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(54) **ACTIVE GAS REGENERATIVE LIQUEFIER SYSTEM AND METHOD**

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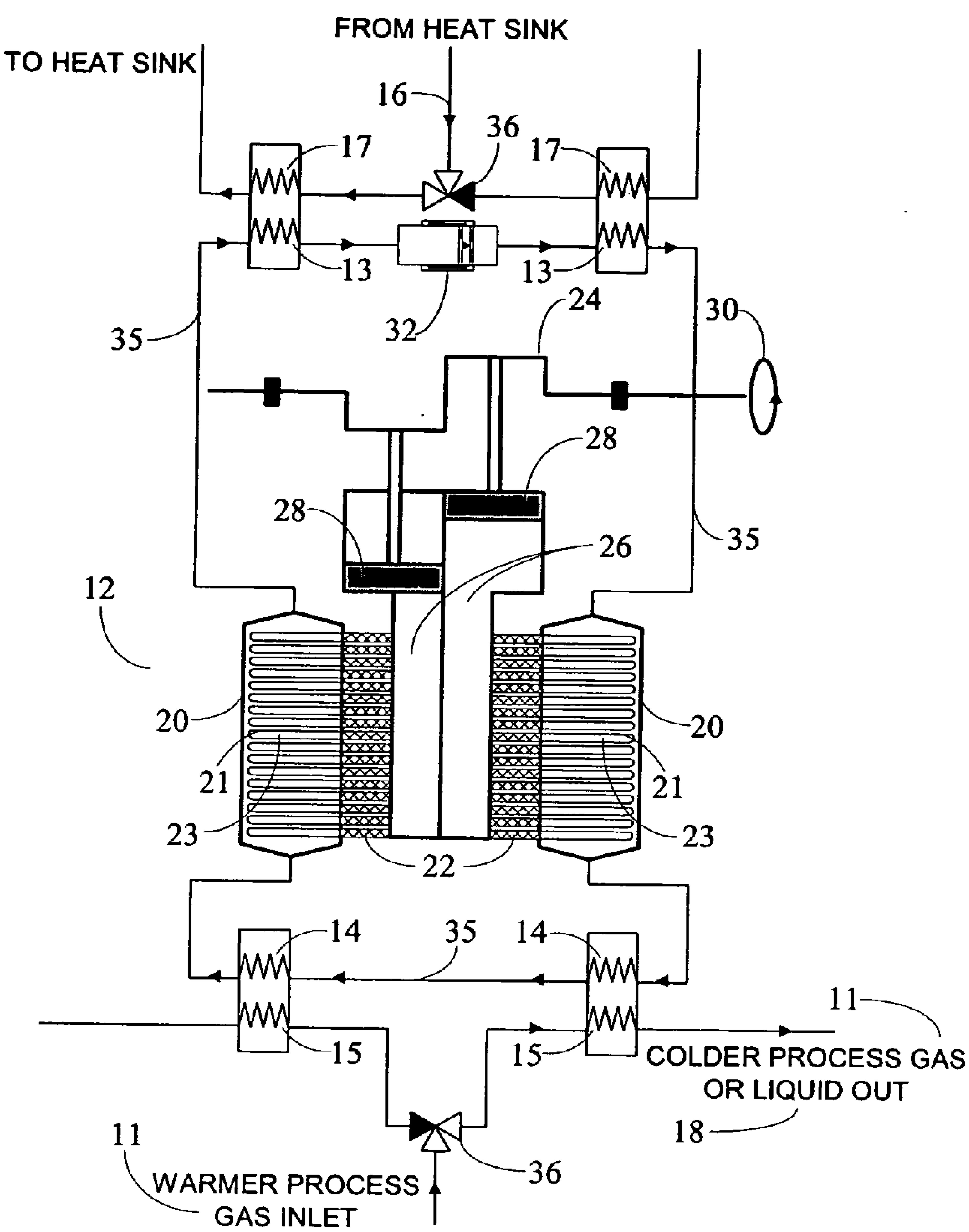
(57) **ABSTRACT**

The present invention provides an active gas regenerative liquefier (AGRL) for efficiently cooling and liquefying a process stream based on the combination of several active gas regenerative refrigerator (AGRR) stages configured to sequentially cool and liquefy the process stream, e.g. natural gas or hydrogen. In specific embodiments, the individual AGRR stages include heat exchangers, dual active regenerators, and a compressor/expander assembly, configured to recover a portion of the work of compression of a refrigerant by simultaneously expanding a refrigerant in one portion of the device while compressing the refrigerant in another portion to effect cooling of a heat transfer fluid, and ultimately the process stream.



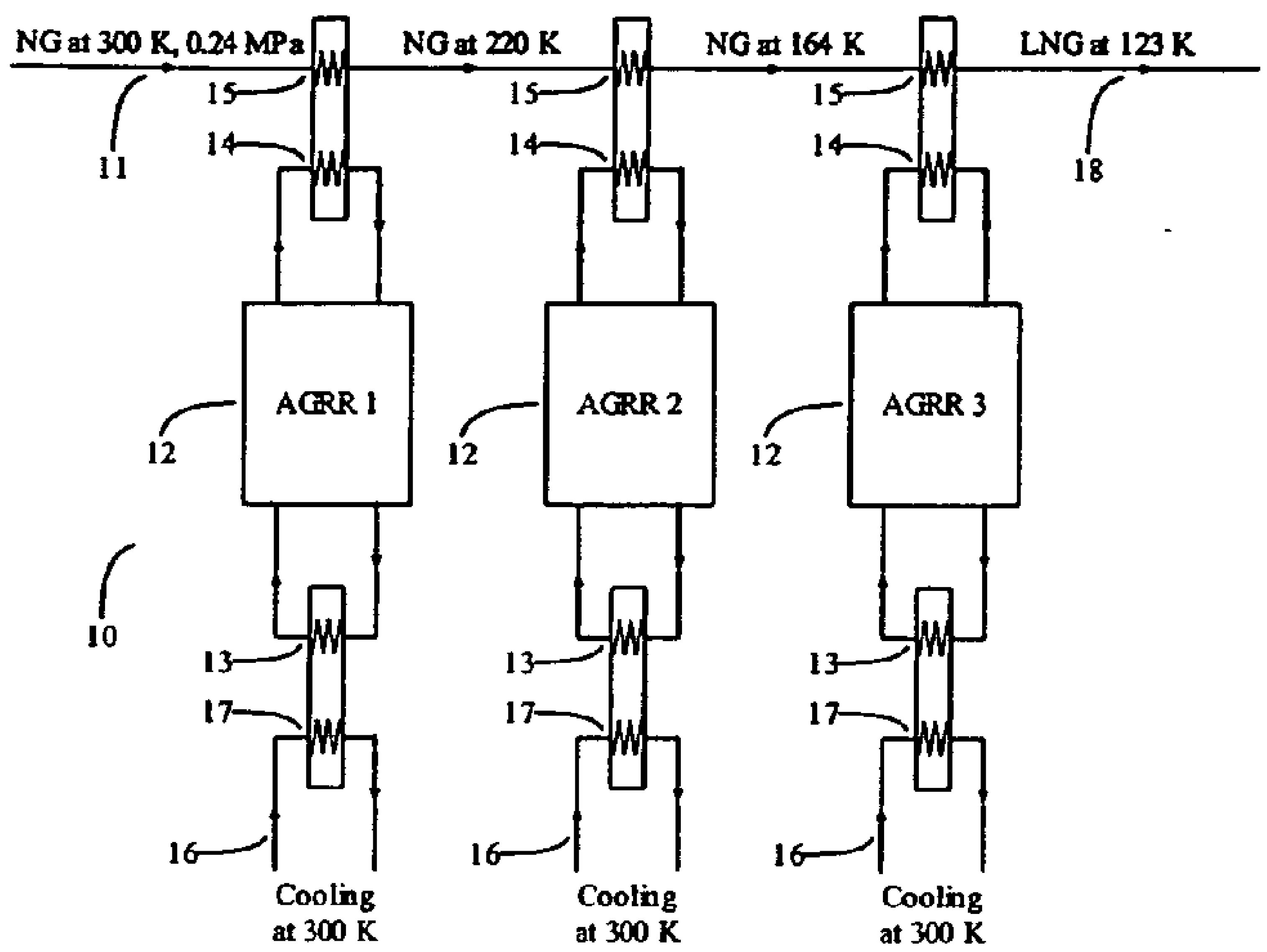

NG at 300 K, 0.24 MPa
NG at 220 K
NG at 164 K
LNG at 123 K
AGRR 1
AGRR 2
AGRR 3
Cooling at 300 K
Cooling at 300 K
Cooling at 300 K
10
11
12
13
14
15
16
17
18

FIG. 1A

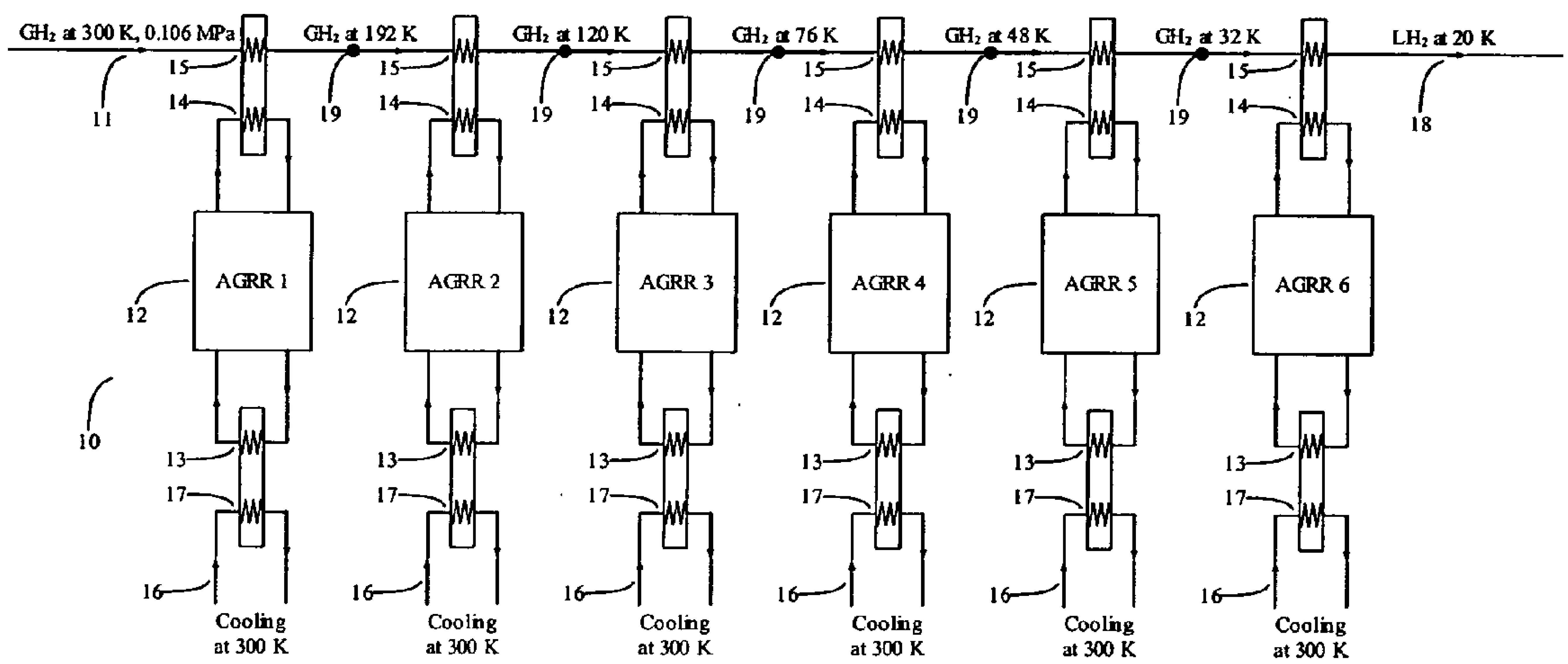

GH_2 at 300 K, 0.106 MPa
GH_2 at 192 K
GH_2 at 120 K
GH_2 at 76 K
GH_2 at 48 K
GH_2 at 32 K
LH_2 at 20 K
AGRR 1
AGRR 2
AGRR 3
AGRR 4
AGRR 5
AGRR 6
Cooling at 300 K
10
11
12
13
14
15
16
17
18
19

FIG. 1B

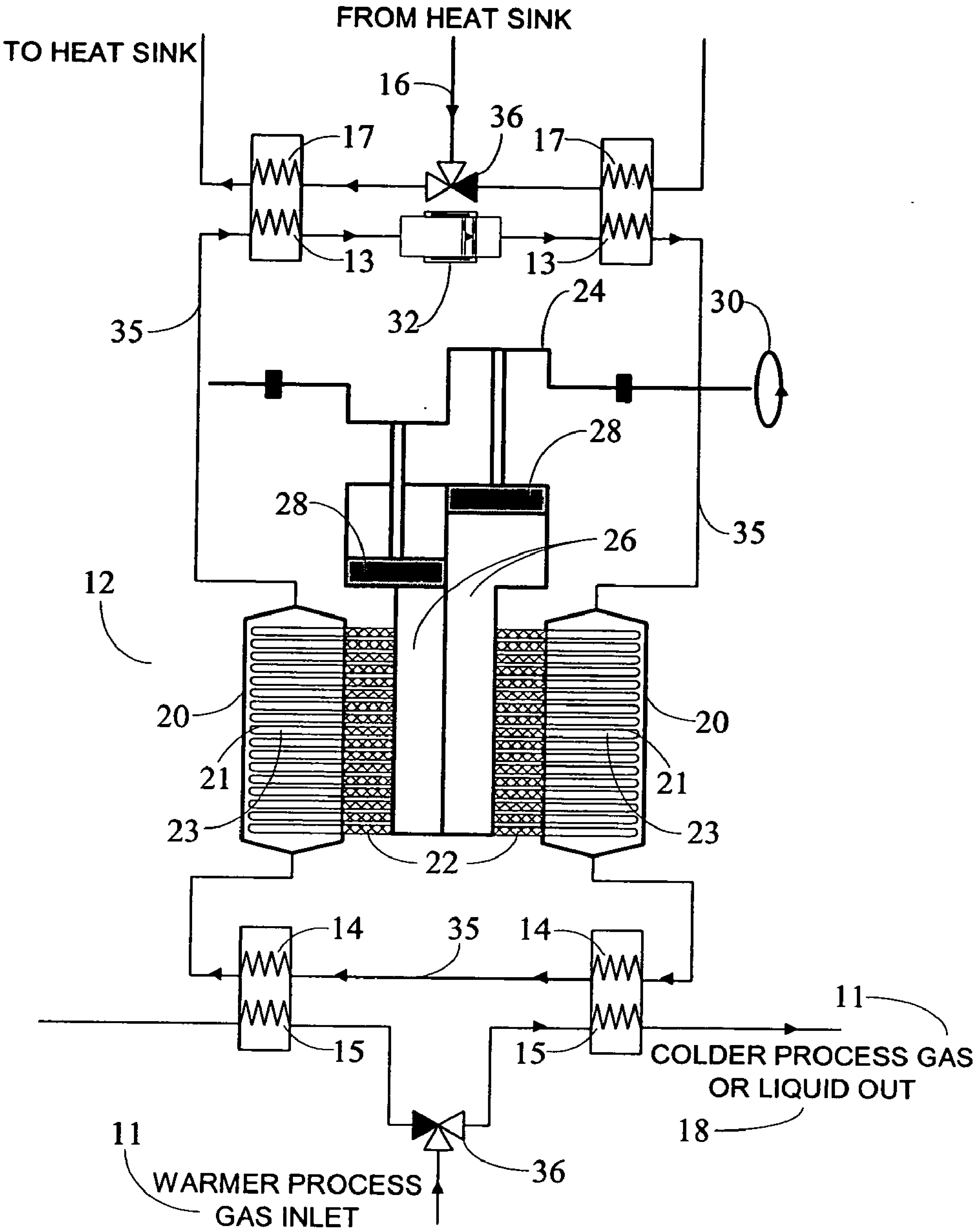

FROM HEAT SINK
TO HEAT SINK
16
36
17
17
13
13
32
24
30
35
28
28
26
35
12
20
20
21
21
23
23
22
14
35
14
11
15
15
COLDER PROCESS GAS
OR LIQUID OUT
18
11
WARMER PROCESS
GAS INLET
36

FIG. 2

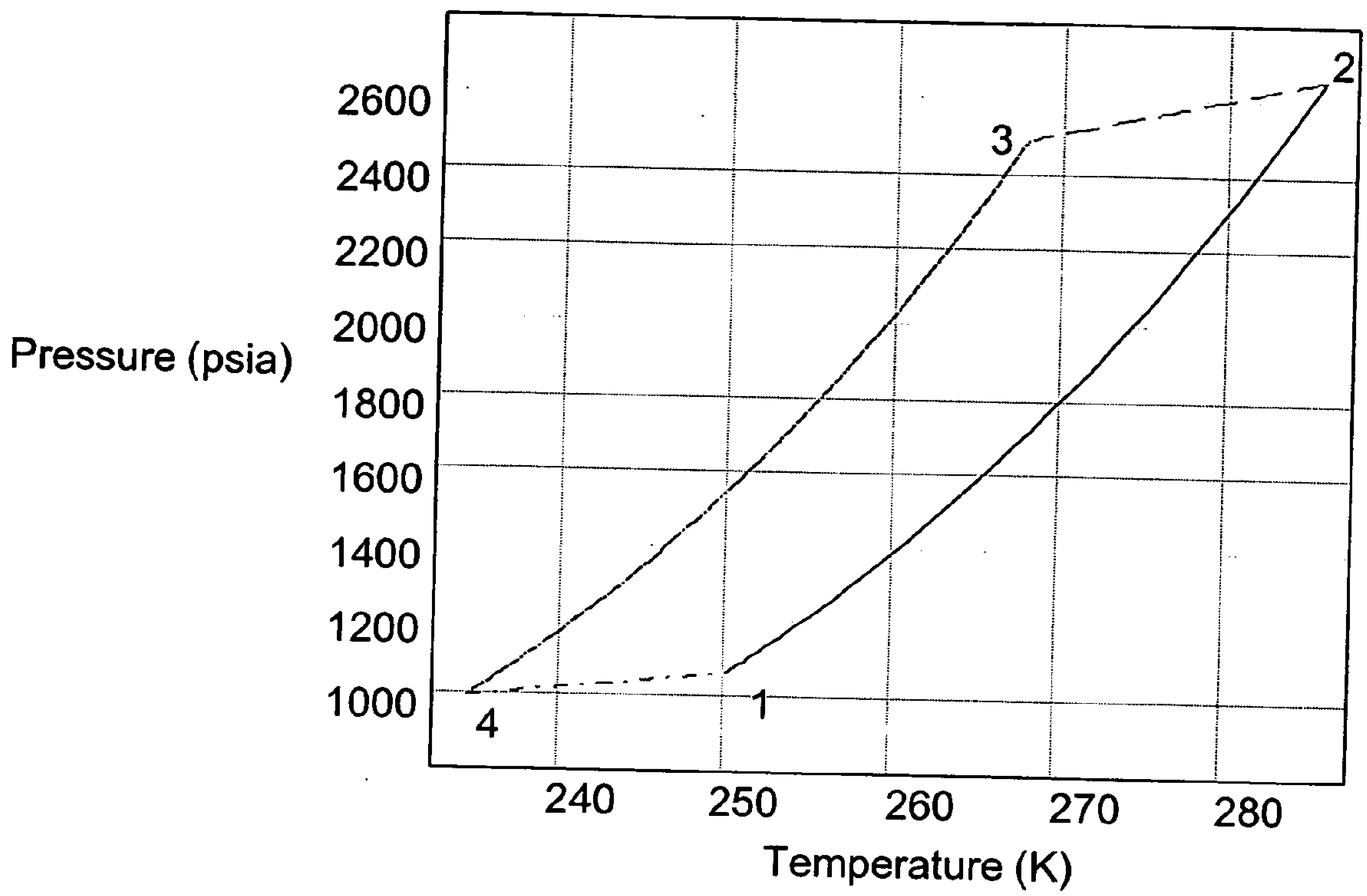

Pressure (psia)
2600
2400
2200
2000
1800
1600
1400
1200
1000
240
250
260
270
280
Temperature (K)
1
2
3
4

FIG. 3

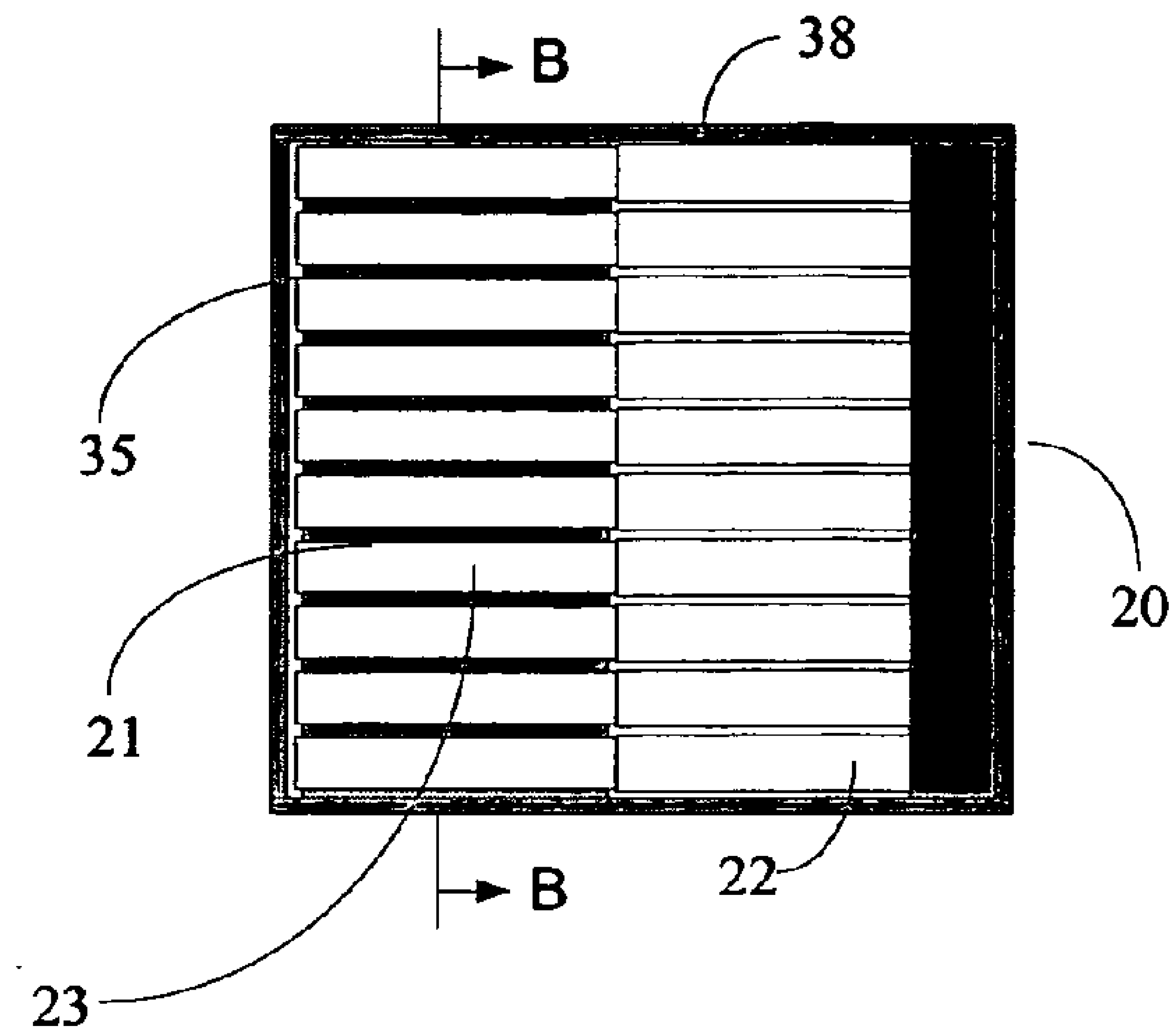


FIG. 4A

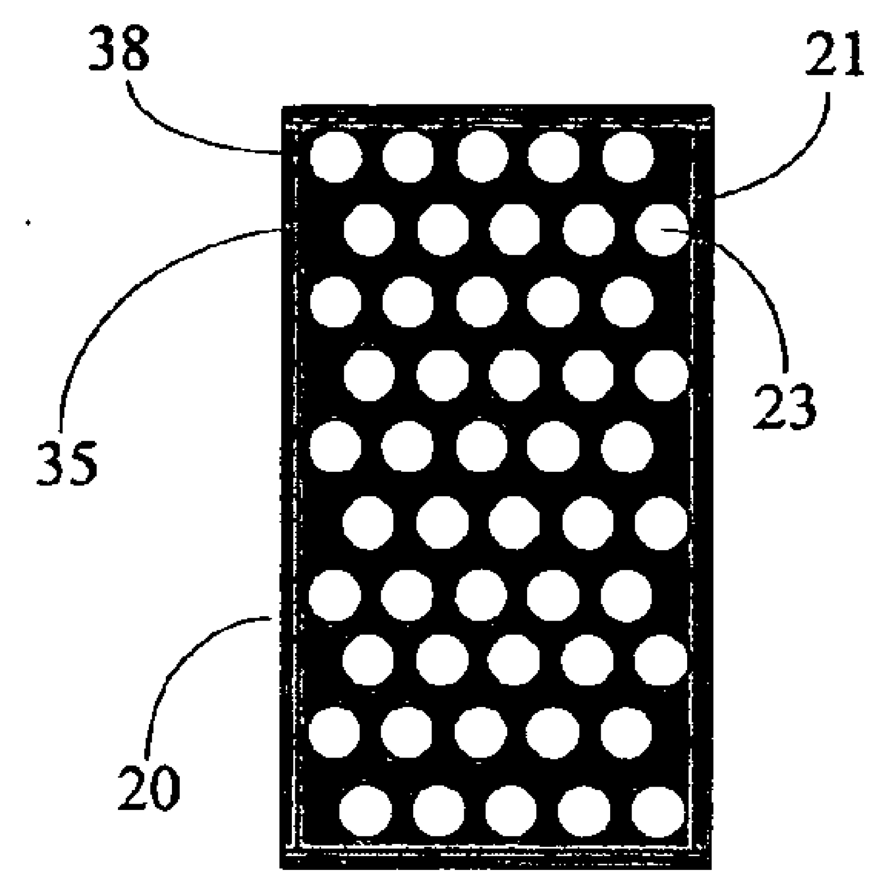


FIG. 4B

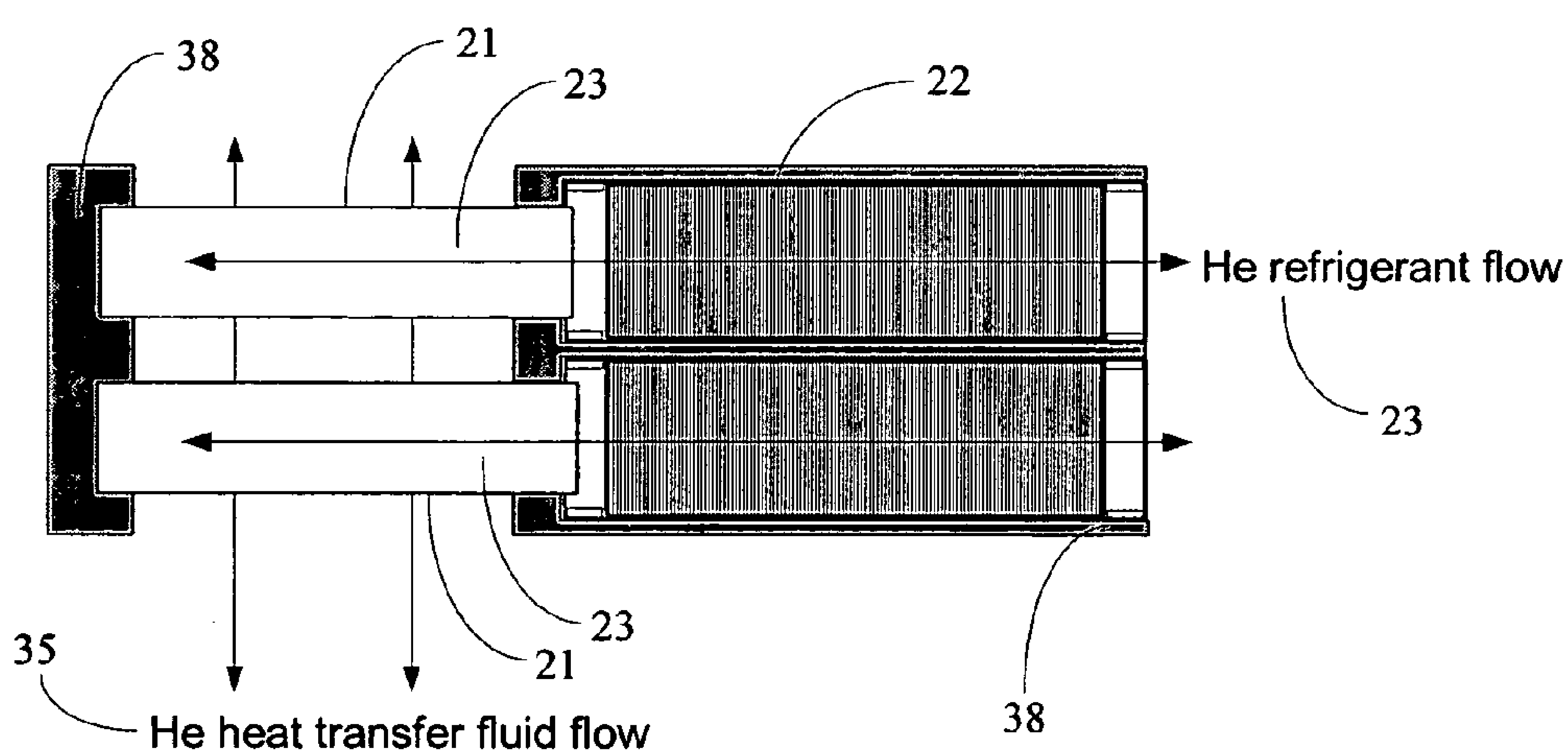


FIG. 4C

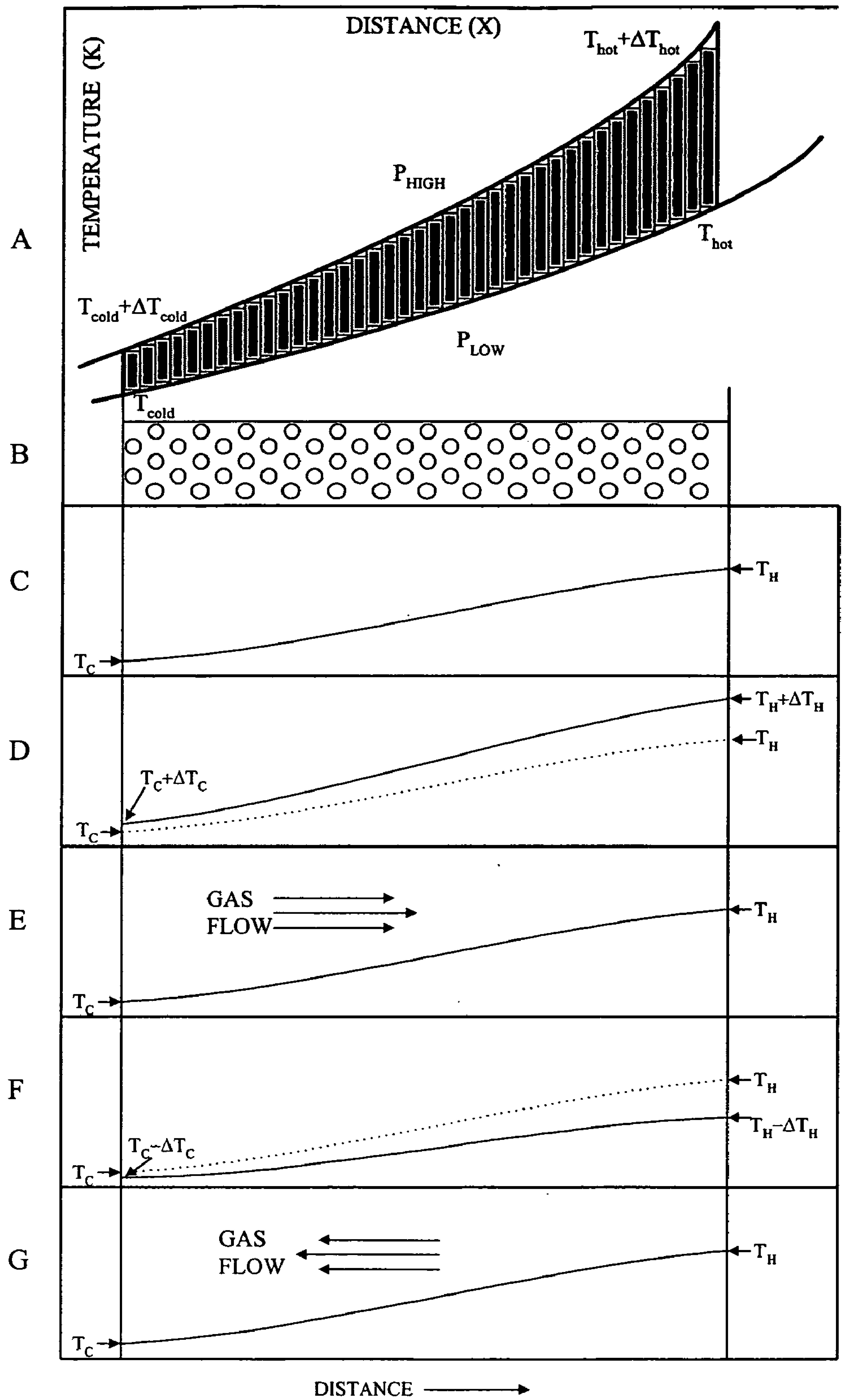

A
B
C
D
E
F
G
DISTANCE (X)
TEMPERATURE (K)
$T_{hot}+\Delta T_{hot}$
P_{HIGH}
T_{hot}
$T_{cold}+\Delta T_{cold}$
P_{LOW}
T_{cold}
T_H
T_C
$T_H+\Delta T_H$
T_H
$T_C+\Delta T_C$
T_C
GAS
FLOW
T_H
T_C
T_H
$T_H-\Delta T_H$
$T_C-\Delta T_C$
T_C
GAS
FLOW
T_H
T_C
DISTANCE

FIG. 5

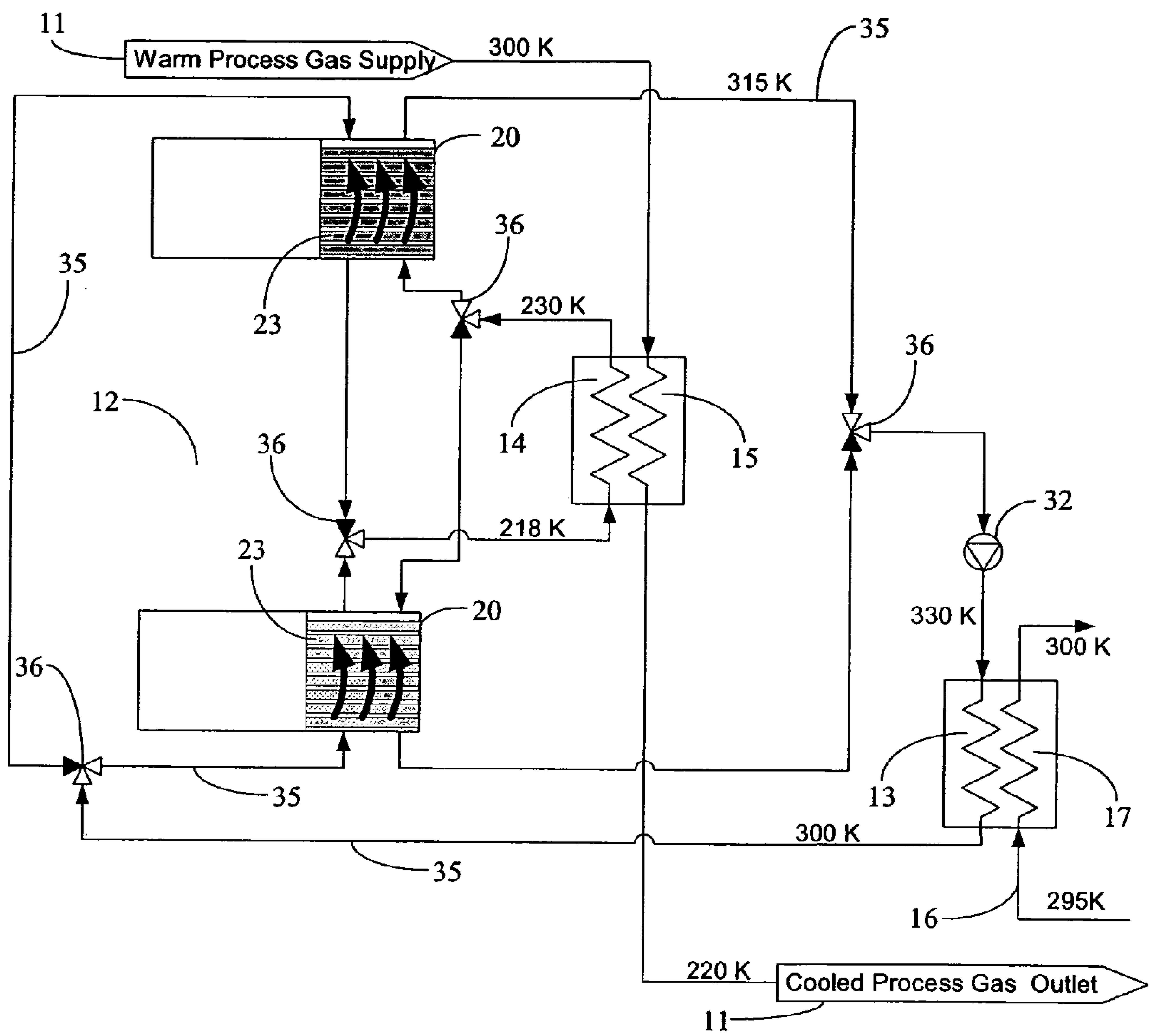

11
Warm Process Gas Supply
300 K
315 K
35
20
36
35
23
230 K
36
12
36
14
15
218 K
23
20
32
330 K
300 K
36
13
17
35
300 K
35
16
295K
220 K
Cooled Process Gas Outlet
11

FIG. 6

ACTIVE GAS REGENERATIVE LIQUEFIER SYSTEM AND METHOD

PRIORITY

[0001] The applicant claims priority from a Provisional Patent Application filed on Jul. 15, 2005, under Application No. 60/699,948.

FEDERALLY SPONSORED RESEARCH

[0002] The invention was created during a Phase I Small Business Innovation and Research award from NASA to CryoFuel Systems, Inc. under contract number NNJ04JC25C completed Jul. 15, 2004, under which the Government may have certain rights in this invention.

FIELD OF THE INVENTION

[0003] The present invention generally relates to the apparatus and method for liquefying natural gas, hydrogen, or other cryogenic fluids using one or more active gas regenerative refrigerators.

BACKGROUND OF THE INVENTION

[0004] Cryogenic liquids such as nitrogen, helium and oxygen are common forms of important industrial commodities. Similarly, liquid natural gas and liquid hydrogen provide storage, transport, and distribution for energy systems. The capital equipment and power required to make such cryogens are key factors in their use.

[0005] Liquefaction requires first cooling the gas from near room temperature to its characteristic boiling temperature. At this temperature, further cooling condenses the gas into liquid. Cryogenic liquefaction of gases can be accomplished through a variety of methods developed since about 1900.

[0006] Two general liquefaction techniques have evolved; those with a combined process and refrigerant stream, and those whose process and refrigerant streams are separate. The process stream is the gas to be liquefied and the refrigerant stream is the substance providing the cooling. In the former case, the Claude, Linde, or Brayton cycles commonly liquefy gases such as methane (the predominant component of natural gas), hydrogen, or nitrogen by processes where the process gas is simultaneously used as the refrigerant fluid. The cascade, mixed refrigerant, magnetic, or Stirling cycles are good examples of existing cycles of the latter case, where methane, hydrogen, or other cryogens are liquefied with separate process and refrigerant streams. The patent literature describes many devices covering systems of both types. A pre-cooled Claude cycle liquefier of the first type is the most common commercial scale hydrogen liquefier. A mixed refrigerant cycle liquefier of the second type is the most common commercial scale natural gas liquefier.

[0007] A refrigerator is a device that transfers or pumps heat from a specific colder temperature to a specific hotter temperature. A well-defined amount of work is required to pump a given amount of heat. An effective liquefier with separate process and refrigerant streams requires several refrigerators or stages combined appropriately to liquefy completely the cryogen. In such liquefiers each refrigerator stage requires work input to pump heat from a colder temperature to a hotter temperature.

[0008] Each refrigerator stage has a thermodynamic coefficient of performance (COP) called the Carnot ideal COP. The COP is the ratio of the heat extracted from a specific colder temperature to the work required to pump that heat to a specific hotter temperature. Real refrigerators require more work than the ideal minimum work due to a variety of well known mechanisms such as friction, pressure drop, and finite heat transfer. The ratio of ideal COP to real COP is called the efficiency of the refrigerator. Thus, in a liquefier comprised of several refrigerator stages, the efficiencies of the stages are combined to determine the real work required for liquefaction. The ratio of the ideal minimum work of liquefaction to the real work of liquefaction is defined as the Figure of Merit or FOM of the liquefier.

[0009] The ideal rate of work input for a liquefier of a certain rate of cryogen production depends on the gas to be cooled and liquefied. For example, starting with a pure process stream at one atmosphere and near room temperature, it ideally takes about 1090 kJ/kg to liquefy natural gas and about 12,100 kJ/kg to liquefy hydrogen. As stated above, the ratio of the ideal minimum liquefaction work to the actual liquefaction work is defined as the FOM. Existing conventional commercial-scale liquefier technology for natural gas and hydrogen is limited to a FOM of about 0.35[1], i.e., it requires about 3 times more work than the ideal to make liquid natural gas (LNG) or liquid hydrogen (LH_2). The technical literature[2] shows clearly that this FOM has remained the limit over the past three decades or more of technology development.

[1] Block, D. L., Dutta, S. and T-Raissi, A., "Hydrogen for Power Applications, Task 2: Storage of Hydrogen in Solid, Liquid and Gaseous Forms," *Contract Report FSEC-CR*-204-88, *Florida Solar Energy Center*, Cape Canaveral, Fla. (1988). Reference to several liquefier efficiency papers

[2] M. T. Syed, et al. Intl. Journal of Hydrogen Energy, Vol. 23, p. 565, 1998

[0010] It has been recently suggested that the FOM in conventional liquefier cycles such as the Claude cycle could approach 60%. However, this increase from about 35% to about 60% requires very high performance components in conventional liquefier designs. These choices translate into very expensive components that increase the capital cost of the liquefiers. For example, a relatively inexpensive turbine expander used in small commercial LNG liquefiers has an isentropic efficiency of about 82%. (Such an expander is used in a turbo-Brayton/Claude cycle in Prometheus Energy's stranded-gas-well to LNG commercial liquefiers making ~5,000 gallons of LNG/day.) It is possible to increase that expander efficiency to as high as 92% but only at about double the cost. The same is true with more effective heat exchangers required to increase FOMs in conventional liquefiers. More efficient gas compressors for advanced liquefiers require more stages of compression with intercoolers that increases prices sharply. These disadvantages reinforce the need for a breakthrough in liquefaction technology to increase liquefier FOM from ~0.35 to ~0.60 or higher while simultaneously reducing the capital cost of the liquefier.

[0011] Several techniques are important in designing more efficient, cost effective liquefiers. These include: means for efficient work input and work recovery; using heat exchange that matches the heat removal from the process stream to minimize heat transfer across large temperature approaches; elimination of large temperature differences between refrigerants and the process stream; reduction of parasitic heat leaks across large temperature differences; and avoidance of irreversible process changes such as excess pressure drops in pipes and valves. Each of these factors affects the FOM of the device. Active regenerative refrigerator technology has been developed over the past three decades with the objective of providing such features with substantially less irreversible entropy production and therefore higher efficiency.

[0012] An active regenerative refrigerator separates the process stream from the refrigerant, i.e. a gas or solid, and the heat transfer fluid. A passive regenerator with periodic heat transfer can be thought of as a thermal flywheel, i.e., storing thermal energy in one stage of the cycle and returning thermal energy in another stage of the cycle. Regenerative heat exchange between the refrigerant and the heat transfer fluid is periodic rather than steady state heat exchange. It is well known that high performance regenerative heat exchangers offer compact highly efficient designs for a regenerative thermodynamic cycle such as disclosed herein. The refrigerants provide the means to introduce and recover work in the cycles, usually with associated temperature changes that cause heat to flow. The refrigerants also provide the cooling for the thermodynamic cycles. In some regenerative cycles, such as a magnetic cycle and the gas cycle disclosed in the present invention, the regenerator function necessary for the cycle is simultaneously provided by the refrigerant. In that sense the refrigerant is "an active regenerator". In known prior art, active regenerators utilize excellent heat transfer geometries that have large thermal masses and huge heat transfer surfaces simultaneously with low pressure drop and low thermal conduction across inherent thermal gradients required for efficient regenerative cycle devices.

[0013] The active magnetic regenerative liquefier (AMRL) technology has been progressing since the mid seventies.[3] As described in U.S. Pat. No. 4,704,871 to Barclay, et al., active magnetic regenerative refrigerators (AMRRs) employ paramagnetic or ferromagnetic materials that, when adiabatically passed into or out of a magnetic field (typically created by a superconducting magnet), increase or decrease in temperature due to a phenomenon known as the magnetocaloric effect. The basic regenerative magnetic cycle consists of: adiabatic temperature increase upon magnetization; heat transfer to a thermal sink; regenerative heat transfer to decrease the magnetized magnetic refrigerant average temperature; adiabatic temperature decrease upon demagnetization; heat transfer from the thermal load; and regenerative heat transfer to increase the demagnetized magnetic refrigerant average temperature back to the starting temperature.

[3] See for example, U.S. Pat. No. 4,332,135 (1982) by W. A. Steyert and J. A. Barclay, NASA KSC reports, and other more recent patents and papers.

[0014] Several AMRR stages can be configured as liquefiers to make an active magnetic regenerative liquefier or AMRL for natural gas or hydrogen. The AMRL disclosed in U.S. Pat. No. 6,467,274 to Barclay, et al. achieves high FOMs by using multi-stage AMRRs in a parallel or a series configuration. Decades of component development and numerous analyzed designs establish a good basis to make efficient AMRLs spanning from about 300 K to about 110 K for LNG or to about 20 K for LH_2. However, there is a substantial cost in utilizing the AMRL. Combinations of multiple magnetic materials must be incorporated into highly effective magnetic regenerators. These regenerators must have a heat transfer fluid passing periodically through them to couple the solid refrigerant to the process stream. Superconducting magnets cooled to near liquid helium temperatures (4 K) are needed to provide magnetic fields of sufficient strength (5-6 Tesla) to cause temperature changes in the magnetic refrigerants essential to achieving high FOMs. The AMRL technology has excellent promise for large-scale hydrogen or natural gas liquefaction, but has the inherent disadvantage of being a complex system to design and operate.

[0015] Other novel active regenerative refrigerators using electrocaloric and elastocaloric effects have also been proposed and/or demonstrated, as in U.S. Pat. No. 5,465,781, issued to DeGregoria. None of these have been proposed or used for liquefaction of natural gas, hydrogen, or other cryogens because of the inability to be effectively implemented at cryogenic temperatures.

[0016] U.S. Pat. No. 6,332,323 to Reid, et al., discloses numerous potential embodiments for an active gas regenerative refrigerator (AGRR). None of the embodiments of the AGRRs in this patent were configured as a liquefier for natural gas, hydrogen, or other cryogens. In addition, none of the AGRRs in this patent allowed for distributed work input and recovery.

[0017] Thus, there is a need for a liquefier for natural gas, hydrogen, and other cryogens that operates with a high Figure of Merit and yet is relatively simple and cost-effective to produce and operate. There is also a need for more efficient active gas regenerative refrigerators for separate applications such as economical cooling high temperature superconductor devices. The present invention fulfills this and other needs.

SUMMARY OF THE INVENTION

[0018] The present invention claimed in this patent application is an active gas regenerative liquefier (AGRL) based on the combination of several active gas regenerative refrigerators (AGRRs). The AGRR stages are configured to sequentially receive and cool a process stream and deliver the process stream to the next colder AGRR stage until the process stream is liquefied.

[0019] Each AGRR stage used in the AGRL may include dual identical active regenerators also containing a refrigerant, a heat transfer fluid and a heat transfer fluid circulator that connects the dual active regenerators to a process stream heat exchanger and a heat sink exchanger, and a refrigerant compressor/expander assembly as disclosed and claimed. In one embodiment, each AGRR stage has two process stream heat exchangers and two heat rejection exchangers, using one set of exchangers in one half of the cycle and the other set of exchangers in the other half. In an alternate embodiment, each AGRR stage incorporates multiple three-way valves to efficiently control the flow of the heat transfer fluid through a single process stream heat exchanger and a single heat sink exchanger during the cycle. Preferably, each AGRR stage will also have instrumentation for operational control and a cold box for effective thermal insulation of the cryogenic components from the environment.

[0020] An efficient AGRL for natural gas is disclosed using at least three AGRR stages. An AGRL for hydrogen is also disclosed using at least six AGRR stages. Use of several stages of refrigeration approximates continuous heat removal from the process stream at the highest possible temperature, one of the elements of a highly efficient liquefier design. Using such configured AGRR stages, the ideal FOM can reach about 0.92 for the three-stage natural gas liquefier of this invention and about 0.81 for the six-stage hydrogen liquefier of this invention. Fewer or more AGRR stages can also be used in other embodiments of an AGRL.

[0021] Work recovery is another feature of a highly efficient liquefier. Gas refrigerant compression is very work intensive and is normally done near ambient temperature. Gas refrigerant expansion is normally done in a device called an expander at cryogenic temperatures. Besides providing cooling of the refrigerant, it can be a means to recover a portion of work of compression[4]. The imbalance of work input in the compressor and recovery in an expander in a conventional gas liquefier is a fundamental limitation to its efficiency. The present invention uniquely provides distributed compression in all AGRR stages at temperatures from near room temperature to near cryogen liquefaction temperatures and simultaneously recovers work from distributed expansion at temperatures from near room temperature to cryogenic temperatures. This feature makes the AGRL design inherently more efficient than conventional liquefiers.

[4] Isentropic expansion of a gaseous refrigerant from high pressure to low pressure cools the refrigerant and provides work output. This work can be used to offset the work of compression of the refrigerant.

[0022] In a parallel-type AGRL each AGRR stage uses a refrigerant to pump a thermal load from a cold temperature unique to each AGRR stage to a common heat rejection temperature, e.g., near room temperature. The final, or coldest, AGRR stage removes primarily latent heat from a process stream to liquefy it and expels rejected heat at near room temperature. The previous successively warmer AGRR stages in a parallel-type AGRL remove primarily sensible heat from the process stream to cool it and expel rejected heat at near room temperature. The efficiency of each AGRR stage depends on its inherent inefficiencies from real heat transfer, fluid flow, refrigerant compression/expansion, and other processes necessary to pump heat from a colder to a warmer temperature. Since each AGRR stage spans a different temperature range in the disclosed AGRL, each stage can be optimally designed to achieve high efficiency by choices that minimize irreversible entropy creation in all aspects of the overall AGRL design. These include minimum temperature approaches in all heat exchangers, small pressure drops, and efficient work input and recovery. By using a parallel-type AGRL configuration with highly efficient AGRR stages as disclosed a FOM of about 0.60 for liquefaction of hydrogen is achievable at relatively low cost. The combination of the several real AGRR stage efficiencies can provide natural gas or hydrogen liquefiers with FOMs of between 0.52 and 0.69. This performance is a quantum increase over that of the best conventional liquefiers with FOMs of about 0.35. This AGRL invention provides a breakthrough in efficient and cost-effective hydrogen and natural gas liquefaction.

[0023] In another embodiment of the invention, the AGRL includes several stages of refrigeration, with each stage including an array of discrete micro compressor-expander units (MCEU)[5] configured as a high performance active regenerator having excellent heat transfer, low pressure drop, and low longitudinal conduction with respect to the heat transfer fluid and regenerator materials. The compressor-expander units are configured such that the compression of the refrigerant within a unit is coupled to the simultaneous expansion of the refrigerant within the other end of the unit, thereby allowing distributed work input and recovery from near ambient temperature to cryogenic temperatures necessary for liquefaction of natural gas or hydrogen. In this embodiment, the net work input is reduced substantially thus providing very efficient regenerative refrigeration, no matter what the temperature span of the liquefier. This input of "distributed net work" is unique among gas liquefiers.

[5] The MCEU or micro compressor expander unit may consist of a small diameter tube, such as mm dimensions, with working refrigerant gas that is separately compressed at one end of the MCEU, and simultaneously separately expanded on the other end of the MCEU such that the work for compression of the refrigerant is partially compensated by work produced by the expansion.

[0024] Another feature of the AGRL is the use of multistage refrigeration to a sequence of separate process stream heat exchangers containing the flowing process stream (natural gas or hydrogen gas) that approximates continuous cooling. In the case of hydrogen, associated ortho-to-para (o-p) exothermic converters at each stage enables removal of the o-p heat as the hydrogen is cooled and liquefied. This multistage AGRR design feature markedly increases the thermal efficiency of an AGRL compared to conventional hydrogen liquefiers, e.g. ones based on a Claude cycle.

[0025] In another embodiment of the disclosed invention, temperature approaches in between refrigerants in the individual tubes or MCEUs and heat exchange fluid in the active regenerators are kept small, thus avoiding the inherent inefficiency of conventional cycles when heat transfers across larger temperature spans. Thus, the temperature differences between the array of distributed MCEUs, between the heat transfer fluid that couples the refrigerants in the MCEUs to the process stream heat exchangers and heat sink exchangers of each AGRR, and between the heat exchange fluid and the process streams can be selected in a manner that optimizes efficient heat transfer and thereby increases thermodynamic efficiency.

[0026] The AGRL as disclosed and claimed is relatively simple in design as compared to other liquefiers, thus requiring fewer components, less expense, and simpler controls for automatic operation. These features are important attributes of commercial liquefiers.

[0027] A method of liquefying a process stream of gas is also disclosed. The steps include: cooling a heat transfer fluid by passing it through a first active regenerator in which a refrigerant has been expanded, circulating the heat transfer fluid through a process stream heat exchanger to pick up a thermal load from the process stream, passing the heat transfer fluid through a second active regenerator in which the refrigerant has been compressed, thereby heating the heat transfer fluid, and circulating the heat transfer fluid through a heat rejection exchanger to expel excess heat to a heat sink. These steps may be repeated several times in each of several stages of refrigeration, each stage successively cooling the process stream until liquefaction occurs.

[0028] The above summary of the present invention is not intended to represent each embodiment, or every aspect, of the present invention. The present invention also includes any additional features and benefits which are apparent from the detailed description and figures set forth below.

BRIEF DESCRIPTION OF THE FIGURES

[0029] FIG. 1A depicts an AGRL of three AGRR stages for liquefaction of natural gas.

[0030] FIG. 1B depicts an AGRL of six AGRR stages for liquefaction of hydrogen.

[0031] FIG. 2 is a schematic of a distributed-tube AGRR with a common compressor/expander assembly. This AGRR is configured as a single stage of a multistage AGRL.

[0032] FIG. 3 is a pressure versus temperature diagram of the gas cycle of the combined refrigerant and a thin-walled tube in a selected micro compressor-expander unit of an

AGRR operating in an active regenerative cycle near room temperature. This diagram assumes instantaneous heat transfer between the refrigerant and the thin-walled tube in the MCEU. The pressures used in this diagram are illustrative rather than prescriptive.

[0033] FIG. **4**A is a side view of a simplified active regenerator configured as an array of thin-walled tubes with helium gas as the refrigerant. This active regenerator has a common supply duct from the compressor/expander and thermally isolates each layer of thin-walled tubes from room temperature using a passive micro-regenerator of stainless steel particles.

[0034] FIG. **4**B is side view of the active regenerator taken along the line **4**B-**4**B in FIG. **4**A that illustrates the array of thin-walled tubes comprising the regenerator geometry.

[0035] FIG. **4**C is an expanded view of two thin-walled tubes of the active regenerator, together with the passive micro-regenerators that thermally isolate the refrigerant in each of the tubes from the refrigerant supplied by a common room temperature compressor/expander assembly.

[0036] FIGS. **5**A to **5**G are diagrams of the typical temperature profiles of the active regenerator refrigerant/thin tubes of an individual AGRR stage of the AGRL at various points of the active gas regenerative cycle. The flow indicated in the diagram is that of the heat transfer fluid through the regenerator in its periodic flow that sequentially connects thermally to the process stream thermal load and to the heat sink heat exchanger.

[0037] FIG. **6** is a schematic showing the flow of the heat transfer fluid in the refrigeration cycle in the first stage of an AGRL system with dual active regenerators. The three-way valve ports are shown as black if closed and white if open. The process stream is cooled in this AGRR stage from about 300 K to about 220 K The heat rejection exchanger shows the heat transfer fluid being cooled from 330 K to 300 K. This embodiment could be the first stage of a natural gas or a hydrogen AGRL.

[0038] While the invention is susceptible to various modifications and alternative forms, specific embodiments are shown by way of example in the drawings and are described in detail herein. It should be understood, however, that the invention is not intended to be limited to the particular forms disclosed. Rather, the invention is to cover all modifications, equivalents, and alternatives falling within the spirit and scope of the invention as described.

DETAILED DESCRIPTION OF THE ILLUSTRATED EMBODIMENTS

[0039] Referring to FIG. **1**A, a preferred embodiment of the active gas regenerative liquefier or AGRL (**10**) is schematically shown. A process stream (**11**) (e.g. natural gas) enters the AGRL (**10**) from the left in the figure. The natural gas process stream (**11**) in this embodiment is initially at a temperature of 300 K and a pressure of about 20 psig (0.24 MPa). The AGRL (**10**) comprises three AGRR stages (**12**). In parallel configuration, each AGRR stage (**12**) has a heat rejection exchanger (**13**), or heat rejection means, at a common temperature (300 K in this embodiment) and a cold heat exchanger (**14**) at a temperature designed to maximize the FOM of the liquefier system. Each AGRR stage (**12**) receives the process stream (**11**) and cools it to about the temperature of the cold heat exchanger (**14**) of that stage. In this embodiment, the first AGRR has a cold heat exchanger at about 220 K, the second AGRR has a cold heat exchanger at about 164 K, and the third AGRR has a cold heat exchanger at about 123 K. The cold heat exchanger (**14**) of each stage removes heat from the process stream (**11**) in a process stream heat exchanger (**15**). The heat rejection exchanger (**13**) of each stage expels heat to a cooling fluid (**16**) (e.g., water) in a heat sink exchanger (**17**). In the final stage of refrigeration in AGRR **3** in FIG. **1**A, the latent heat of vaporization is removed, liquefying the process stream (**11**). Thus, after passing through the third AGRR, the process stream output (**18**) is liquid natural gas at 123 K in this embodiment.

[0040] In one embodiment, the cooling fluid will pass through each heat sink exchanger to a separate water-to-air heat exchanger, where the water is cooled and returned to the liquefier system to cycle through the heat sink exchangers again. In another embodiment, a common heat sink exchanger may be used that is capable of handling the heat rejected from each AGRR stage and connected to a closed loop water chiller. This water chiller will have a circulation pump for the water as well as a fan to drive air convection through the water radiator. The pump and fan powers for the chiller will be small and supplied by small motors. The process stream heat exchangers (**15**) and the cryogenic AGRR cold heat exchangers (**14**) may be contained within a cold box or otherwise thermally isolated from the surroundings to reduce parasitic heat leaks into the cryogenic liquid product (**18**).

[0041] FIG. **1**B schematically shows a preferred embodiment of the AGRL (**10**) for production of LH_2. The hydrogen process stream (**11**) in this embodiment is initially at a temperature of 300 K and a pressure of about 1 atmosphere (0.1 MPa). The process stream (**11**) enters from the left in the figure and passes through six successive AGRR stages (**12**) with cold heat exchangers (**14**) at temperatures of about 192 K, 120 K, 76 K, 48 K, 32 K and 20 K. Between each stage, a continuous ortho-to-para catalytic converter (**19**) converts the ortho form of hydrogen in the process stream (**11**) to an equilibrium concentration of the para form of hydrogen at that particular temperature, thereby increasing the efficiency of the AGRL (**10**). The remaining elements are as described for FIG. **1**A, resulting in a process stream output (**18**) of liquid hydrogen at 20 K in this embodiment.

[0042] The designed cooling capacity of the AGRR stages scales with the rate of production of LNG or LH_2. The AGRL design can be scaled from several hundred or thousand gallons/day upwards to much larger liquefaction capacities. This type of AGRL for natural gas or for hydrogen has been designed to have a FOM within the range of about 0.52 to 0.69.

[0043] Referring to FIG. **2**, a unique and novel AGRR stage (**12**) is depicted. This AGRR design can be used for the several AGRR stages required to make the AGRLs shown in FIGS. **1**A and **1**B. The AGRR stage (**12**) includes dual regenerators (**20**) comprised of rectangular arrays of many small tubes (**21**) and a refrigerant compressor/expander assembly (**24**) operating at room temperature, comprised of a manifold (**26**), pistons (**28**), and a drive mechanism (**30**). The tubes (**21**) are filled with a refrigerant (**23**) (e.g. helium gas) and a heat transfer gas displacer or circulator (**32**) is also filled with a heat transfer fluid (**35**) (e.g., nitrogen gas for LNG and helium gas for LH_2). The tubes (**21**) in each active regenerator (**20**) are connected to the refrigerant manifold (**26**) by an array of passive micro-regenerators (**22**) located at the entrance of each of the tubes (**21**) to the common refrigerant manifold (**26**). Pistons (**28**) are located within the manifolds (**26**) to alternately compress and expand the refrigerant (**23**) within the regenerators (**20**). The pistons (**28**) are coupled such that one piston will compress the working refrigerant (**23**) in a first (left most in FIG. **2**) portion of the manifold (**26**) while another piston simultaneously expands the refrigerant (**23**) in a second (right most in FIG. **2**) portion of the manifold (**26**). The pistons (**28**) are driven by the drive mechanism (**30**).

[0044] After temperature gradients are established in arrays of tubes (**21**) during a short startup sequence, the AGRR stage (**12**) operates as follows: A piston (**28**) expands the refrigerant (**23**) in all the small tubes (**21**) of one of the dual regenerators (the right regenerator in FIG. **2**) (**20**) and compresses the refrigerant (**23**) in all the small tubes (**21**) of the second of the dual regenerators (the left regenerator in FIG. **2**) (**20**). The pressure decrease of the refrigerant (**23**) in the tubes (**21**) of the right regenerator (**20**) causes a polytropic expansion with a corresponding temperature decrease of the refrigerant (**23**) in each tube (**21**) such that the temperature in the active regenerator spans from near room temperature at the top to near a cryogenic thermal load temperature at the bottom. A displacer or circulator (**32**) drives the heat transfer fluid (**35**) from a heat rejection exchanger (**13**) cooled by a cooling fluid (**16**) downward through the array of tubes (**21**) in the right regenerator (**20**), further cooling the heat transfer fluid (**35**) to near a cryogenic thermal load temperature. The heat transfer fluid (**35**) then flows through a cold heat exchanger (**14**) that is thermally coupled to a process stream heat exchanger (**15**) (the right most process stream heat exchanger in FIG. **2**) where it picks up a thermal load from the process stream (**11**) before passing through a second cold heat exchanger (**15**) (the left most cold heat exchanger in FIG. **2**) and into the array of tubes (**21**) in the left active regenerator (**20**). There is no process stream flow through the left most process stream heat exchanger (**15**) in FIG. **2** so the heat transfer fluid temperature does not change in the left most cold heat exchanger (**14**). The refrigerant (**23**) in the small tubes (**21**) of the left active regenerator (**20**) has been heated by a corresponding polytropic compression caused by the motion of the second piston (**28**) of the compressor/expander assembly (**24**). The heat transfer fluid (**35**) flowing from the bottom to the top of the left regenerator (**20**) is warmed as it picks up heat from the left regenerator (**20**) and is directed to a heat rejection exchanger (**13**), where the heat transfer fluid (**35**) is cooled when heat is transferred to a cooling fluid (**16**) via a heat sink exchanger (**17**). Three way valves (**36**) direct the flow of the process stream gas (**11**) and the heat sink cooling fluid (**16**) to the appropriate process stream heat exchanger (**15**) and heat sink exchanger (**17**) in a counterflow direction. The displacer or circulator (**32**) then reverses the flow of the heat transfer fluid (**35**) and the drive mechanism (**30**) of the compressor/expander assembly (**24**) moves the pistons (**28**) simultaneously to compress the refrigerant (**23**) in the small tubes (**21**) of the right active regenerator (**20**) and expand the refrigerant (**23**) in the small tubes (**21**) of the left active regenerator (**20**). The heat transfer fluid (**35**) circulates in a reverse direction through its flow path, now being cooled in the left regenerator (**20**), picking up a thermal load from the left process stream heat exchanger (**15**), receiving heat from the right regenerator (**20**), and expelling the heat out to the heat rejection exchanger (**13**) to the cooling fluid (**16**).

[0045] In the embodiment depicted in FIG. **2**, there are two process stream heat exchangers (**15**) and two heat sink exchangers (**17**). Three-way valves (**36**) control the flow direction of the cooling fluid (**16**) from the heat sink and the flow direction of the process stream gas (**11**), allowing the periodic flow of the dual active regenerators (**20**) to be coupled in counterflow with the process stream heat exchangers (**15**) and the heat sink exchangers (**17**), one set of exchangers being used in the first half of the cycle, and the second set of exchangers being used in the second half of the cycle. An alternate embodiment utilizes two additional three-way valves to control the flow of the heat transfer fluid without the need for two sets of exchangers, as will be described in detail below.

[0046] During the cycle, the average temperature of the refrigerant (**23**) in the array of tubes (**21**) is increased in one regenerator (**20**) and decreased in the other regenerator (**20**) as the heat transfer fluid (**35**) flows. The direction of flow of the heat transfer fluid through the active regenerator (**20**) is reversed by the displacer or circulator (**32**) when the temperature of the bottommost layer of tubes (**21**) increases by about half the temperature decrease of the small tubes caused by the polytropic expansion of the refrigerant in bottommost layer of tubes in the active regenerator (**20**). This flow reversal of the heat transfer fluid is synchronized with the compression/expansion of the refrigerant in the dual regenerators.

[0047] The work required to pump the heat from the thermal load to the heat sink is distributed over all the tubes (**21**) comprising the dual active regenerators (**20**) of each AGRR stage. By coupling the pistons (**28**) together via a direct linkage, the net work of refrigerant compression that must be externally supplied by the drive mechanism (**30**) is the net work required for the thermodynamic refrigeration provided by the AGRR stage. The offset of most of the work of compression of the refrigerant by work recovery from simultaneous expansion of the refrigerant and the distributed work input as a function of temperature are two of the fundamental reasons for a high efficiency in an individual AGRR stage (**12**) of an AGRL. The cooling power of the AGRR stage (**12**) is proportional to the heat transfer fluid flow rate and the effective temperature changes of the refrigerant caused by compression/expansion. As described, the heat transfer fluid (**35**) is cooled or heated by the effective temperature changes fluid, helium gas at a modest pressure, (e.g., 1-2 MPa), in the process stream heat exchanger for each AGRR stage. The thermal cooling load at each stage is related to the He and H_2 mass flow rates via the equation

$$\dot{m}_{He}c_{He}\Delta T_{cold}=\dot{Q}_{load}=\dot{m}_{H2}(h_f-h_i)$$

where c_{He} is the heat transfer fluid heat capacity, h_f and h_i are enthalpies of the H_2 at the entrance and exit of the process stream heat exchanger for the respective AGRR stage, ΔT_{cold} is the effective temperature change of the He heat transfer fluid as it passes in counterflow through the process stream heat exchanger with an average H_2 exit temperature of about T_{cold} for that particular process stream heat exchanger. After passing through the process stream heat exchanger, the warmer He heat transfer fluid then flows back through the other dual active regenerator of the AGRR stage where the refrigerant within all tubes in this array is compressed with corresponding effective adiabatic temperature increases above the mean temperature at a particular longitudinal position along the active regenerator. At the beginning of this "hot blow" period, the temperature spanned by the dual active regenerator with the compressed refrigerant in each tube is ~T_{cold} to ~$T_{hot}+\Delta T_{hot}$. The heat transfer fluid picks up heat from each of the tubes in the active regenerator as it flows and eventually leaves the hot end of the regenerator at a temperature higher than the mean heat sink temperature.

[0048] The periodic motion of the heat transfer fluid is synchronous with the operation of the refrigerant-filled tubes and only shifted in phase by the ratio of the time for the compression/expansion step to the time for the blow periods (this ratio is usually small, i.e., 0.05 to 0.1). To accomplish this effectively with a single room temperature heat transfer fluid displacer or circulator, the preferred embodiment uses a valve arrangement to create a periodic of the refrigerant (**23**) in the array of tubes (**21**) of the active regenerators (**20**) caused by the compressor/expander assembly (**24**).

[0049] The frequency of periodic flow reversal in the displacer or circulator (**32**) and the operation of the three-way valves (**36**) is properly phased with the temperature changes in the array of tubes (**21**) and can typically operate at reasonable frequencies near 1 Hz. The dual regenerator configuration in this AGRR stage (**12**) allows the heat transfer fluid loop to be hermetic and reversible according to the motion of the displacer or circulator (**32**) and pistons (**28**). The dual regenerators (**20**) are identical and operate 180° out of phase with each other. In other words, the compression of the refrigerant (**23**) by a piston (**28**) connected via manifold (**26**) to the array of tubes (**21**) in one of the dual regenerators (**20**) is synchronous with the expansion of the refrigerant (**23**) by a piston (**28**) connected via a manifold (**26**) to the other dual regenerator (**20**). Similarly, the heat transfer fluid flow or cold blow (from top to bottom) in one active regenerator (**20**) is synchronous with the heat transfer fluid flow or hot blow (from bottom to top) in the other regenerator (**20**). The heat transfer fluid (**35**) reciprocatively flows through each of the dual regenerators (**20**) while flowing semi-continuously in counterflow through the cold heat exchanger (**14**) coupled to the process stream heat exchanger (**15**) and the heat rejection exchanger (**13**) coupled to the heat sink exchanger (**17**).

[0050] In one embodiment, each passive micro-regenerator (**22**) for each individual tube in a given layer of tubes in the two-dimensional array of tubes (**21**) comprising the dual active regenerators (**20**) has a thermal mass of ~30-50 times the thermal mass of the refrigerant (helium gas) that flows in and out of each tube (**21**) during compression or expansion. A typical passive micro-regenerator material is stainless steel spheres with a diameter of ~200-300 microns (0.2-0.3 mm). The temperature span across each layer of passive micro-regenerators (**22**) feeding each layer of tubes (**21**) in the active regenerator will be from the average cold temperature of the refrigerant plus tube combination in that layer of tubes (**21**) and the near room temperature refrigerant in the manifold (**26**) connecting the compressor/expander to the active regenerator. The diameter of each cylindrical passive micro-regenerator (**22**) is the same as the diameter of the small tube it is connected to in the dual active regenerators. The length of these passive micro-regenerators (**22**) is a design variable and was chosen as a few centimeters in one tested embodiment. The pressure drop for the helium gas refrigerant as it flows through the passive micro-regenerators (**22**) in and out of the individual tubes (**21**) was designed to be very low at an operational cycle time of ~1 Hz. The pressure changes of the helium refrigerant (**23**) within the tubes (**21**) can be from ~215 psia to ~430 psia in a typical operation. The mean pressure of the He heat transfer fluid that flows through the dual active regenerators (**20**) and the heat transfer fluid in the reversible displacer or circulator (**32**) is an operating variable and was about 200 psia in a test embodiment. It may be higher or lower if desired. The pressure drop of the heat transfer fluid (helium gas in the preferred embodiment for LH_2) through optimally designed dual regenerators (**20**) is typically very small, i.e., 10's to 100's of Pa.

[0051] The pressure versus temperature diagram of FIG. **3** illustrates the thermodynamic cycle of a typical tube filled with refrigerant gas to a mean pressure of about 1800 psia. This figure shows how the temperature of the refrigerant changes throughout the active gas regenerative cycle. The pressure increase from the compression of the refrigerant in the tubes in the same layer within an array of tubes comprising an active regenerator increases the pressure from about 1800 psia to about 2600 psia. The corresponding temperature of the refrigerant gas plus the thin tube shell increases from about 270 K to about 285 K, i.e. to point **2** on P-T coordinates of FIG. **3**. This point corresponds to the temperature $T_{hot}+\Delta T_{hot}$. The heat transfer fluid is circulated by and around the tubes in this particular layer of tubes to partially remove the heat of compression and cool the refrigerant and tube to point **3** (at temperature T_{hot}) on the P-T coordinates in FIG. **3**. The expansion of the refrigerant gas by the compressor/expander assembly from about 2500 psia to about 1000 psia cools the refrigerant to about 235 K as shown by point **4** on the coordinates of FIG. **3**, at temperature $T_{cold}-\Delta T_{cold}$. The heat transfer fluid again flows by and around the tubes in this layer of the active regenerator in the opposite direction while heat is added to the cold refrigerant and tube until the temperature reaches about 250 K as indicated by point **1** (at temperature T_{cold}) on the coordinates in FIG. **3**. The compression of the refrigerant by the compressor/expander assembly is repeated to increase the pressure from about 1000 psia to about 2600 psia with a corresponding temperature increase to about 285 K as shown in point **2**. The cycle is now complete and repeats at the frequency of operation of about 1 Hz.

[0052] FIG. **4**A shows a cross side view of a simplified active regenerator (**20**). The layers of tubes (**21**) are shown attached to the passive micro-regenerators (**22**) of each tube in each layer of the active regenerator (**20**). These passive micro-regenerators (**22**) allow the common compressed and expanded refrigerant to always be near room temperature in the manifolds (**26**) by highly effective heat transfer to or from the refrigerant as it flows in and out of individual thin-walled tubes (**21**) of the active regenerators (**20**). The thermal mass of the passive micro-regenerators is chosen to be much larger than the refrigerant as required for high performance designs. From the top of the active regenerator, each layer of passive micro-regenerators (**22**) will have a slightly lower temperature on the tube end than the layer above it, thereby creating a temperature span across the active regenerator in the AGRR. For example, in the first stage AGRR of a three-stage AGRL for LNG, the upper layer of micro-regenerators (**22**) will have its average tube-end temperature close to 290 K and the bottommost layer of passive micro-regenerators (**22**) will have an average tube-end temperature close to 215 K. The passive micro-regenerators (**22**) at the coldest layer of this AGRR stage will have a temperature gradient along their longitudinal axis (from the tube-end towards the manifold (**26**)) from about 215 K to about 300 K.

[0053] FIG. **4**B is a side view of the active regenerator to illustrate the staggered array of thin-walled tubes (**21**) filled with refrigerant (**23**). In the preferred embodiment, these tubes are typically 0.25" in diameter with a wall thickness of ~0.001". FIG. **4**C illustrates the two helium flows (refrigerant (**23**) and heat transfer fluid (**35**)) required for the operation of the AGRR stage. One transverse flow in the active regenerator is the refrigerant (**23**) that is compressed and expanded by the combined compressor/expander assembly. The other vertical flow is the heat transfer fluid (**35**) through the array of tubes to periodically transfer heat to and from the refrigerant-filled tubes (**21**) and coupling these heat flows to the heat rejection exchanger and cold exchanger of each AGRR stage within the AGRL. The active regenerator (**20**) is hermetically sealed by a suitably chosen assembly container (**38**).

[0054] FIG. **5** illustrates the combined operation of one of the dual active regenerators of an AGRR stage. The temperatures span from about 300 K on the hot end to about 20 K on the coldest end of the sixth AGRR stage of an AGRL for hydrogen or to about 123 K on the coldest end of the third AGRR stage of an ARGL for natural gas.

[0055] FIG. **5**A shows the temperature changes of the individual tubes along the longitudinal axis of the active regenerator after the AGRR cycle as described earlier in FIG. **3**. In other words, FIG. **5**A depicts the temperature span from a cold temperature to a hot temperature of the individual refrigerant-filled tubes along the longitudinal axis of each AGRR active regenerator. FIG. **5**B shows a side view of the layers of thin-walled tubes as described in FIG. **4**. FIG. **5**C shows the steady state of temperature of the array of layered tubes along the active regenerator after a significant period of operation. FIG. **5**D shows the temperature profile of the refrigerant-filled tubes after the compression step of the cycle, the temperature in each refrigerant-filled tube having increased. FIG. **5**E shows the temperature profile after the hot blow period of the active regenerator with heat transfer fluid going from left to right in the figure. FIG. **5**F shows the temperature reduction of the refrigerant-filled tubes in the regenerator after the expansion step. FIG. **5**G shows the temperature profile after the cold blow of the cycle where the heat transfer fluid flows from right to left in the figure.

[0056] In the hot blow, the heat transfer gas comes out of the right end of the active regenerator at a temperature $T_{hot}+\phi\Delta T_{hot}$ where ϕ ranges from 1 to 0 during the blow period (usually ϕ averages about 0.5) and ΔT_{hot} is the temperature change of the hottest layer of tubes. This hot gas can reject heat to the cooling fluid as the heat transfer fluid cools back toward T_{hot}. Similarly, during the cold blow, the heat transfer gas comes out of the left end of active regenerator at a temperature of $T_{cold}-\phi\Delta T_{cold}$ and it can absorb heat from the thermal load from the process stream as it warms toward T_{cold}. The operation of this active regenerator is similar to that of high performance regenerators in other regenerative refrigerators with the added feature that each refrigerant-filled tube in the regenerator has the ability to actively change its temperature and thus enable distributed refrigeration within the regenerator rather than just be a passive heat sink/source as is traditionally the case. Hence, there is a large surface area for heat transfer between the tubes and the heat transfer fluid in the preferred embodiment. Also, the pressure drop to flow, the longitudinal (axial) conduction, and porosity of the regenerator are as small as possible in the preferred embodiment. However, other active regenerator configurations may be used in other embodiments. The temperature increases of the refrigerant-filled tubes along the regenerator longitudinal axis must satisfy the second law of thermodynamics and be in the ideal ratio of absolute temperatures along the regenerator as has been schematically illustrated in FIG. **5**; frames D and F.

[0057] The cooling power of the AGRR stage at the cold temperature, the hot sink temperature, the thermal load temperature, the heat transfer fluid pressure, the effective tube wall temperature change of the cold heat transfer fluid leaving the cold end of the dual active regenerators, and the ϕ factor that specifies the fraction of the effective tube wall temperature change that can be used for a given heat transfer fluid flow period are all design variables that can be set. The flow rate of the He heat transfer fluid, $\dot{m}_{He}$, is calculated from the equation:

$$\dot{Q}_C=\dot{m}_{He}c_p\Delta T_C\Phi$$

where $\dot{Q}_C$ is the cooling power at the cold temperature, T_C, c_p is the heat capacity of He at constant pressure, and ΔT_C is the effective tube wall temperature change at the coldest row of refrigerant-filled tubes in AGRR active regenerator. The typical variables for an active regenerator with 50 W of cooling power at 240 K are presented in Table 1 below.

TABLE 1

Typical AGRR stage specifications for efficiency design calculations

Q_{Cdot} (W)	T_H (K)	T_C (K)	ΔT_C (K)	Φ (dim)	He heat transfer gas pressure (psia)	He heat transfer flow rate (g/sec)
50	290	240	10	0.5	200	1.93

[0058] In the preferred embodiment, a rectangular tube array is used with a staggered tube arrangement in successive layers from the cold end to the hot end of the dual active regenerators. The length in the x direction is the active length of the refrigerant-filled tubes, or the longitudinal axis of the regenerator. The length in the y direction is the length of each row of refrigerant-filled tubes where the number of tubes depends upon the tube diameter and separation between each tube in a row. The z direction is the heat transfer fluid flow direction and this length is determined by the tube diameter and the separation between the layers of rows of tubes. The total number of tubes in the dual active regenerators on each side of the AGRR stage is the product of the number of tubes in each row and the number of layers of rows. In one embodiment, the length of x and y have the same value and the z length has a value three times the y length, although other dimensions are possible. The primary independent variable becomes the tube diameter once the rectangular dimensions of the dual regenerators are chosen with the constraints above.

[0059] The average He heat transfer gas properties at 200 psia can be calculated at the average temperature of the dual active regenerators. The density, heat capacity, viscosity, and thermal conductivity can be obtained and used to calculate the Prandtl number. The Reynolds number of the heat transfer fluid flow can be determined using the accepted equation from the literature for this geometry. For the various configurations, the heat transfer coefficient, the friction factor, and the effective thermal conductivity can be calculated. These values can then be used to calculate the entropy generated from the three mechanisms described above and the total entropy generated can then be used to obtain the FOM of the AGRL.

[0060] Of immediate note is that the AGRR stage efficiencies ranges from a relatively small value in the case with 10 total tubes of 0.635 cm (0.25") outer diameter, 2.5 cm length, and a reasonable ΔT of 10 K to a very impressive value in the case where 0.15875 cm (1/16") outer diameter, 7.5 cm length, and a higher but achievable ΔT of 15 K. These dimensions along with Table 2 are provided as examples of one embodiment only and should not be construed to limit the scope of the disclosed invention.

TABLE 2

Calculated efficiencies for various AGRR stage geometries with staggered tubes. AGRR dimensions are shown.

ΔT_C (K)	L_x (cm)	L_y (cm)	L_z (cm)	D_t (cm)	S_y (cm)	S_z (cm)	$N_{tubes/row}$	$N_{layers/AGRR}$	N_{total}	Efficiency
15	7.5	7.5	22.5	0.3175	0.158	0.3175	15	35	525	0.77
15	7.5	7.5	22.5	0.1588	0.079	0.1588	31	70	2170	0.82

[0061] In the preferred embodiment, each of the dual regenerators in each AGRR stage has the following characteristics:

[0062] high specific area (~10,000 m^2/m^3)

[0063] very thin wall tubes

[0064] appropriate ΔT vs. T characteristics

[0065] mechanically strong enough for modest pressures

[0066] leak tight and able to withstand cyclic mechanical pressure loads

[0067] low pressure drop

[0068] high transverse and low longitudinal (axial) conductivity and

[0069] mass producible at low/modest cost.

[0070] As mentioned, one variable that can be manipulated to affect the efficiency of the AGRR stage is the effective adiabatic temperature change of the refrigerant-filled small diameter thin-walled tubes. For example, a 5/32" outer diameter×0.003" wall stainless tubing with a compression pressure ratio of 3 has a possible ΔT of about 19 K with a mean operating temperature of 300 K and an initial He refrigerant pressure of 215 psia.

[0071] In the preferred embodiment of a hydrogen AGRL, the thermal load, $\dot{Q}_{load}$, from the hydrogen process stream is transferred via convective heat transfer to the heat transfer flow stream for dual active regenerators from a continuous stream in the circulator and heat rejection exchanger. The pressure drop of the active regenerator and heat exchanger combinations can be kept reasonably low by design choices, so this circulator needs high volumetric efficiency at relatively low head. The extra work for this component of each AGRR stage is modest so its effect on overall efficiency of each stage and FOM of the AGRL is relatively small. The operation of the active regenerator in this AGRR stage is similar to that in any regenerative refrigerator with the added feature that each refrigerant-filled tube in the active regenerator has the ability to provide refrigeration rather than acting only as a passive heat sink/source. The selection of many small diameter tubes in the preferred embodiment creates a large surface area for heat transfer between the refrigerant-filled tubes and the heat transfer fluid, thereby reducing the dominant irreversible entropy mechanism in the active regenerator, in each AGRR stage, and in the AGRL as a whole.

[0072] The preferred embodiment of a hydrogen AGRL includes the following features:

[0073] Active regenerator tube arrays, refrigerant, and passive micro-regenerator arrays for each AGRR stage

[0074] Heat transfer fluid periodic flow assemblies for each AGRR stage

[0075] mechanical drive for displacer or circulators

[0076] fluid transfer system pipes, heat exchangers, flow control valves, seals, insulation, circulators and/or blowers

[0077] Compressor/expander assemblies for each AGRR stage

[0078] mechanical drive for the compressor/expander for each AGRR stage

[0079] System integration, heat exchanger assemblies, skid for mounting all vessels, etc.

[0080] cold box for He/H_2 process stream heat exchangers

[0081] He/H_2O heat sink heat exchangers at ~300 K

[0082] He/H_2 heat exchangers with ortho-to-para hydrogen catalysts

[0083] H_2 supply and LH_2 storage vessels

[0084] Auxiliary systems

[0085] electrical power for the AGRL

[0086] vacuum station for start-up and cryopump for long-term operation

[0087] Instrumentation and control

[0088] temperature, pressure, velocity, loads, power, and flow rate transducers

[0089] flammable gas detectors, shut-down valves, process stream pressure and flow rate controls, level and pump controls

[0090] control panel, DAQ racks, PC and PC interface/software.

Note that although the preferred embodiment would contain all of the foregoing elements, one could manufacture an AGRL that incorporates only some of the above.

[0091] A hydrogen AGRL also preferably has the characteristics described in Table 3 for each AGRR stage:

TABLE 3

Design calculations of the six hydrogen AGRR stages. Key parameters for each stage are: hi = 0.75, i = 1–6, ΔT_{ad} = 15 K, Φ = ½, f = 1 Hz.

Stage	He temperatures at the Cold HEX T_{cold}[K]	ΔT_{cold}[K]	Heat transfer fluid (He) flow rate [g/sec]	Total length of Regenerator tubes per stage [m]
1	200	7.85	0.39	41.0
2	134	4.95	0.40	81.6
3	90	3.19	0.43	113
4	60	2.07	0.53	141
5	40	1.36	0.66	150
6	27	0.91	1.62	281

[0092] With these total regenerator tube lengths, the geometries of each AGRR stage can be obtained with choices of 1_x, 1_y, and 1_z, the respective lengths of the rectangular regenerator as described above. Table 4 summarizes the selected geometries of each AGGR stage of one embodiment of the AGRL with tubes of diameter 0.3175 cm. Again, these geometries could be varied.

TABLE 4

Selected/calculated dimensions and the total number of regenerator tubes of the six AGRR stages of a 1 kg/day LH2 AGRL.

AGRR Stage	L_x (cm)	L_y (cm)	L_z (cm)	Tubes/layer	Total tubes
1	7.5	10	13	21	547
2	7.5	10	25	21	1088
3	7.5	10	34	21	1518
4	7.5	15	29	32	1884
5	7.5	15	30	32	1998
6	7.5	15	57	32	3753

[0093] FIG. **6** schematically illustrates the operation of the first AGRR stage (**12**) of one embodiment of an AGRL configured as a hydrogen liquefier. As described above, the He heat transfer fluid system connects the dual active gas regenerators (**20**) of each AGRR stage to a heat sink exchanger (**17**) and a process stream heat exchanger (**15**). The operation of the circulator (**32**) is synchronized with the reciprocating motion of the pistons (**28**) of the compressor/expander assembly (**24**) to pump heat from the process stream (**11**) at the cold heat exchanger (**14**) via the process stream heat exchangers (**15**) to the heat rejection exchanger (**13**) via the heat sink exchangers (**17**). A three-way rotary valve (**36**) facilitates this operation, as will be described here in detail. The three-way valves are white if open for flow and black if closed to flow. Note that to simplify depiction of the flow paths in FIG. **6**, the cold ends of the dual active regenerators (**20**) are at the ends nearest each other in the figure in contrast to that orientation shown in FIG. **2**.

[0094] For purposes of this description, the refrigerant (**23**) in the upper dual active regenerator (**20**) of FIG. **6** has been compressed, resulting in a temperature increase in the refrigerant-filled tubes (**21**). The refrigerant (**23**) in the lower dual active regenerator (**20**) of FIG. **6** has been expanded, resulting in a temperature increase in the refrigerant-filled tubes (**21**). The heat transfer fluid (**35**) enters through the lower regenerator (**20**) and flows toward the top of the figure, executing a cold blow. During the cold blow, the lower regenerator (**20**) accepts heat from the He heat transfer fluid (**35**), thereby lowering the temperature of the heat transfer fluid (**35**) to about $T_{cold}-\Delta T_{cold}$. This cold heat transfer fluid (**35**) then circulates through the cold heat exchanger (**14**) thermally coupled to the process stream heat exchanger (**15**), thus accepting heat from the thermal load of the hydrogen process stream (**11**) as explained above. After accepting heat from the process stream (**11**), the He heat transfer fluid (**35**) returns through the upper active regenerator (**20**) of this AGRR stage and flows toward the top of the figure, executing a hot blow. During the hot blow, the heat transfer fluid accepts heat from the upper regenerator (**20**), thereby elevating the temperature of the heat transfer fluid (**27**) to about $T_{hot}+\Delta T_{hot}$ at the exit of the upper dual active regenerator. The heat transfer fluid (**35**) then circulates through the heat rejection exchanger (**13**), where the cooling fluid (**16**) accepts heat from the AGRR stage.

[0095] Each of the three-way valves (**36**) can be switched, and the pistons (**28**) of the compressor/expander expand the refrigerant (**23**) in the upper active regenerator (**20**) and compress the refrigerant (**23**) in the lower regenerator (**20**). The use of four three-way valves (**36**) eliminate the necessity of reversing the flow of the heat transfer fluid (**35**) through the system, insure counterflow fluid flows in both the cold heat exchanger and heat rejection exchanger at all times, and eliminate two heat exchangers as depicted in the embodiment of FIG. **2**. These valves (**36**) are positioned on either side of the active regenerators (**20**).

[0096] The second half of the cycle is analogous to the first half of the cycle. In this instance the heat transfer fluid (**35**) flows through the upper regenerator (**20**) toward the bottom of the figure and is cooled by the refrigerant-filled tubes before passing through the cold heat exchanger (**14**) thermally coupled to the process stream heat exchanger (**15**) to accept heat from the process stream (**11**). The heat transfer fluid (**35**) then circulates through the lower regenerator (**20**), flowing toward the bottom of the figure, where it accepts heat from the refrigerant-filled tubes. This heat is expelled when the heat transfer fluid (**35**) passes through the heat rejection exchanger (**13**) that is thermally coupled to the heat sink exchanger (**17**). The valves (**36**) are then switched back to the configuration in FIG. **6**. This sequence of events is continuously repeated at the frequency of about 1 Hz. The layout of each stage of an AGRL will be approximately the same except the temperature spans across the subsequent AGRR stages will increase as the process stream is further cooled toward liquefaction.

[0097] Various heat exchangers may be used for the AGRL. In one embodiment, small brazed-plate heat exchangers are used for the process stream heat exchangers. The heat rejection and heat sink exchangers may be compact plate-fin liquid-to-gas exchangers that have been used successfully in previous LNG liquefier designs and are available in the prior art. The duty of these exchangers is significantly more than the process stream heat exchangers. In the preferred embodiment, each AGRR stage rejects heat to a common water stream (the cooling fluid) that subsequently rejects heat to the environment through a water to air heat exchanger. The heat exchanger for removing excess heat from the cooling fluid can take the form of a finned tube with air cross flow as used in many air conditioners or similar vapor compression cycle refrigerators. These two operations are well defined and understood by those skilled in the art.

[0098] As mentioned, the preferred embodiment includes a cold box. Preferably, this cold box is vacuum insulated and of a simple design with a top plate for mounting the six stages of the AGRL. The assembly can be done easily with a crane to raise or lower the AGRL into the cold box. In this embodiment, the cold box is evacuated with a high vacuum turbo pump prior to operation. The cold box vacuum can be maintained by Cryopumping with zeolite containers attached to the cold end of one or the stages (e.g., an about 40 K stage because zeolite will adsorb helium at this temperature). Superinsulation is wrapped on all the cryogenic sections of the AGRL to reduce radiative heat leaks. All instrumentation can be mounted to the individual AGRR stages and the process stream may enter through vacuum feed throughs in the top plate of the cold box. The controls for the flows, the compressor/expanders, all valve drive motor controllers, and other operational components can be located outside the cold box.

[0099] In the preferred embodiment, the cryogenic liquid produced (i.e., LNG or LH_2) is stored in a small vessel within the cold box with a double-walled vacuum jacketed or suitably insulated transfer line out of the vessel through the top plate for storage in an external cryogenic storage vessel. A level detector and or a mass flow meter may be placed in the vessel to directly measure the rate of liquefaction.

[0100] While the invention is susceptible to various modifications and alternative forms, specific embodiments thereof have been shown by way of example in the drawings and herein described in detail. For example, although the refrigerant has been described throughout as helium gas, any fluid that can be compressed or expanded to cause a temperature change in the desired range may be used. Other mixed refrigerants or combined compressor/expander assemblies are also possible. One skilled in the art will recognize suitable substitutions. Similarly, the dimensions of the various components may be varied. The AGRL may be configured to liquefy other fluids. It should be understood, however, that it 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 described.

We claim:

1. An active gas regenerative liquefier (AGRL), comprising at least a first active gas regenerative refrigerator (AGRR) stage and a second AGRR stage, the first AGRR stage configured to receive and cool a process stream, and deliver the process stream to the second AGRR stage, wherein the first and second AGRR stages have means for heat rejection to a common heat sink.

2. The AGRL of claim **1**, in which the second AGRR stage and any subsequent AGRR stage is configured to successively receive and cool the process stream.

3. The AGRL of claim **2**, further comprising at least a third AGRR stage for the liquefaction of natural gas.

4. The AGRL of claim **2**, further comprising at least a third AGRR stage, wherein the first through third AGRR stages have respective cold reservoirs at temperatures of about 220 K, 164 K, and 123 K, respectively.

5. The AGRL of claim **2**, further comprising at least a third, a fourth, a fifth, and a sixth AGRR stage for the liquefaction of hydrogen.

6. The AGRL of claim **2**, further comprising at least a third, a fourth, a fifth, and a sixth AGRR stage, wherein the first through sixth AGRR stages have respective cold reservoirs at temperatures of about 192 K, 120 K, 76 K, 48 K, 32 K, and 20 K, respectively.

7. The AGRL of claim **2** in which the process stream is hydrogen gas, further comprising at least one ortho to para converter located between any two sequential AGRR stages.

8. An active gas regenerative liquefier (AGRL), comprising:

at least one active gas regenerative refrigerator (AGRR) stage, said AGRR stage comprising at least two active regenerators, means for heat rejection to a common heat sink near room temperature, and means to provide work to compress a refrigerant and simultaneously recover work from expansion of the refrigerant;

said AGRR stage configured to receive and cool a process stream to the point of liquefaction, and deliver the liquefied process stream to a storage vessel.

9. The AGRL of claim **2**, in which the AGRR stage comprises:

a first active regenerator and a second active regenerator, the first active regenerator and the second active regenerator each comprising an array of tubes, wherein at least one passive micro-regenerator is located at an entrance of each of said tubes, said tubes containing a refrigerant;

a compressor/expander assembly, said assembly comprising a manifold having a first portion and a second portion, a first piston in said first portion of said manifold, and a second piston in said second portion of said manifold, said array of tubes of the first active regenerator being connected to said first portion of said manifold and said array of tubes of the second active regenerator being connected to said second portion of said manifold, said manifold also containing the refrigerant, said pistons being configured to separately periodically compress and expand the refrigerant to thereby increase or decrease the temperature of the refrigerant in said tubes;

means to drive the pistons such that one piston compresses the refrigerant in one portion of said manifold while the other piston expands the refrigerant in the other portion of said manifold, thereby enabling work recovery from expansion to offset work required for compression;

means to circulate a heat transfer fluid between the process stream and the heat sink, said heat transfer fluid circulating past the first active regenerator and the second active regenerator in order to accept heat from or transfer heat to said active regenerators;

at least a first process stream heat exchanger for exchanging heat from said process stream to said heat transfer fluid; and

at least a first heat rejection exchanger for exchanging heat from said heat transfer fluid to said heat sink.

10. The AGRL of claim **9**, in which the means to circulate the heat transfer fluid is coupled in phase with the means to drive the pistons, such that the heat transfer fluid circulates along a first flow path while the refrigerant in the first portion of the manifold is expanded and along a second flow path while the refrigerant in the second portion of the manifold is expanded.

11. The AGRL of claim **10**, further comprising:

a second process stream heat exchanger, a second heat rejection exchanger, a first valve connecting the first process stream heat exchanger, the second process stream heat exchanger, and the process stream, and a second valve connecting the first heat rejection exchanger, the second heat rejection exchanger, and the heat sink;

where said means to circulate the heat transfer fluid comprises a circulator that directs the heat transfer fluid in an oscillatory manner along the first flow path in which the heat transfer fluid flows sequentially through the first active regenerator, the first process stream heat exchanger, the second process stream heat exchanger, the second active regenerator, and the second heat rejection exchanger and along the second flow path sequentially through the second active regenerator, the second process stream heat exchanger, the first process stream heat exchanger, the first active regenerator, and the first heat rejection exchanger;

said first valve configured to direct the process stream through the first process stream heat exchanger when the heat transfer fluid flows along the first flow path and to direct the process stream through the second process stream heat exchanger when the heat transfer fluid flows along the second flow path;

said second valve configured to direct a cooling fluid through the second heat rejection exchanger when the heat transfer fluid flows along the first flow path and to direct the cooling fluid through the first heat rejection exchanger when the heat transfer fluid flows along the second flow path.

12. The AGRL of claim **10**, in which the means to circulate the heat transfer fluid comprises a plurality of valves configured to direct the flow of the heat transfer fluid alternately along the first flow path in which the heat transfer fluid passes sequentially through the first active regenerator, the process stream heat exchanger, the second active regenerator, and the heat rejection exchanger, and the second flow path in which the heat transfer fluid passes sequentially through the second active regenerator, the process stream heat exchanger, the first active regenerator, and the heat rejection exchanger.

13. The AGRL of claim **12**, in which a first valve is positioned between the first active regenerator and the heat rejection exchanger, a second valve is positioned between the first active regenerator and the process stream heat exchanger, a third valve is positioned between the second active regenerator and the heat rejection exchanger, and a fourth valve is positioned between the second active regenerator and the process stream heat exchanger.

14. The AGRL of claim **12**, in which the valves maintain continuous counterflow of the heat transfer fluid in the process stream heat exchanger and the heat rejection exchanger with periodic heat transfer fluid flow from the first and the second active regenerators.

15. The AGRL of claim **9**, wherein the tubes of each active regenerator are arranged in a plurality of layers with temperatures in the tubes spanning from a cold temperature in a bottom layer to a hot temperature in a top layer, such that the first piston performs distributed work as a function of the temperature in the tubes during compression of the refrigerant and the second piston simultaneously recovers distributed work as a function of the temperature in the tubes during expansion of the refrigerant.

16. The AGRL of claim **9**, wherein the passive micro-regenerator comprises spheres with a diameter less than the diameter of the entrance of the tube and with a thermal mass several times greater than the thermal mass of the refrigerant.

17. The AGRL of claim **9**, wherein the passive micro-regenerator comprises screens with a diameter approximately equal to the diameter of the entrance of the tube and with a thermal mass several times greater than the thermal mass of the refrigerant.

18. The AGRL of claim **9**, in which the temperature differences between the heat transfer fluid and the refrigerant within the active regenerators are 2 K or less.

19. The AGRL of claim **9**, in which the temperature differences between the heat transfer fluid and a cooling fluid in the heat rejection exchanger, and between the heat transfer fluid and the process stream in the process stream heat exchanger are 10's of K or less.

20. A natural gas liquefier, comprising at least three active gas regenerative refrigerator (AGRR) stages situated and configured to receive and sequentially cool a natural gas process stream.

21. A hydrogen liquefier, comprising at least six active gas regenerative refrigerator (AGRR) stages situated and configured to receive and sequentially cool a hydrogen process stream and an ortho to para converter situated between each of the AGRR stages.

22. A method of liquefying a process stream of gas, comprising the steps of:

A. Cooling a heat transfer fluid by passing it through a first active regenerator in which a refrigerant has been cooled by expanding said refrigerant;

B. Circulating said heat transfer fluid through a process stream heat exchanger for exchanging heat from the process stream to the heat transfer fluid;

C. Passing said heat transfer fluid through a second active regenerator in which a refrigerant has been heated by compression of said refrigerant;

D. Circulating said heat transfer fluid through a heat rejection exchanger for exchanging heat from said heat transfer fluid to a heat sink;

E. Repeating steps A-D multiple times in a first stage of refrigeration to cool said process stream to a first temperature; and

F. Repeating steps A-D multiple times in at least one subsequent stage of refrigeration to further cool said process stream to successively lower temperatures until liquefaction occurs.

23. The method of claim **22**, in which expansion of the refrigerant in step A is simultaneous with compression of the refrigerant in step C, such that a distributed work of compression of the refrigerant is offset by a distributed work of expansion of the refrigerant.

24. A method of making a highly efficient liquefier, comprising the steps of

A. providing at least one stage of refrigeration, said stage comprising a first active regenerator, a second active regenerator, and a refrigerant;

B. providing means to input work via compression of the refrigerant, said input work distributed over a first temperature span in the first active regenerator of each stage;

C. providing means to recover work via expansion of the refrigerant, said recovered work distributed over a second temperature span in the second active regenerator of each stage, said second temperature span being lower than said first temperature span;

D. providing means to couple said means for work input and said means for work recovery enabling the work input to be offset by the work recovered;

E. maintaining small temperature differences wherever heat transfer occurs; and

F. simultaneously optimizing heat transfer, pressure drops, and longitudinal conduction, friction losses, and other parasitic heat leaks in the liquefier operation.

25. The method of claim **24**, in which the first and the second temperature spans range from near cryogenic temperatures to near room temperature and in which the means to input and recover work are accomplished by polytropic temperature changes of the refrigerant from distributed compression or expansion.

26. The method of claim **25**, wherein the distributed compression or expansion of the refrigerant causes temperature changes of between 15 K and 20 K.

27. The method of claim **24**, wherein the means to couple means for work input and said means for work recovery comprises a resonant piston compressor/expander in a common cylinder.

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