(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
International Bureau

(43) International Publication Date
12 June 2014 (12.06.2014)

WIPO | PCT

(10) International Publication Number
WO 2014/088732 A1

(51) International Patent Classification:
F25J 1/00 (2006.01)

(21) International Application Number:
PCT/US2013/067814

(22) International Filing Date:
31 October 2013 (31.10.2013)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:
61/733,304 4 December 2012 (04.12.2012) US
14/068,248 31 October 2013 (31.10.2013) US

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(81) Designated States *(unless otherwise indicated, for every kind of national protection available)*: AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BN, BR, BW, BY, BZ, CA, CH, CL, CN, CO, CR, CU, CZ, DE, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IR, IS, JP, KE, KG, KN, KP, KR, KZ, LA, LC, LK, LR, LS, LT, LU, LY, MA, MD, ME, MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PA, PE, PG, PH, PL, PT, QA, RO, RS, RU, RW, SA, SC, SD, SE, SG, SK, SL, SM, ST, SV, SY, TH, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, ZA, ZM, ZW.

(84) Designated States *(unless otherwise indicated, for every kind of regional protection available)*: ARIPO (BW, GH, GM, KE, LR, LS, MW, MZ, NA, RW, SD, SL, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, RU, TJ, TM), European (AL, AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV, MC, MK, MT, NL, NO, PL, PT, RO, RS, SE, SI, SK, SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, KM, ML, MR, NE, SN, TD, TG).

Published:

— *with international search report (Art. 21(3))*

(54) Title: USE OF ALTERNATE REFRIGERANTS IN OPTIMIZED CASCADE PROCESS

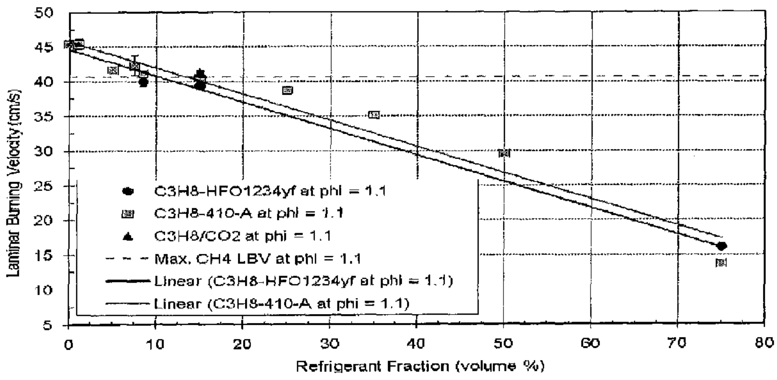

FIG. 1

(57) **Abstract**: Methods and systems for liquefying natural gas using nonflammable refrigerants are provided. Methods of liquefaction include cooling a natural gas stream via indirect heat exchange with a first nonflammable refrigerant selected from the group consisting of: difluoromethane, pentafluoromethane, trifluoromethane, hexafluoroethane, tetrafluoroethane, pentafluorethane, trifluoroethane, pentafluoroethane, any derivative thereof, and any combination thereof during a first refrigeration cycle; and cooling the natural gas stream via indirect heat exchange with a second refrigerant during a second refrigeration cycle.

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USE OF ALTERNATE REFRIGERANTS IN OPTIMIZED CASCADE PROCESS

FIELD OF THE INVENTION

[0001] The present invention relates generally to methods for liquefying natural gas. More particularly, but not by way of limitation, embodiments of the present invention include methods and systems for liquefying natural gas using nonflammable refrigerants.

BACKGROUND OF THE INVENTION

[0002] Natural gas is an important resource widely used as energy source or as industrial feedstock used in, for example, manufacture of plastics. Comprising primarily of methane, natural gas is a mixture of naturally occurring hydrocarbon gases and is typically found in deep underground natural rock formations or other hydrocarbon reservoirs. Other components of natural gas may include, but are not limited to, ethane, propane, carbon dioxide, nitrogen, and hydrogen sulfide. Typically, natural gas is transported from source to consumers through pipelines that physically connect reservoir to market.

[0003] Because natural gas is sometimes found in remote areas devoid of certain infrastructure (e.g., pipelines), alternative methods for transporting natural gas must be used. This situation commonly arises when the source of natural gas and the market are separated by great distances, for example, a large body of water. Bringing this natural gas from remote areas to market can have significant commercial value if the cost of transporting natural gas is minimized.

[0004] One alternative method of transporting natural gas involves converting natural gas into a liquefied form via liquefaction process. Because natural gas is gaseous under standard atmospheric conditions, it is typically subjected to thermodynamic processes in order to be liquefied. In its liquefied form, natural gas has a specific volume that is significantly lower than its specific volume in its gaseous form. Thus, the liquefaction process greatly increases the ease of transporting and storing natural gas, particularly in cases where pipelines are not available. For example, ocean liners carrying liquefied natural gas tanks can effectively link a natural gas source to a distant market when separated by an ocean.

[0005] Converting natural gas to its liquefied form can have other economic benefits as well. For example, storing liquefied natural gas (LNG) can help balance out periodic fluctuations in natural gas supply and demand. In particular, LNG can be more easily "stockpiled" for later use when natural gas demand is low and/or supply is high. As a result, future demand peaks can be met with LNG from storage, which can be vaporized as demand requires.

[0006] At least several conventional methods exist for liquefying natural gas. In one method, a propane pre-cooled mixed refrigerant is used to cool natural gas. The mixed refrigerant typically includes, but is not limited to, nitrogen, methane, ethane, and propane. In another method (e.g., optimized cascade process), natural gas is converted into LNG by utilizing multiple refrigerants in one or more mechanical refrigeration cycles that are used to lower the temperature of a natural gas stream. During the optimized cascade process, natural gas is first treated to remove contaminants including, but not limited to, CO_2, water, and mercury before entering the liquefaction section of an LNG plant. The treated gas is then chilled to approximately –260 °F in successively colder heat exchangers that use propane, ethylene, and methane as refrigerants. In some cases, the refrigerants are pure or substantially pure substances. In other cases, the refrigerants can be mixtures comprising more than one component. The product leaving the methane exchangers is LNG that is ready for storage. The LNG product is then pumped into insulated storage tanks before being loaded on special ships to be transported to LNG import terminals around the world.

[0007] While LNG and LNG facilities are generally considered safe there are certain inherent safety risks associated with hydrocarbon processing techniques. For example, conventional LNG refrigerants such as propane and ethylene are flammable materials. One potential catastrophic outcome arising from an accidental release of flammable materials is a vapor cloud explosion. Vapor cloud explosion can start the released flammable material forms a vapor cloud within a congested or confined area. Ignition of this cloud produces a flame front that accelerates through the congestion and creates a pressure wave. The severity of the pressure wave depends on several factors including, but not limited to, type of fuel released, size of the cloud within the congested/confined area, and degree of congestion/confinement within the cloud. As processing plants become more congested and confined, risk of explosion can increase. Most vapor cloud explosions have subsonic flame speeds and are classified as deflagrations. Even short-duration deflagrations can result in significant damage to buildings, equipments, and people. Potential damage is primarily a function of total amount of fuel burned, the maximum flame velocity that is achieved, and the manner in which the expansion of the combustion gases is contained.

[0008] Typically, LNG facilities are built in sufficiently open spaces in order to reduce the chances of a vapor cloud explosion in the unlikely case that flammable material is released. Other design considerations can also reduce the risk of explosion.

[0009] Moreover, recent expansion of LNG technology for offshore developments have prompted new studies analyzing safety risks of offshore LNG facilities. While these studies generally demonstrate that offshore LNG technology does not present unsafe risk levels, additional risk reduction efforts should always be considered.

BRIEF SUMMARY OF THE DISCLOSURE

[0010] The present invention relates generally to methods for liquefying natural gas. More particularly, but not by way of limitation, embodiments of the present invention include methods and systems for liquefying natural gas using nonflammable refrigerants.

[0011] One example of a method for liquefaction of natural gas comprises the steps of: a) cooling a natural gas stream via indirect heat exchange with a first nonflammable refrigerant selected from the group consisting of: difluoromethane, pentafluoromethane, trifluoromethane, hexafluoroethane, tetrafluoroethane, pentafluorethane, trifluoroethane, pentafluoroethane, any derivative thereof, and any combination thereof during a first refrigeration cycle; and b) cooling the natural gas stream via indirect heat exchange with a second refrigerant during a second refrigeration cycle.

[0012] Another example of a method for liquefaction of natural gas comprises the steps of: a) providing at least one nonflammable refrigerant selected from the group consisting of: difluoromethane, pentafluoroethane, trifluoromethane, hexafluoroethane, tetrafluoroethane, pentafluorethane, trifluoroethane, pentafluoroethane, any derivative thereof, and any combination thereof; and b) cooling a natural gas stream in an LNG facility via indirect heat exchange with the nonflammable refrigerant.

[0013] Yet another example of a method for liquefaction of natural gas comprises the steps of: a) cooling a natural gas stream in a LNG facility via indirect heat exchange with a first nonflammable refrigerant selected from the group consisting of: difluoromethane, pentafluoroethane, trifluoromethane, hexafluoroethane, tetrafluoroethane, pentafluorethane, trifluoroethane, pentafluoroethane, any derivative thereof, and any combination thereof during a first refrigeration cycle; b) cooling the natural gas stream in the LNG facility via indirect heat exchange with a second refrigerant during a second refrigeration cycle; and c) cooling the natural gas stream in the LNG facility via indirect heat exchange with a third refrigerant during the third refrigeration cycle.

BRIEF DESCRIPTION OF THE DRAWINGS

[0014] A more complete understanding of the present disclosure and advantages thereof may be acquired by referring to the follow description taken in conjunction with the accompanying figures, wherein:

[0015] Figure 1 is a plot summarizing effects of nonflammable refrigerants on laminar burning velocity as described in Example 1.

[0016] Figure 2 is a plot summarizing effects of nonflammable refrigerants on laminar burning velocity as described in Example 1.

[0017] While the present invention is susceptible to various modifications and alternative forms, specific exemplary embodiments thereof have been shown by way of example in the drawings and are herein described in detail. It should be understood, however, that the description herein of specific embodiments is not intended to limit the invention to the particular forms disclosed, but on the contrary, the intention is to cover all modifications, equivalents, and alternatives falling within the spirit and scope of the invention as defined by the appended claims.

DETAILED DESCRIPTION

[0018] The present invention relates generally to methods for liquefying natural gas. More particularly, but not by way of limitation, embodiments of the present invention include methods and systems for liquefying natural gas using nonflammable refrigerants.

[0019] There are certain inherent safety risks related to transporting and processing of LNG. These include, but are not limited to, hydrocarbon and LNG process and tanks, helicopter transportation, occupational, and ship collision. Certain risk evaluations indicate the general risk perspective that hydrocarbon (including LNG) processing and tanks can contribute to ~25–50% of the overall risk to personnel engaged in a large LNG operation. As most refrigerants used in large LNG operations are flammable, they can contribute to the overall safety risk to personnel. It is believed that the portion of risk attributed to flammable refrigerants can be on the order of 2-5%. In addition to the personnel risk, potential impacts of an explosion (e.g., vapor cloud explosion) arising from the use of flammable refrigerants include damage and/or loss of facility assets and major business interruption. While current use of flammable refrigerants are generally considered safe in the context of overall risk, there may be safer alternative refrigerants that can reduce inherent risks to personnel.

[0020] Various parameters can lessen or heighten the risk of a vapor cloud explosion. Some of the parameters affecting the risk of vapor cloud explosions include, but are not limited to, degree of congestion, degree of confinement, gas cloud size gas concentration, gas type (reactivity), ignition location, active mitigation measures, and the like. The risk of vapor cloud explosion may be lowered by addressing any one (e.g., gas type reactivity) or more of the parameters. Conventional refrigerants used during LNG process such as methane have relatively low reactivity while other conventional refrigerants have high reactivity (e.g., ethylene) or medium reactivity (e.g., propane). Fuels are typically considered low reactivity if their laminar burning velocities (LBVs) are lower than about 40 cm/s. Medium reactivity fuels typically have LBVs between about 40 to about 75 cm/s. High reactivity fuels have LBVs of greater than about 75 cm/s. In general, reactivity increases as LBV increases. Thus, lowering the reactivity of refrigerants used during LNG processes can lower the risk of vapor cloud explosion as well as the overall safety risk basis arising from LNG facilities and related activities.

[0021] Some of the approaches for reducing safety risks associated with LNG processes involve increasing equipment spacing or designing safety systems to mitigate fire and explosion risks. To date the use of nonflammable refrigerants, particularly in certain LNG processes (e.g., cascade LNG processes, floating LNG facilities, etc.) have been non-existent or limited. Alternative approaches involving nonflammable refrigerants are often limited by technical challenges. The physical properties of many nonflammable refrigerants differ from conventional LNG refrigerants (i.e., propane, ethylene, etc.) such that significant modifications to the design of LNG facilities may be required in order to achieve desirable operating efficiency. For example, use of nonflammable refrigerants may require higher operating pressures on the heavies removal unit. Other design modifications may involve addressing one or more of the following: possible combination of nonflammable refrigerant with physical solvent removal in upstream units; possible combination of nonflammable refrigerant with combustible refrigerants; novel compressor driver arrangement and higher methane compressor discharge pressures; and the like. There may also be considerable technical challenges in scaling the use of nonflammable refrigerants in LNG processes.

[0022] Reference will now be made in detail to embodiments of the invention, one or more examples of which are illustrated in the accompanying drawings. Each example is provided by way of explanation of the invention, not as a limitation of the invention. It will be apparent to those skilled in the art that various modifications and variations can be made in the present invention without departing from the scope or spirit of the invention. For instance, features illustrated or described as part of one embodiment can be used on another embodiment to yield a still further embodiment. Thus, it is intended that the present invention cover such modifications and variations that come within the scope of the invention.

[0023] The present invention provides compositions and methods related to lowering safety risk associated with LNG facilities and associated activities. In certain embodiments, a nonflammable refrigerant that is compatible with LNG processes is provided. The nonflammable refrigerant may be used in place of conventional LNG refrigerants (e.g., methane, propane, ethylene, etc.) or may be used in conjunction with conventional LNG refrigerants to form a refrigerant mixture. Other additives may be added to the refrigerant or refrigerant mixture as desired.

[0024] As compared to many conventional methods, advantages of certain embodiments of liquefying natural gas methods and systems described herein include, but are not limited to, one or more of the following:

- lower safety risks of significant fires and explosions,
- elimination of certain safety equipments,
- reduced spacing requirements,
- significant reduction in capital expenditure,
- well-suited for offshore or near populated areas where space is limited.

Other advantages will be apparent from the disclosure herein.

Cascade LNG

[0025] The present invention can be implemented in a process/facility used to cool natural gas to its liquefaction temperature, thereby producing LNG. The LNG process generally employs one or more refrigerants to extract heat from the natural gas and then reject the heat to the environment. Certain LNG processes may comprise multiple refrigerants. For example, a first refrigerant may be used to cool a first refrigeration cycle. A second refrigerant may be used to cool a second refrigeration cycle. A third refrigerant may be used to cool a third refrigeration cycle. As used herein, the terms “first”, “second”, and “third” refer to the relative position of the cycle with respect to each other. For example, the first refrigeration cycle is positioned just upstream of the second refrigeration cycle while the second refrigeration cycle is positioned upstream of the third refrigeration cycle and so forth. An optimized cascade LNG process typically utilizes propane, ethylene, and methane as the first, second, and third refrigerant respectively.

[0026] In one embodiment, the LNG process in accordance with one or more embodiments of the present invention employs a cascade-type refrigeration process that uses a plurality of multi-stage cooling cycles, each employing a different refrigerant composition, to sequentially cool the natural gas stream to lower and lower temperatures. In another embodiment, the LNG process is a mixed refrigerant process that employs a combination of two or more refrigerants to cool the natural gas stream in at least one cooling cycle.

[0027] Natural gas can be delivered to the LNG process at an elevated pressure in the range of from about 500 to about 3,000 pounds per square in absolute (psia), about 500 to about 1,000 psia, or 600 to 800 psia. Depending largely upon the ambient temperature, the temperature of the natural gas delivered to the LNG process can generally be in the range of from about 0 to about 180 °F (about -18 to about 82 °C), or about 20 to about 150 °F (about -7 to about 66 °C), or 60 to 125 °F (about 16 to about 52 °C).

[0028] While reference to a specific cascade LNG process comprising 3 refrigeration cycles involving 3 refrigerants is made, this is not intended to be limiting. It is recognized that a cascade LNG process involving more or less refrigerants and/or refrigeration cycles may be contemplated. Other variations to the cascade LNG process as well as alternative compatible LNG processes may also be contemplated.

[0029] In one embodiment, the present invention can be implemented in an LNG process that employs cascade-type cooling followed by expansion-type cooling. In such a liquefaction process, the cascade-type cooling may be carried out in a mechanical refrigeration cycle at an elevated pressure (e.g., about 650 psia) by sequentially passing the natural gas stream through first, second, and third refrigeration cycles employing respective first, second, and third refrigerants. In one embodiment, the first and second refrigeration cycles are closed refrigeration cycles, while the third refrigeration cycle is an open refrigeration cycle that utilizes a portion of the processed natural gas as a source of the refrigerant. Further, the third refrigeration cycle can include a multi-stage expansion cycle to provide additional cooling of the processed natural gas stream and reduce its pressure to near atmospheric pressure.

[0030] In the sequence of first, second, and third refrigeration cycles, the refrigerant having the highest boiling point can be utilized first, followed by a refrigerant having an intermediate boiling point, and finally by a refrigerant having the lowest boiling point. In one embodiment, the refrigerant can be a hydrocarbon-containing refrigerant. In another embodiment, the first refrigerant has a mid-boiling point at standard temperature and pressure (i.e., an STP mid-boiling point) within about 20, about 10, or 5 °F of the STP boiling point of pure propane. The first refrigerant can contain predominately propane, propylene, or mixtures thereof. The first refrigerant can contain at least about 75 mole percent propane, at least 90 mole percent propane, or can consist essentially of propane. In one embodiment, the second refrigerant has an STP mid-boiling point within about 20, about 10, or 5 °F of the STP boiling point of pure ethylene. The second refrigerant can contain predominately ethane, ethylene, or mixtures thereof. The second refrigerant can contain at least about 75 mole percent ethylene, at least 90 mole percent ethylene, or can consist essentially of ethylene. In one embodiment, the third refrigerant has an STP mid-boiling point within about 20, about 10, or 5 °F of the STP boiling point of pure methane. The third refrigerant can contain at least about 50 mole percent methane, at least about 75 mole percent methane, at least 90 mole percent methane, or can consist essentially of methane. At least about 50, about 75, or 95 mole percent of the third refrigerant can originate from the processed natural gas stream.

[0031] The first refrigeration cycle can cool the natural gas in a plurality of cooling stages/steps (e.g., two to four cooling stages) by indirect heat exchange with the first refrigerant. Each indirect cooling stage of the refrigeration cycles can be carried out in a separate heat exchanger. In the one embodiment, core-and-kettle heat exchangers are employed to facilitate indirect heat exchange in the first refrigeration cycle. After being cooled in the first refrigeration cycle, the temperature of the natural gas can be in the range of from about -45 to about -10 °F (about -43 to about -23 °C), or about -40 to about -15 °F (about -40 to about -26 °C), or about -20 to -30 °F (-29 to about -34 °C). A typical decrease in the natural gas temperature across the first refrigeration cycle may be in the range of from about 50 to about 210 °F (about 10 to about 99 °C), about 75 to about 180 °F (about 24 to about 82 °C), or about 100 to about 140 °F (about 38 to about 60 °C).

[0032] The second refrigeration cycle can cool the natural gas in a plurality of cooling stages/steps (e.g., two to four cooling stages) by indirect heat exchange with the second refrigerant. In one embodiment, the indirect heat exchange cooling stages in the second refrigeration cycle can employ separate, core-and-kettle heat exchangers. Generally, the temperature drop across the second refrigeration cycle can be in the range of from about 50 to about 180 °F (about 10 to about 82 °C), about 75 to about 150 °F (about 24 to about 66 °C), or about 100 to about 120 °F (about 38 to about 49 °C). In the final stage of the second refrigeration cycle, the processed natural gas stream can be condensed (i.e., liquefied) in major portion, preferably in its entirety, thereby producing a pressurized LNG-bearing stream. Generally, the process pressure at this location is only slightly lower than the pressure of the natural gas fed to the first stage of the first refrigeration cycle. After being cooled in the second refrigeration cycle, the temperature of the natural gas may be in the range of from about -205 to about -70 °F (about -132 to about -57 °C), about -175 to about -95 °F (about -115 to about -71 °C), or about -140 to about -125 °F (about -96 to about -87 °C).

[0033] The third refrigeration cycle can include both an indirect cooling section and an expansion-type cooling section. To facilitate indirect heat exchange, the third refrigeration cycle can employ at least one brazed-aluminum plate-fin heat exchanger. The total amount of cooling provided by indirect heat exchange in the third refrigeration cycle can be in the range of from about 5 to about 60 °F, about 7 to about 50 °F, or 10 to 40 °F.

[0034] The expansion-type cooling section of the third refrigeration cycle can further cool the pressurized LNG-bearing stream via sequential pressure reduction to approximately atmospheric pressure. Such expansion-type cooling can be accomplished by flashing the LNG-bearing stream to thereby produce a two-phase vapor-liquid stream. When the third refrigeration cycle is an open refrigeration cycle, the expanded two-phase stream can be subjected to vapor-liquid separation and at least a portion of the separated vapor phase (i.e., the flash gas) can be employed as the third refrigerant to help cool the processed natural gas stream. The expansion of the pressurized LNG-bearing stream to near atmospheric pressure can be accomplished by using a plurality of expansion steps (i.e., two to four expansion steps) where each expansion step is carried out using an expander. Suitable expanders include, for example, either Joule-Thomson expansion valves or hydraulic expanders. In one embodiment, the third stage refrigeration cycle can employ three sequential expansion cooling steps, wherein each expansion step can be followed by a separation of the gas-liquid product. Each expansion-type cooling step can further cool the LNG-bearing stream in the range of from about 10 to about 60 °F, about

15 to about 50 °F, or 25 to 35 °F. The reduction in pressure across the first expansion step can be in the range of from about 80 to about 300 psia, about 130 to about 250 psia, or 175 to 195 psia. The pressure drop across the second expansion step can be in the range of from about 20 to about 110 psia, about 40 to about 90 psia, or 55 to 70 psia. The third expansion step can further reduce the pressure of the LNG-bearing stream by an amount in the range of from about 5 to about 50 psia, about 10 to about 40 psia, or 15 to 30 psia. The liquid fraction resulting from the final expansion stage is an LNG product. Generally, the temperature of the LNG product can be in the range of from about -200 to about -300 °F (-129 to about -184 °C), about -225 to about -275 °F (about -143 to about -170 °C), or about -240 to about -260 °F (about -151 to about -162 °C). The pressure of the LNG product can be in the range of from about 0 to about 40 psia, about 10 to about 20 psia, or 12.5 to 17.5 psia.

[0035] The natural gas feed stream to the LNG process will usually contain such quantities of C2+ components so as to result in the formation of a C2+ rich liquid in one or more of the cooling stages of the second refrigeration cycle. Generally, the sequential cooling of the natural gas in each cooling stage is controlled so as to remove as much of the C2 and higher molecular weight hydrocarbons as possible from the gas, thereby producing a vapor stream predominating in methane and a liquid stream containing significant amounts of ethane and heavier components. This liquid can be further processed via gas-liquid separators employed at strategic locations downstream of the cooling stages. In one embodiment, one objective of the gas/liquid separators is to maximize the rejection of the C5+ material to avoid freezing in downstream processing equipment. The gas/liquid separators may also be utilized to vary the amount of C2 through C4 components that remain in the natural gas product to affect certain characteristics of the finished LNG product.

[0036] The exact configuration and operation of gas-liquid separators may be dependant on a number of parameters, such as the C2+ composition of the natural gas feed stream, the desired BTU content of the LNG product, the value of the C2+ components for other applications, and other factors routinely considered by those skilled in the art of LNG plant and gas plant operation. In one embodiment of the present invention, the C2+ hydrocarbon stream or streams may be demethanized via a single stage flash or a fractionation column. The gaseous methane-rich stream can be directly returned at pressure to the liquefaction process. The resulting heavies-rich liquid stream may then be subjected to fractionation in one or more fractionation zones to produce individual streams rich in specific chemical constituents (e.g., C2, C3, C4, and C5+).

[0037] The nonflammable refrigerants of the present invention may be used during liquefaction of natural gas. Refrigerants utilized in cascade-type refrigeration processes can have successively lower boiling points in order to maximize heat removal from the natural gas stream being liquefied. Additionally, cascade-type refrigeration processes can include some level of heat integration. For example, a cascade-type refrigeration process can cool one or more refrigerants having a higher volatility via indirect heat exchange with one or more refrigerants having a lower volatility. In addition to cooling the natural gas stream via indirect heat exchange with one or more refrigerants, cascade and mixed-refrigerant LNG systems can employ one or more expansion cooling stages to simultaneously cool the LNG while reducing its pressure to near atmospheric pressure. In some embodiments, the nonflammable refrigerant may be used in a floating LNG (FLNG) process. In one or more embodiments, the nonflammable refrigerant may be used in an optimized cascade LNG process.

Refrigerants

[0038] A refrigerant is a substance used in a heat cycle, which can undergo a reversible phase transition from a liquid to a gas during an LNG process. A nonflammable refrigerant according to one or more embodiments generally comprises simple fluorohydrocarbons that are nonflamamble. In some embodiments, the nonflammable refrigerant may include, but not limited to, one or more of the following: difluoromethane (sometimes referred to as “R-32”), pentafluoroethane (sometimes referred to as “R-125” or “FE-25”), trifluoromethane (sometimes referred to as “R-23” or “FE-13”), hexafluoroethane (sometimes referred to as “R-116”), derivatives thereof, and mixtures thereof. Other suitable examples of fluorohydrocarbons may include, but are not limited to, difluoropropane, trifluoropropane, tetrafluoropropane, pentafluoropropane, and the like. The hydrocarbon portion of the fluorohydrocarbon may comprise one or more carbons. In some embodiments, the fluorohydrocarbon may comprise between one carbon to about ten carbons. In those embodiments, the fluorohydrocarbon may comprise one or more fluorines.

[0039] In some embodiments, the nonflammable refrigerant may comprise a mixture of difluoromethane and pentafluoroethane. A 1:1 mixture (by weight) of difluoromethane and pentafluoroethane is sometimes referred to as "R-410A" and has a boiling point of -55.3 °F (-48.5 °C). In another example, the nonflammable refrigerant may comprise a mixture of trifluoromethane and hexafluoroethane. A 46:54 mixture (by weight) of trifluoromethane and hexafluoroethane is sometimes referred to as "R-508B" and has a boiling of -126.94 °F (-88.3 °C). In those embodiments comprising a mixture of nonflammable refrigerants, each nonflammable refrigerant may be present in about 0.1% up to about 100% by weight. The specific amount of nonflammable refrigerant present in the mixture may be modified by one of ordinary skill in the art as desired. Such modifications may depend on a number of factors including, but not limited to, desired boiling point, cost, availability, and desired maximum LBV to minimize or eliminate the possibility of ignition and/or reduce the overpressure from an ignited vapor cloud. In one or more embodiments, the nonflammable refrigerant may comprise at least one selected from the group consisting of: R-410A, R-508B, R-23, R-125, and any combination thereof. In some embodiments, the nonflammable refrigerant may be an azeotropic mixture. In other embodiments, the nonflammable refrigerant may be a zeotropic mixture or near-azeotropic mixture.

[0040] In some embodiments, the nonflammable refrigerant is substantially free of hydrocarbons. In other embodiments, the nonflammable refrigerant includes a hydrocarbon component in an amount ranging from about 0.1% to about 99.9% by volume or about 0.1% to about 99.9% by volume. In such embodiments, the hydrocarbon may be selected from the group consisting of: ethylene, propane, methane, and any combination thereof.

[0041] In accordance with one or more embodiments, a nonflammable refrigerant comprising a mixture of difluoromethane and pentafluoroethane (e.g., R-410A) may be used as the first refrigerant of an optimized cascade LNG process. In other embodiments, R-134a (1,1,1,2-tetrafluoroethane), R-125 (pentafluoroethane), R-404a (a blend of 52 wt% trifluoroethane, 44 wt% R-125, and 4 wt% R-134a), or combinations thereof may be used as the first refrigerant. In accordance with one or more embodiments, a nonflammable refrigerant comprising a mixture of trifluoromethane and hexafluoroethane (e.g., R-508B) may be used as the second refrigerant of an optimized cascade LNG process. When a nonflammable refrigerant is used in place of a flammable refrigerant (e.g., ethylene, propane), the chance of fire and explosion in an LNG facility is reduced. In one aspect, the chance of deflagration to detonation transition may be reduced at least in part due to the reduction in equipment spacing. The use of nonflammable refrigerants also allows greater flexibility in design LNG processes. For example, personnel quarters of a floating LNG facility may be located closer to a nonflammable refrigerant loop which reduces plot spacing and allows the nonflammable refrigerant to be used as a utility for other facility spaces. Moreover, mixtures of difuloromethane and pentafluoroethane (e.g., R-410A) condense at a lower temperature than propane which reduces energy and capital requirement for the LNG process.

[0042] In some embodiments, difluoromethane, pentafluoroethane, or both may be added to a flammable refrigerant (e.g., propane) as an additive that can reduce the flame speed of the flammable refrigerant. In some embodiments, the flame speed may be reduced below deflagration to detonation transition. In some embodiments, the flame speed may be reduce to less than or about the flame speed of methane (see Example 2).

[0043] The use of trifluoromethane and hexafluoroethane and other nonflammable refrigerants may require modifications to conventional optimized cascade LNG processes. For example, such use may require increased pressure in heavies removal column which causes the column to operate in retrograde condensation region. Furthermore, this increased pressure may require, for example, a booster motor for methane compression system or other rearrangement of gas turbine generator drivers. Methane compressor discharge pressures are typically limited to a value lower than that required for the use of trifluoromethane and hexafluoromethane. Thus, it may be desirable to use a physical solvent and include the heavies removal column upstream. Moreover, since the mixture of trifluoromethane and hexafluoromethane condenses at a lower pressure than ethylene, a reduced driving force will reduce refrigerant loss through compressor seals.

[0044] In some embodiments, at least one of trifluoromethane and hexafluoroethane may be added to a flammable refrigerant (e.g., ethylene) as an additive which can reduce the flame speed of the flammable refrigerant. In some embodiments, the flame speed may be reduced below deflagration to detonation transition. In some embodiments, the flame speed may be reduce to less than or about the flame speed of methane (~0.4 meters per second). The flame speed of ethylene is about 0.75 meters per second.

[0045] The following examples of certain embodiments of the invention are given. Each example is provided by way of explanation of the invention, one of many embodiments of the invention, and the following examples should not be read to limit, or define, the scope of the invention.

Example 1

[0046] In this Example, laminar burning velocities for various compounds were measured. The tests were performed using a constant-volume vessel in which gases were introduced using a partial pressure method, allowed to mix, and ignited. The resulting dynamic pressure trace was analyzed in order to calculate the LBV.

[0047] The measured LBV values are summarized in Table 1 below. The reactivity of a compound increases as LBV increases. This reactivity is typically a function of the strength of hydrogen bonding in the compound and not the heat of combustion or the thermal unit (e.g., BTU) value.

Table 1

Compound	LBV (m/s)
Methane	~ 0.40
Propane	~ 0.46
Ethylene	~ 0.75
Acetylene	~ 1.55
Hydrogen	~ 3.25

[0048] FIG. 1 illustrates the results of an experiment in which various nonflammable refrigerants and compounds such as hydrofluoro-olefin (HFO1234yf), R-410A, and CO_2 were added to propane. As the nonflammable refrigerant fraction increases, the laminar burning velocity decreases.

[0049] FIG. 2 illustrates the results of an experiment in which various nonflammable refrigerants (R-508B and R-23) were added to ethylene. As the nonflammable refrigerant fraction increases, the laminar burning velocity decreases. FIGS.1 and 2 illustrate that nonflammable refrigerants may be added to flammable refrigerants to significantly reduce laminar burning velocity of flammable refrigerants. Such plots may be used to determine mixtures having desired LBV values.

Example 2

[0050] Certain characteristics of an optimized cascade process improved when R-410A was used as a nonflammable refrigerant in place of propane. These characteristics were simulated using REFPROP (version 8) in the Aspen Physical Property System and verified against National Institute of Standards and Technology (NIST) tables. For example, the actual air compressor capacity (ACFM) decreased by 10-30% which allows for a smaller compressor bundle. R-410A also provides a greater vapor density (69.4 kg/m^3) as compared to propane (25.6 kg/m^3) which permits higher system mass flow and reduces pressure drop losses, allowing smaller diameter piping and smaller equipment to obtain the same refrigerant duty achieved by propane. Table 2 below summarizes simulated properties of R410A using Aspen HYSYS®, a modeling software available through Aspentech Technology, Inc. (Burlington, Massachusetts). These results suggest that R410A may be stable and behaves predictably throughout the given temperature range.

Table 2.

	R410A modeling in HYSYS®								
Temp-erature [F]	Vapor Pressure [psia]	Phase Mass Density (Liquid Phase) [lb/ft3]	Phase Mass Density (Vapor Phase) [lb/ft3]	Phase Mass Enthalpy (Liquid Phase) [Btu/lb]	Phase Mass Enthalpy (Vapor Phase) [Btu/lb]	Phase change delta H (Btu/lb)	Phase Mass Entropy (Liquid Phase) [Btu/lb-F]	Phase Mass Entropy (Vapor Phase) [Btu/lb-F]	Phase change delta S (Btu/lb -F)
-40	-0.94	-0.06	-0.48		0.00	0.00		0.00	0.00
-35	-0.64	-0.06	-0.41	1.02	-0.02	-0.04	0.92	0.00	0.00
-30	-0.60	-0.07	-0.33	1.02	-0.05	-0.08	1.03	-0.02	0.00
-25	-0.67	-0.07	-0.26	0.99	-0.09	-0.14	0.99	-0.06	0.00
-20	-0.52	-0.06	-0.20	0.96	-0.05	-0.11	0.98	-0.07	0.00
-15	-0.60	-0.07	-0.14	0.95	-0.11	-0.20	0.94	-0.07	0.00
-10	-0.41	-0.06	-0.08	0.95	-0.10	-0.20	0.94	-0.09	0.00
-5	-0.33	-0.07	-0.01	0.93	-0.10	-0.21	0.93	-0.13	0.00
0	-0.29	-0.07	0.01	0.93	-0.11	-0.24	0.89	-0.11	0.00
5	-0.26	-0.07	0.05	0.86	-0.13	-0.28	0.88	-0.12	0.00
10	-0.31	-0.08	0.16	0.87	-0.17	-0.34	0.88	-0.16	0.00
15	-0.28	-0.07	0.14	0.84	-0.13	-0.32	0.87	-0.15	0.00
20	-0.16	-0.08	0.21	0.83	-0.20	-0.41	0.84	-0.17	0.00
25	-0.14	-0.08	0.26	0.84	-0.20	-0.43	0.84	-0.18	0.00
30	-0.18	-0.09	0.29	0.83	-0.21	-0.47	0.84	-0.22	0.00
35	-0.14	-0.10	0.34	0.84	-0.16	-0.44	0.83	-0.21	-0.01
40	-0.11	-0.11	0.36	0.79	-0.22	-0.52	0.82	-0.24	-0.01
45	-0.06	-0.12	0.40	0.82	-0.21	-0.54	0.81	-0.23	-0.01
50	-0.04	-0.12	0.42	0.80	-0.22	-0.58	0.82	-0.26	-0.01
55	-0.02	-0.14	0.43	0.82	-0.26	-0.66	0.81	-0.25	-0.01
60	0.01	-0.15	0.45	0.81	-0.24	-0.67	0.82	-0.28	-0.01
65	0.02	-0.17	0.49	0.81	-0.25	-0.71	0.82	-0.28	-0.01
70	0.03	-0.19	0.52	0.82	-0.29	-0.81	0.83	-0.32	-0.01
75	0.05	-0.22	0.53	0.83	-0.28	-0.84	0.82	-0.33	-0.01
80	0.05	-0.23	0.55	0.83	-0.30	-0.92	0.82	-0.35	-0.01
85	0.06	-0.25	0.55	0.83	-0.29	-0.94	0.83	-0.35	-0.01
90	0.04	-0.28	0.57	0.84	-0.31	-1.03	0.84	-0.36	-0.01
95	0.05	-0.33	0.60	0.84	-0.39	-1.22	0.83	-0.41	-0.01
100	0.08	-0.35	0.65	0.82	-0.36	-1.21	0.88	-0.43	-0.02
105	0.07	-0.38	0.67	0.84	-0.38	-1.33	0.85	-0.45	-0.02
110	0.07	-0.42	0.70	0.84	-0.47	-1.57	0.86	-0.50	-0.02
115	0.06	-0.45	0.73	0.83	-0.47	-1.67	0.84	-0.51	-0.02
120	0.07	-0.50	0.78	0.83	-0.49	-1.82	0.80	-0.55	-0.02
125	0.07	-0.56	0.83	0.82	-0.53	-2.02	0.85	-0.57	-0.02
130	0.06	-0.63	0.92	0.84	-0.63	-2.40	0.84	-0.59	-0.03
135	0.06	-0.74	1.03	0.86	-0.65	-2.69	0.90	-0.67	-0.03

140	0.05	-0.92	1.12	0.91	-0.73	-3.24	0.91	-0.69	-0.04

DEFINITIONS

[0051] As used herein, “confinement” and related terms refer to the presence of obstructions that prevent flame propagation in any one of three directions (x, y, or z directions). Objects may be confined in one dimension, two dimensions, or three dimensions.

[0052] As used herein, “congestion” and related terms refer to the presence of obstacles that cause a flame front to flow around the obstacles thus generating turbulence and accelerating the flame front. More specifically, the terms “low congestion”, “medium congestion”, and “high congestion” may be a context dependent term. For example, “low congestion” may be defined as having about 15% or less area blockage ratio (ABR) and a pitch of greater than about 8D. In some embodiments, “low congestion” may refer to an area that is easy to walk through relatively unimpeded. The term “medium congestion” may refer to an area having between about 15% to about 30% ABR and a pitch of about 4D to about 8D. In some embodiments, “medium congestion” may refer to an area that can be walked through but requires taking an indirect path. The term “high congestion” may refer to an area having more than about 30% ABR and a pitch of less than about 4D. In some embodiments, “high congestion” may refer to an area that cannot be walked through.

[0053] As used herein, the term “area blockage ratio” refers to the ratio of the volume of congestion to the total volume available.

[0054] As used herein, the term “pitch” refers to the distance between rows of repeated congestion obstacles. Pitch is oftentimes measured as a multiple of the average congestion diameter (i.e., 8D = 8 diameter lengths).

[0055] As used herein, the term “risk” refers to the probability and the consequence of an accidental event.

[0056] As used herein, the term “azeotropic mixture” refers to a mixture made up of two or more refrigerants with similar boiling points that acts as a single fluid. The components of an azeotropic mixture typically do not separate under normal operating conditions and can be charged as a vapor or liquid.

[0057] As used herein, the term "near-azeotropic mixture" refers to a mixture made up of two or more refrigerants with different boiling points that, when in a totally liquid or vapor state, act as one component. However, when changing from vapor to liquid or liquid to vapor, the individual refrigerants evaporate or condense at different temperatures.

[0058] As used herein, the term "derivative" refers to a compound that is derived from a similar compound.

[0059] In closing, it should be noted that the discussion of any reference is not an admission that it is prior art to the present invention, especially any reference that may have a publication date after the priority date of this application. At the same time, each and every claim below is hereby incorporated into this detailed description or specification as a additional embodiments of the present invention.

[0060] Although the systems and processes described herein have been described in detail, it should be understood that various changes, substitutions, and alterations can be made without departing from the spirit and scope of the invention as defined by the following claims. Those skilled in the art may be able to study the preferred embodiments and identify other ways to practice the invention that are not exactly as described herein. It is the intent of the inventors that variations and equivalents of the invention are within the scope of the claims while the description, abstract and drawings are not to be used to limit the scope of the invention. The invention is specifically intended to be as broad as the claims below and their equivalents.

REFERENCES

[0061] All of the references cited herein are expressly incorporated by reference. The discussion of any reference is not an admission that it is prior art to the present invention, especially any reference that may have a publication data after the priority date of this application. Incorporated references are listed again here for convenience:

1. US 7,849,691
2. US 20100122551
3. US 20100281915

CLAIMS

1. A method for liquefaction of natural gas comprising:

a) cooling a natural gas stream via indirect heat exchange with a first nonflammable refrigerant selected from the group consisting of: difluoromethane, pentafluoromethane, trifluoromethane, hexafluoroethane, tetrafluoroethane, pentafluorethane, trifluoroethane, pentafluoroethane, any derivative thereof, and any combination thereof during a first refrigeration cycle; and

b) cooling the natural gas stream via indirect heat exchange with a second refrigerant during a second refrigeration cycle.

2. The method of claim 1 wherein the first refrigerant or the second refrigerant includes a hydrocarbon component in an amount ranging from about 0.1% to about 99% by volume.

3. The method of claim 2 wherein the hydrocarbon is selected from the group consisting of: ethylene, propane, and any combination thereof.

4. The method of claim 1 wherein the first refrigerant or the second refrigerant is an azeotropic mixture or a near-azeotropic mixture.

5. The method of claim 1 wherein the first refrigerant is selected from a group consisting of: a 1:1 mixture of difluoromethane and pentafluoromethane; pentafluoroethane; a mixture of trifluoroethane, pentafluoroethane and tetrafluoroethane; and any derivative thereof.

6. The method of claim 1 wherein the second refrigerant is a nonflammable refrigerant comprising a mixture of trifluoromethane and hexafluoroethane.

7. The method of claim 1, further comprising:

c) producing liquefied natural gas.

8. The method of claim 7, further comprising:

d) vaporizing the liquefied natural gas.

9. A method for liquefaction of natural gas comprising:

a) providing at least one nonflammable refrigerant selected from the group consisting of: difluoromethane, pentafluoroethane, trifluoromethane, hexafluoroethane, tetrafluoroethane, pentafluorethane, trifluoroethane, pentafluoroethane, any derivative thereof, and any combination thereof; and

b) cooling a natural gas stream in an LNG facility via indirect heat exchange with the nonflammable refrigerant.

10. The method of claim 9 wherein the LNG facility is located offshore.

11. The method of claim 9 wherein the refrigerant includes a hydrocarbon component in an amount ranging from about 0.1% to about 99% by volume.

12. The method of claim 11 wherein the hydrocarbon is selected from the group consisting of: ethylene, propane, and any combination thereof.

13. The method of claim 9 wherein at least three nonflammable refrigerants are used to cool the natural gas stream and produce liquefied natural gas.

14. The method of claim 13 wherein the liquefied natural gas is vaporized.

15. The method of claim 9 wherein a first nonflammable refrigerant is used to cool the natural gas stream in a closed-loop refrigeration cycle and second nonflammable refrigerant is used to cool the natural gas stream in an open-loop refrigeration cycle.

16. The method of claim 9 wherein the refrigerant is an azeotropic or near-azeotropic mixture.

17. A method for liquefaction of natural gas comprising:

a) cooling a natural gas stream in a LNG facility via indirect heat exchange with a first nonflammable refrigerant selected from the group consisting of: difluoromethane, pentafluoroethane, trifluoromethane, hexafluoroethane, tetrafluoroethane, pentafluorethane, trifluoroethane, pentafluoroethane, any derivative thereof, and any combination thereof during a first refrigeration cycle;

b) cooling the natural gas stream in the LNG facility via indirect heat exchange with a second refrigerant during a second refrigeration cycle; and

c) cooling the natural gas stream in the LNG facility via indirect heat exchange with a third refrigerant during the third refrigeration cycle.

18. The method of claim 17 wherein the first refrigerant is selected from a group consisting of: a 1:1 mixture of difluoromethane and pentafluoromethane; pentafluoroethane; a mixture of trifluoroethane, pentafluoroethane and tetrafluoroethane; and any derivative thereof.

19. The method of claim 17 wherein the second refrigerant is a mixture of trifluoromethane and hexafluoroethane.

20. The method of claim 17 wherein the third refrigerant is methane.

21. The method of claim 17 wherein the first refrigerant or the second refrigerant includes a hydrocarbon component in an amount ranging from about 0.1% to about 99% by volume.

22. The method of claim 21 wherein the hydrocarbon is selected from the group consisting of: ethylene, propane, and any combination thereof.

23. The method of claim 17 wherein the LNG facility is located offshore.

24. The method of claim 17, further comprising:
d) producing liquefied natural gas.

25. The method of claim 24, further comprising:
e) vaporizing the liquefied natural gas.

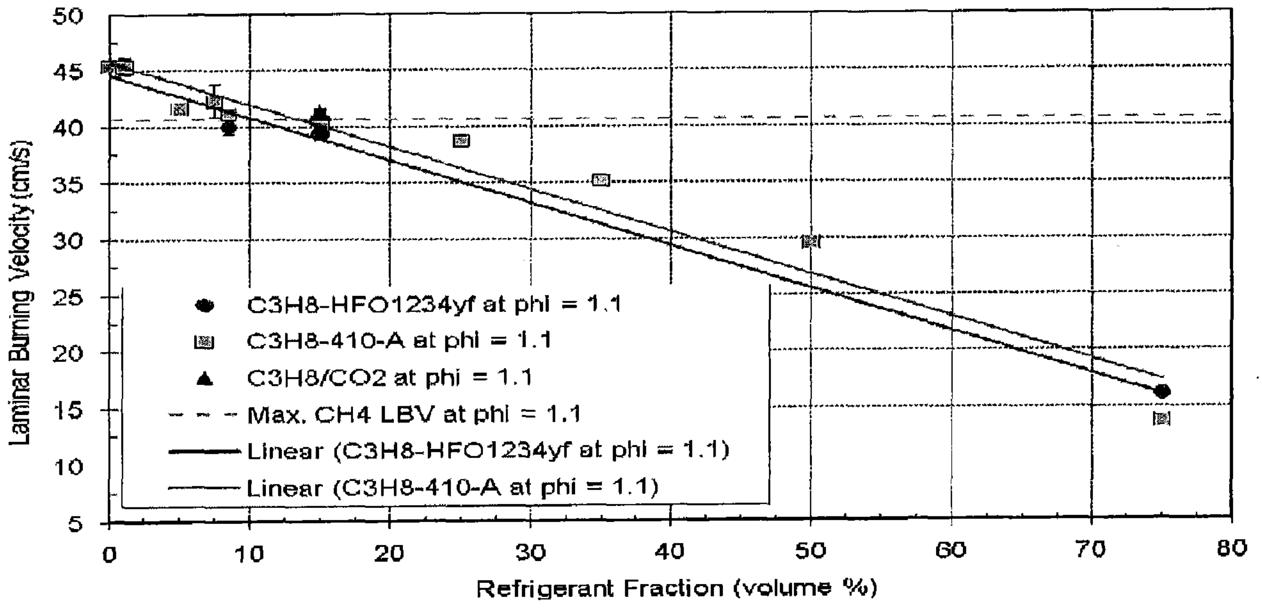
Laminar Burning Velocity (cm/s)
50
45
40
35
30
25
20
15
10
5
0
10
20
30
40
50
60
70
80
Refrigerant Fraction (volume %)
● C3H8-HFO1234yf at phi = 1.1
▨ C3H8-410-A at phi = 1.1
▲ C3H8/CO2 at phi = 1.1
Max. CH4 LBV at phi = 1.1
Linear (C3H8-HFO1234yf at phi = 1.1)
Linear (C3H8-410-A at phi = 1.1)

FIG. 1

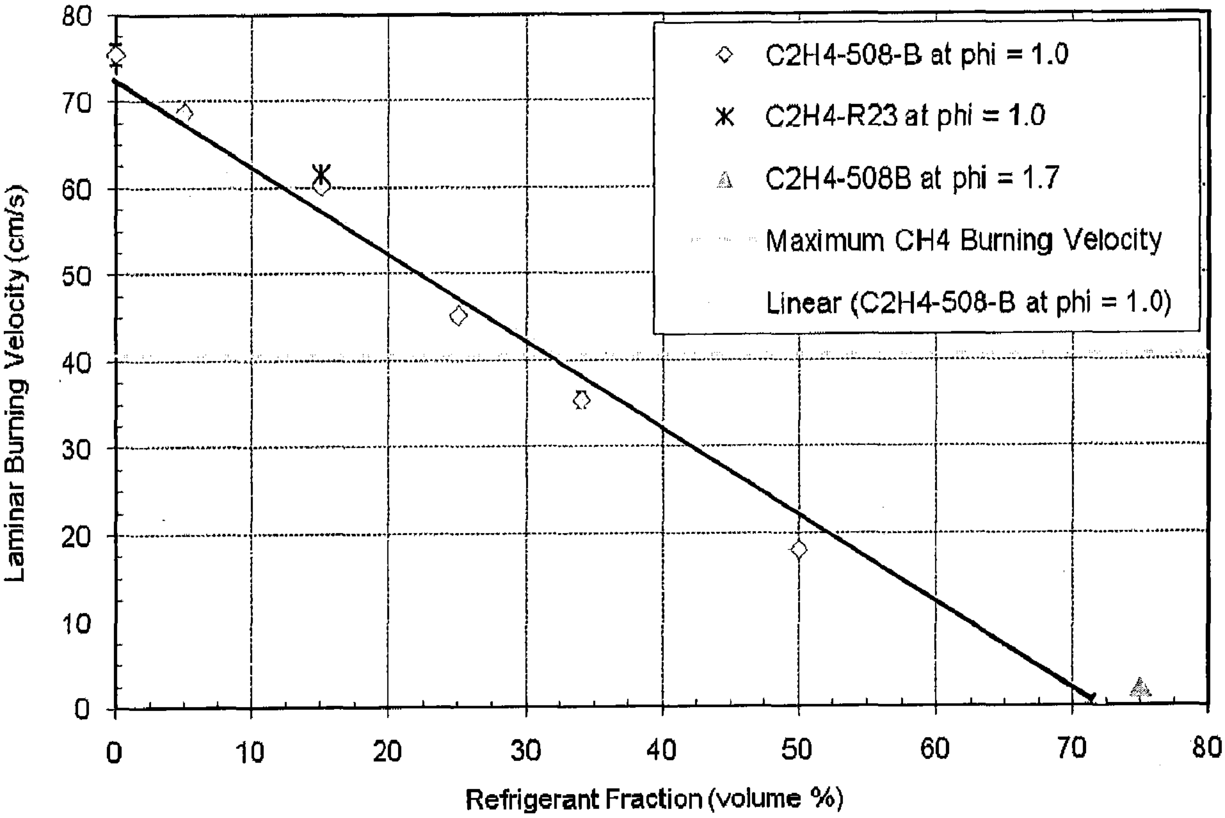
◇ C2H4-508-B at phi = 1.0
✳ C2H4-R23 at phi = 1.0
△ C2H4-508B at phi = 1.7
Maximum CH4 Burning Velocity
Linear (C2H4-508-B at phi = 1.0)
Laminar Burning Velocity (cm/s)
80
70
60
50
40
30
20
10
0
0
10
20
30
40
50
60
70
80
Refrigerant Fraction (volume %)

FIG. 2

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US2013/067814

A. CLASSIFICATION OF SUBJECT MATTER
IPC(8) - F25J 1/00 (2014.01)
USPC - 62/611
According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC(8) - F25J 1/00, 1/02 (2014.01)
USPC - 62/611, 612, 613

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched
CPC - F25J 1/00, 1/0022, 1/0025, 1/0211 (2013.01)

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

Orbit, Google Scholar

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 2010/0281915 A1 (ROBERTS et al) 11 November 2010 (11.11.2010) entire document	1-5, 7, 9-12, 16
Y		6, 8, 13-15, 17-25
Y	US 6,357,257 B1 (GOBLE JR et al) 19 March 2002 (19.03.2002) entire document	6, 19
Y	US 6,564,579 B1 (MCCARTNEY) 20 May 2003 (20.05.2003) entire document	8, 14, 25
Y	US 2004/0182108 A1 (ROBERTS) 23 September 2004 (23.09.2004) entire document	13, 14, 17-25
Y	US 2010/0058803 A1 (RANSBARGER et al) 11 March 2010 (11.03.2010) entire document	15

☐ Further documents are listed in the continuation of Box C. ☐

* Special categories of cited documents:
"A" document defining the general state of the art which is not considered to be of particular relevance
"E" earlier application or patent but published on or after the international filing date
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
"O" document referring to an oral disclosure, use, exhibition or other means
"P" document published prior to the international filing date but later than the priority date claimed
"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"&" document member of the same patent family

Date of the actual completion of the international search 21 February 2014	Date of mailing of the international search report 27 MAR 2014
Name and mailing address of the ISA/US Mail Stop PCT, Attn: ISA/US, Commissioner for Patents P.O. Box 1450, Alexandria, Virginia 22313-1450 Facsimile No. 571-273-3201	Authorized officer: Blaine R. Copenheaver PCT Helpdesk: 571-272-4300 PCT OSP: 571-272-7774