas may be returned to the pipeline 104 from which it was taken, downstream from the location where it was originally removed from the pipeline 104. It is noted that, if the pressures in the hydrogen stream 120 and the natural gas stream 134 are below atmospheric pressures as they exit the heat exchanger 116, vacuum pumps (not shown) may be coupled to the hydrogen and natural gas streams 120 and 134 in order to obtain proper operation of the plant 102. While the use of vacuum pumps would add cost to the overall cost of the system 100, operation at such low pressures would result in extremely pure hydrogen (e.g., purity levels of approximately 99% or higher) at the outlet 132.

[0028] Referring briefly to FIG. 2, a three-dimensional plot depicts the impact of the inlet pressure (i.e. the pressure of the incoming stream of gas 106 or the pressure to which the incoming stream is compressed by compressor 110) and the pressure to which the gas stream or streams are expanded (i.e., the pressure of the hydrogen stream 120 after flowing through the expansion valve 124 and the pressure of the natural gas stream 122 after flowing through the expansion valve 126) on the resulting purity of the hydrogen. Generally, at relatively high expansion pressures, the hydrogen purity tends to be low. However, the level of hydrogen purity rises as the inlet pressure rises. At low expansion pressures the purity of hydrogen is generally above 95% and the inlet pressure has relatively little effect on the level of hydrogen purity. As seen in FIG. 2, if inlet pressures are high enough (e.g., approximately 300 psia or higher) the level of hydrogen purity is above 90% even for expansion pressures that are in the range of approximately 55 psia. Thus, controlling the inlet pressures and expansion pressure of the plant, such as by using precompression (i.e., with compressor 110) and various expansion devices, also controls the purity level of the hydrogen separated from the natural gas.

[0029] It is also noted that the level of other constituents present within the gas stream (i.e., prior to entering the plant inlet 108) may also have an impact on the purity level of hydrogen obtained from the separation process. For example, it has been determined that as the nitrogen level increases, as measured at the inlet 108, the purity of the hydrogen decreases at the outlet 132. It is believed that this is because the nitrogen does not liquefy with the natural gas and, therefore, is withdrawn with the hydrogen from the gas-liquid separator 118. Additionally, it is noted that the amount of methane at the outlet 132 (in the hydrogen stream 120) decreases with such an increase of nitrogen.

[0030] Depending on the intended use of the hydrogen, the existence of either nitrogen or methane in certain quantities may have an undesirable effect on the performance of the hydrogen in its final use. For example, if the hydrogen is used in a fuel cell, the methane can mix with oxygen to form carbon monoxide and carbon dioxide, which can block the access of hydrogen to the fuel cell's catalyst sites. On the other hand, the presence of nitrogen may adversely affect the performance of a fuel cell by forming a nitrogen blanket around the cathode. Thus, depending on the intended use of the hydrogen, the composition of the incoming stream of gas 106 may need to be taken into account as another factor in adjusting inlet and outlet pressures and controlling the purity level of the hydrogen.

[0031] Consideration may also need to be given to the potential of impurities (e.g., carbon dioxide and water) present in the natural gas of the incoming stream 106. It is generally noted that impurities such as carbon dioxide and water will become solids at the cryogenic temperatures that exist in the plant 102. Solid carbon dioxide or water could build up and block the passages within one of the heat exchangers (e.g., heat exchanger 116), the valves 124 and 126 (or other expansion devices) or any of the associate piping or separator. Various options may be used to deal with such potential solid-forming impurities.

[0032] For example, in one embodiment, an amine scrubber system may be used wherein an amine solution captures the impurities from the incoming stream of gas 106 by way of a pack column. The impurities may be subsequently separated from the amine solution by way of energy input such that the amine solution may be recycled and reused.

[0033] In another embodiment, a ceramic palladium membrane system may be used to remove the impurities from the incoming stream of gas 106. The ceramic membrane allows the hydrogen molecule (H₂) to pass through while preventing the passage of larger molecules contained in the natural gas. While such a system would likely be costly, purity levels of hydrogen approaching 100% could conceivably be obtained.

[0034] In yet another embodiment, water management and carbon dioxide management systems may be used, including configurations similar to those described in U.S. Pat. No. 6,581,409 entitled APPARATUS FOR THE LIQUEFACTION OF NATURAL GAS AND METHODS RELATING TO SAME, the disclosure of which is incorporated by reference herein in its entirety.

[0035] For example, with reference to FIG. 3, a plant 102' may be configured generally similar to the plant 102 described with respect to FIG. 1, but with some modifications to accommodate the implementation of a water management system 150, a carbon dioxide management system 152 or both. In such an embodiment, methanol, or some other water absorbing material, may be injected into the incoming stream of gas 106 to enable the removal of water from the natural gas stream during the processing of the incoming stream of gas 106 and for prevention of ice formation throughout the plant 102'. In connection with the water management system 150, a source of methanol 160, or some other water absorbing product, may be injected into the gas stream, via a pump 162, at a location prior to the gas being passed through the high efficiency heat exchanger 116. The pump 162 may include variable flow capability to inject methanol into the gas stream by way of, for example, an atomizing or a vaporizing nozzle. Alternatively, valving may be used to accommodate multiple types of nozzles such that an appropriate nozzle may be used depending on the flow characteristics of the incoming stream of gas 106.

[0036] A suitable pump 162 for injecting the methanol may include variable flow control in the range of 0.4 to 2.5 gallons per minute (GPM) at a design pressure of approximately 1000 psia for a water content of approximately 2 to 7 pounds mass per millions of standard cubic feet (lbm/mm scf). The variable flow control may be accomplished through the use of a variable frequency drive coupled to a motor of the pump 162. One such pump is available from America LEWA located in Holliston, Mass.

[0037] The methanol is mixed with the incoming stream of gas 106 to lower the freezing point of any water which may be contained therein. The methanol mixes with the incoming stream of gas 106 and binds with the water to prevent the formation of ice. Part way through the heat exchange process in the high efficiency heat exchanger 116 (e.g., at temperatures between approximately -60° F. and -90° F) the methanol and water form a liquid. The compressed gas stream 112 is temporarily diverted from the heat exchanger 116 and passed through a water management 150 system which, in one embodiment may include a separating tank wherein the methanol/water liquid is separated from the compressed gas stream 112. The liquid may then be discharged from the separator tank and the gas may flow, for example, through a coalescing filter to remove an additional amount of the methanol/water mixture. The methanol/water mixture may be discharged from the coalescing filter through appropriate piping and the dried gas may then reenter the heat exchanger 116 for further cooling and processing.

[0038] Additionally, the plant 102' may include other systems to handle other impurities such as a carbon dioxide management system 152. In one example of such a system, instead of reducing the temperature of the compressed gas stream 112 within the heat exchanger 116 to a temperature that would result in the production of solid carbon dioxide and potential blockage of the heat exchanger 116, the heat exchanger 116 may be used to lower the temperature of the compressed gas stream to a temperature slightly above the temperature at which carbon dioxide becomes a solid (e.g., from approximately -185° F. to -195° F.). Just prior to entry into the gas liquid separator 118, the cooled gas stream 117 may pass through an expansion device 166, such as a JT valve, to expand the gas and lower the temperature of the gas stream to a level such that LNG and solid carbon dioxide are produced within the gas-liquid separator 118. The hydrogen may be removed from the gas-liquid separator 118 as discussed previously with respect to the plant described in FIG. 1.

[0039] The slurry of solid carbon dioxide and LNG may be removed from the gas-liquid separator 118 to separate the carbon dioxide from the LNG, if desired, using a carbon dioxide separation system 152 which may include the use of hydrocyclones, filters or both (such as detailed in U.S. Pat. No. 6,581,409). The LNG may be processed and used for cooling in the heat exchanger 116 as previously described herein. The solid carbon dioxide may be passed, for example, to a sublimation tank 170 where the carbon dioxide returns to a gaseous state in a manner similar to that which is described in U.S. Pat. No. 6,581,409. The gaseous carbon dioxide may then be passed through heat exchanger 116 to provide additional cooling, returned directly to the pipeline 104, or otherwise processed or disposed of as desired as generally indicated on FIG. 3 by dashed lines.

[0040] The system and process shown in FIG. 3 thus provides for efficient separation of hydrogen from a carrier medium, such as natural gas, while integrating processes to remove impurities such as water or carbon dioxide without expensive equipment and preprocessing.

[0041] It is noted that implementation of such a carbon dioxide management system 152 may require further compression of the incoming stream of gas 106 to accommodate

the expansion activity taking place before the entrance of the cooled stream into the gas-liquid separator **118**.

[0042] Various control schemes may be used to operate the plants **102** and **102'**. Also, additional piping, valving and other process equipment may be utilized such as described in the various documents incorporated by reference herein and as will be appreciated by those of ordinary skill in the art.

EXAMPLE

[0043] The system **100** described with respect to FIG. **1** was modeled to determine the characteristics of the fluid within the system **100** at specific state points for separation of hydrogen from natural gas in a mixed stream having 20% hydrogen and 80% natural gas (molar fractions). FIG. **4** is a table setting forth such state point characteristics. The locations of state points are identified on FIG. **1** wherein state point **200** is at the inlet **108** of the plant **102**. State point **202** refers to the compressed gas stream **112** at a location after the compressor **110** and prior to the ambient heat exchanger **114**. State point **204** refers to the compressed gas stream **112** at a location between the two heat exchangers **114** and **116**. State point **206** refers to the cooled stream **117** at a location between the heat exchanger **116** and the separator **118**. State point **208** refers to the hydrogen stream **120** as it leaves the gas-liquid separator **118**. State point **210** refers to the hydrogen stream **120** at a location between the expansion valve **124** and the heat exchanger **116**. State point **212** refers to the hydrogen stream **120** at a location between the heat exchanger **116** and the compressor **130**. State point **214** refers to the hydrogen stream at a location after it passes through the compressor **130**.

[0044] State point **216** refers to the LNG stream **122** as it leaves the gas-liquid separator **118**. State point **218** refers to the LNG stream **122** at a location between the expansion valve **126** and the heat exchanger **116**. State point **220** refers to the natural gas stream **134** at a location between the heat exchanger **116** and the compressor **136**. And state point **222** refers to the natural gas stream **134** after it leaves the compressor **136**.

[0045] FIG. **5** further shows the composition of various gas streams at identified state points. The compositions are set forth in molar fractions of the identified constituents.

[0046] It is noted that, for the purposes of the model described with respect to FIGS. **1**, **4** and **5**, that the power input for the compressor **110** will be approximately 153 kiloWatts (kW), the power input for the natural gas compressor **136** will be approximately 395 kW, and the power input for the hydrogen compressor **130** will be approximately 98 kW.

[0047] Thus, various embodiments of the invention provide systems and methods for transporting, delivering and separating hydrogen and a carrier medium, such as natural gas, with the mixture containing up to approximately 20% hydrogen while obtaining purity levels of the separated hydrogen from, for example, approximately 80% to approximately 99%. Such capability will enable the use of existing pipelines for the transport and delivery of hydrogen without major modifications to such pipelines which would require substantial time and capital investment.

[0048] While the invention may be susceptible to various modifications and alternative forms, specific embodiments

have been shown by way of example in the drawings and have been described in detail herein. However, it should be understood that the invention is not intended to be limited to the particular forms disclosed. Rather, the invention includes all modifications, equivalents, and alternatives falling within the spirit and scope of the invention as defined by the following appended claims.

What is claimed is:

1. A method of transporting hydrogen, the method comprising:

adding a volume of hydrogen to a volume of natural gas to form a mixed gas volume;

flowing the mixed gas volume as a mixed gas stream from a first location to a second location;

cooling the mixed gas stream and liquefying a substantial portion of the volume of natural gas;

substantially separating the liquefied natural gas from the volume of hydrogen to provide a natural gas stream and a hydrogen stream; and

using at least one of the natural gas stream and the hydrogen stream to cool the mixed gas stream.

2. The method according to claim 1, further comprising compressing the mixed gas stream prior to cooling the mixed gas stream.

3. The method according to claim 2, wherein compressing the mixed gas stream includes compressing the mixed gas stream to a pressure of approximately 300 pounds per square inch absolute or greater.

4. The method according to claim 2, wherein cooling the mixed gas stream includes flowing the mixed gas stream sequentially through a first heat exchanger and a second heat exchanger.

5. The method according to claim 4, wherein flowing the mixed gas stream through the first heat exchanger includes cooling the mixed gas stream to a temperature of approximately 60° F.

6. The method according to claim 5, wherein flowing the mixed gas stream through the second heat exchanger includes cooling the mixed gas stream to a temperature of approximately -265° F.

7. The method according to claim 4, wherein substantially separating the liquefied natural gas from the volume of hydrogen to provide a natural gas stream and a hydrogen stream includes flowing the mixed gas stream from the second heat exchanger into a gas-liquid separator.

8. The method according to claim 4, wherein using at least one of the natural gas stream and the hydrogen stream to cool the mixed gas stream includes expanding at least one of the natural gas stream and the hydrogen stream and flowing the expanded stream through the second heat exchanger.

9. The method according to claim 4, wherein using at least one of the natural gas stream and the hydrogen stream to cool the mixed gas stream includes expanding the natural gas stream, expanding the hydrogen stream, and flowing both the expanded natural gas stream and the expanded hydrogen stream through the second heat exchanger.

10. The method according to claim 9, further comprising compressing the expanded natural gas stream.

11. The method according to claim 10, further comprising compressing the expanded hydrogen stream.

12. The method according to claim 9, further comprising controlling a purity of the hydrogen stream, at least in part, by selecting the pressure to which at least one of the hydrogen stream and the natural gas stream is expanded.

13. The method according to claim 12, further comprising expanding the at least one of the hydrogen stream and the natural gas stream to a pressure of approximately 65 pounds per square inch absolute (psia) or lower.

14. The method according to claim 15, further comprising controlling a purity of the hydrogen stream, at least in part, by selecting the pressure to which the mixed gas stream is compressed.

15. The method according to claim 1, further comprising expanding at least one of the hydrogen stream and the natural gas stream and controlling a purity of the hydrogen stream, at least in part, by selecting the pressure to which the at least one of the hydrogen stream and the natural gas stream is expanded.

16. The method according to claim 15, further comprising expanding the at least one of the hydrogen stream and the natural gas stream to a pressure of approximately 65 pounds per square inch absolute (psia) or lower.

17. The method according to claim 16, further comprising compressing the mixed gas stream prior to cooling the mixed gas stream and controlling a purity of the hydrogen stream, at least in part, by selecting the pressure to which the mixed gas stream is compressed.

18. The method according to claim 1, further comprising removing impurities from the natural gas.

19. The method according to claim 18, wherein removing impurities from the natural gas further includes removing water from the natural gas.

20. The method according to claim 19, wherein removing water from the natural gas includes injecting methanol into the mixed gas stream, liquefying the water and methanol, and separating the water and methanol from the mixed gas stream.

21. The method according to claim 18, wherein removing impurities from the natural gas includes removing carbon dioxide from the natural gas.

22. The method according to claim 21, wherein removing carbon dioxide from the natural gas includes producing a slurry of solid carbon dioxide and natural gas and substantially separating the solid carbon dioxide from the natural gas.

23. The method according to claim 22, further comprising subliming the solid carbon dioxide.

24. The method according to claim 1, wherein adding a volume of hydrogen to a volume of natural gas to provide a mixed gas stream includes providing a mixed gas stream with up to approximately 20% molar fraction of hydrogen.

25. A method of separating hydrogen from natural gas contained in a mixed gas stream, the method comprising:

cooling the mixed gas stream and liquefying a substantial portion of the volume of natural gas;

substantially separating the liquefied natural gas from the volume of hydrogen to provide a natural gas stream and a hydrogen stream; and

using at least one of the natural gas stream and the hydrogen stream to cool the mixed gas stream.

26. The method according to claim 25, further comprising compressing the mixed gas stream prior to cooling the mixed gas stream.

27. The method according to claim 26, wherein cooling the mixed gas stream includes flowing the mixed gas stream sequentially through a first heat exchanger and a second heat exchanger.

28. The method according to claim 27, wherein using at least one of the natural gas stream and the hydrogen stream to cool the mixed gas stream includes expanding the natural gas stream, expanding the hydrogen stream, and flowing both the expanded natural gas stream and the expanded hydrogen stream through the second heat exchanger.

29. The method according to claim 28, further comprising compressing the expanded natural gas stream.

30. The method according to claim 29, further comprising compressing the expanded hydrogen stream.

31. The method according to claim 28, further comprising controlling a purity of the hydrogen stream, at least in part, by selecting the pressure to which at least one of the hydrogen stream and the natural gas stream is expanded.

32. The method according to claim 31, further comprising expanding the at least one of the hydrogen stream and the natural gas stream to a pressure of approximately 65 pounds per square inch absolute (psia) or lower.

33. The method according to claim 25, further comprising expanding at least one of the hydrogen stream and the natural gas stream and controlling a purity of the hydrogen stream, at least in part, by selecting the pressure to which the at least one of the hydrogen stream and the natural gas stream is expanded.

34. The method according to claim 25, further comprising compressing the mixed gas stream prior to cooling the mixed gas stream and controlling a purity of the hydrogen stream, at least in part, by selecting the pressure to which the mixed gas stream is compressed.

35. The method according to claim 25, further comprising removing impurities from the natural gas.

36. The method according to claim 25, wherein adding a volume of hydrogen to a volume of natural gas to provide a mixed gas stream includes providing a mixed gas stream with up to approximately 20% molar fraction of hydrogen.

37. A system for separating hydrogen from natural gas contained in a mixed stream, the system comprising:

a first compressor;

a first heat exchanger;

a second heat exchanger;

a gas-liquid separator;

at least one expansion device;

a first flow path defined and configured to deliver a mixed gas stream sequentially through the first compressor, the second compressor, and into the gas-liquid separator; and

a second flow path defined and configured to deliver at least one of liquid natural gas and hydrogen gas from the gas liquid separator, through the at least one expansion device and through the second heat exchanger.

38. The system of claim 37, wherein the second flow path is defined and configured to deliver liquid natural gas from the gas liquid separator, through a first expansion device of the at least one expansion device and through the second

heat exchanger, and wherein the system further comprises a third flow path defined and configured to deliver hydrogen from the gas liquid separator, through a second expansion device of the at least one expansion device and through the second heat exchanger.

39. The system of claim 38, wherein first and second expansion devices each include Joule-Thomson valves.

40. The system of claim 38, further comprising a second compressor, and wherein the second flow path extends from the second heat exchanger to the second compressor.

41. The system of claim 40, further comprising a third compressor, and wherein the third flow path extends from the second heat exchanger to the third compressor.

42. The system of claim 41, further comprising a source of methanol coupled with the first flow path and configured to introduce a volume of methanol thereinto.

43. The system of claim 42, further comprising a separating system coupled with the first flow path and configured to remove water and methanol from the mixed stream of gas at a location between the first heat exchanger and the gas-liquid separator.

44. The system of claim 43, further comprising another separating system coupled with second flow path and configured to remove carbon dioxide from the liquid natural gas.

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