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(54) Title: HYDROCARBON GAS PROCESSING

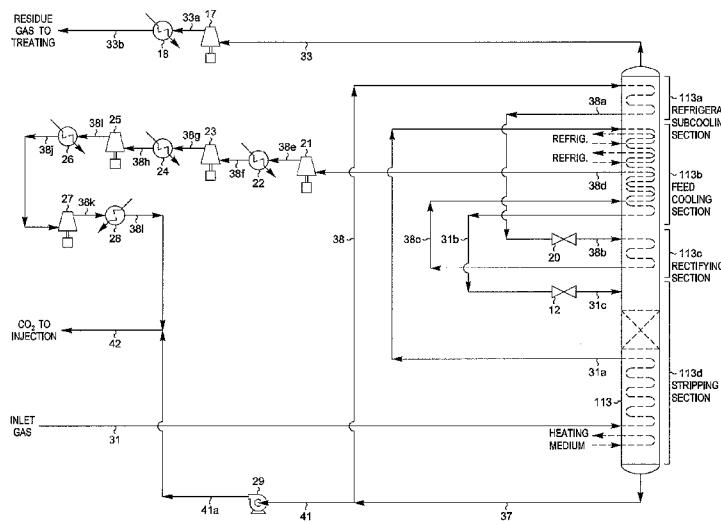


FIG. 2

(57) Abstract: A process and an apparatus are disclosed for removal of carbon dioxide from hydrocarbon gas streams in a processing assembly. The stream is cooled, expanded to intermediate pressure, and supplied as a top feed to a mass transfer means inside the processing assembly. A distillation liquid stream is collected from the lower region of the mass transfer means and directed into a first heat and mass transfer means inside the processing assembly to heat it and strip out its volatile components while cooling the gas stream, thereafter discharging the heated and stripped distillation liquid stream from the processing assembly as the bottom liquid product therefrom. The quantities and temperatures of the feeds to the mass transfer means are effective to maintain the temperature of the upper region of the mass transfer means at a temperature whereby the major portion of said carbon dioxide is recovered in the bottom liquid product.

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## HYDROCARBON GAS PROCESSING

### SPECIFICATION

#### BACKGROUND OF THE INVENTION

[0001] This invention relates to a process and apparatus for the separation of a gas containing hydrocarbons and carbon dioxide. The applicants claim the benefits under Title 35, United States Code, Section 119(e) of prior U.S. Provisional Application Number 61/186,361 which was filed on June 11, 2009. The applicants also claim the benefits under Title 35, United States Code, Section 120 as a continuation-in-part of U.S. Patent Application No. 12/781,259 which was filed on May 17, 2010, and as a continuation-in-part of U.S. Patent Application No. 12/772,472 which was filed on May 3, 2010, and as a continuation-in-part of U.S. Patent Application No. 12/750,862 which was filed on March 31, 2010, and as a continuation-in-part of U.S. Patent Application No. 12/717,394 which was filed on March 4, 2010, and as a continuation-in-part of U.S. Patent Application No.

12/689,616 which was filed on January 19, 2010, and as a continuation-in-part of U.S. Patent Application No. 12/372,604 which was filed on February 17, 2009. Assignees S.M.E. Products LP and Ortloff Engineers, Ltd. were parties to a joint research agreement that was in effect before the invention of this application was made.

[0002] Hydrocarbons are found in a variety of gases, such as natural gas, refinery gas, and synthetic gas streams obtained from other hydrocarbon materials such as coal, crude oil, naphtha, oil shale, tar sands, and lignite. The hydrocarbon constituents of natural gas usually have a major proportion of methane and ethane, i.e., methane and ethane together comprise at least 50 mole percent of the total hydrocarbon components. The gas also usually contains relatively lesser amounts of heavier hydrocarbons such as propane, butanes, pentanes, and the like, as well as carbon dioxide, hydrogen, nitrogen, and other gases.

[0003] In many cases, the gas streams from these sources are contaminated with high concentrations of carbon dioxide, making the gas streams unsuitable for use as fuel, chemical plant feedstock, or other purposes. There are a variety of processes that have been developed to remove the carbon dioxide using chemical, physical, and hybrid solvents. Other processes have been developed that use a refrigerated absorbent stream composed of heavy (C<sub>4</sub>-C<sub>10</sub> typically) hydrocarbons to remove the carbon dioxide in a distillation column, such as the process described in U.S. Patent No. 4,318,723. All of these processes have increasingly higher capital cost and operating cost as the carbon dioxide concentration in the gas stream increases, which often makes processing of such gas streams uneconomical.

[0004] One method for improving the economics of processing gas streams containing high concentrations of carbon dioxide is to provide bulk separation of the carbon dioxide from the gas stream prior to processing with solvents or absorbents, so that only a minor fraction of the carbon dioxide must then be removed from the gas stream. For example, semi-permeable membranes have often been used for bulk removal of carbon

dioxide. However, a significant fraction of the lighter hydrocarbons in the gas stream are often "lost" in the carbon dioxide stream that is separated by bulk removal processes of this type.

[0005] A better alternative for bulk removal of carbon dioxide is to use distillation to fractionate the gas stream into a light hydrocarbon stream and a carbon dioxide stream, so that removal of the residual carbon dioxide from the light hydrocarbon stream is all that is required to produce pipeline-quality gas for use as fuel, chemical plant feedstock, etc. The majority of the carbon dioxide that is removed is recovered as a liquid rather than a vapor, allowing the carbon dioxide to be pumped (rather than compressed) for subsequent use in tertiary oil recovery operations or for other purposes, resulting in substantial reductions in capital cost and operating cost.

[0006] The present invention is generally concerned with the removal of the majority of the carbon dioxide from such gas streams. A typical analysis of a gas stream to be processed in accordance with this invention would be, in approximate mole percent, 31.0% methane, 0.4% ethane and other C<sub>2</sub> components, 0.1% propane and other C<sub>3</sub> components, 0.1% butanes plus, and 67.7% carbon dioxide, with the balance made up of nitrogen. Sulfur containing gases are also sometimes present.

[0007] In a typical distillation process for removing carbon dioxide, a feed gas stream under pressure is cooled by heat exchange with other streams of the process and/or external sources of refrigeration such as a Freon compression-refrigeration system. The gas is condensed as it is cooled, and the high-pressure liquid is expanded to an intermediate pressure, resulting in further cooling of the stream due to the vaporization occurring during expansion of the liquids. The expanded stream, comprising a mixture of liquid and vapor, is fractionated in a distillation column to separate residual methane, nitrogen, and other volatile gases as overhead vapor from the carbon dioxide and the heavier hydrocarbon components as

bottom liquid product. A portion of the liquid carbon dioxide can be flash expanded to lower pressure and thereafter used to provide low level refrigeration to the process streams if desired.

[0008] The present invention employs a novel means of performing the various steps described above more efficiently and using fewer pieces of equipment. This is accomplished by combining what heretofore have been individual equipment items into a common housing, thereby reducing the plot space required for the processing plant and reducing the capital cost of the facility. Surprisingly, applicants have found that the more compact arrangement also reduces the power consumption required to achieve a given level of carbon dioxide removal, thereby increasing the process efficiency and reducing the operating cost of the facility. In addition, the more compact arrangement also eliminates much of the piping used to interconnect the individual equipment items in traditional plant designs, further reducing capital cost and also eliminating the associated flanged piping connections. Since piping flanges are a potential leak source for carbon dioxide (a greenhouse gas) and for hydrocarbons (which are volatile organic compounds, VOCs, that contribute to greenhouse gases and may also be precursors to atmospheric ozone formation), eliminating these flanges reduces the potential for atmospheric emissions that can damage the environment.

[0009] In accordance with the present invention, it has been found that more than 87% of the carbon dioxide can be removed while leaving more than 99% of the methane in the residue gas stream. The present invention, although applicable at lower pressures and warmer temperatures, is particularly advantageous when processing feed gases in the range of 400 to 1500 psia [2,758 to 10,342 kPa(a)] or higher under conditions requiring distillation column overhead temperatures of -50°F [-46°C] or colder.

[0010] For a better understanding of the present invention, reference is made to the following examples and drawings. Referring to the drawings:

[0011] FIG. 1 is a flow diagram of a prior art natural gas processing plant;

[0012] FIG. 2 is a flow diagram of a natural gas processing plant in accordance with the present invention; and

[0013] FIGS. 3 and 4 are flow diagrams illustrating alternative means of application of the present invention to a natural gas stream.

[0014] In the following explanation of the above figures, tables are provided summarizing flow rates calculated for representative process conditions. In the tables appearing herein, the values for flow rates (in moles per hour) have been rounded to the nearest whole number for convenience. The total stream rates shown in the tables include all non-hydrocarbon components and hence are generally larger than the sum of the stream flow rates for the hydrocarbon components. Temperatures indicated are approximate values rounded to the nearest degree. It should also be noted that the process design calculations performed for the purpose of comparing the processes depicted in the figures are based on the assumption of no heat leak from (or to) the surroundings to (or from) the process. The quality of commercially available insulating materials makes this a very reasonable assumption and one that is typically made by those skilled in the art.

[0015] For convenience, process parameters are reported in both the traditional British units and in the units of the Système International d'Unités (SI). The molar flow rates given in the tables may be interpreted as either pound moles per hour or kilogram moles per hour. The energy consumptions reported as horsepower (HP) and/or thousand British Thermal Units per hour (MBTU/Hr) correspond to the stated molar flow rates in pound moles per hour. The energy consumptions reported as kilowatts (kW) correspond to the stated molar flow rates in kilogram moles per hour.

DESCRIPTION OF THE PRIOR ART

[0016] FIG. 1 is a process flow diagram showing the design of a processing plant to remove carbon dioxide from natural gas using a prior art process. In this simulation of the process, inlet gas enters the plant at 81°F [27°C] and 1110 psia [7,653 kPa(a)] as stream 31. The feed stream is usually dehydrated to prevent hydrate (ice) formation under cryogenic conditions. Solid and liquid desiccants have both been used for this purpose.

[0017] The feed stream 31 is cooled to 1°F [-17°C] in heat exchanger 10 by heat exchange with column reboiler liquids at 43°F [6°C] (stream 36), column side reboiler liquids at 30°F [-1°C] (stream 35), and Freon R134a refrigerant. Stream 31a is further cooled in heat exchanger 11 by heat exchange with cool carbon dioxide vapor at -49°F [-45°C] (stream 39), cold residue gas at -60°F [-51°C] (stream 33), and pumped liquid at -60°F [-51°C] (stream 34a). The further cooled stream 31b at -14°F [-25°C] and 1060 psia [7,308 kPa(a)] is expanded to the operating pressure (approximately 617 psia [4,254 kPa(a)]) of fractionation tower 13 by expansion valve 12, cooling stream 31c to -35°F [-37°C] before it is supplied to fractionation tower 13 at its top column feed point.

[0018] Overhead vapor stream 32 leaves fractionation tower 13 at -35°F [-37°C] and is cooled and partially condensed in heat exchanger 14. The partially condensed stream 32a enters separator 15 at -60°F [-51°C] and 610 psia [4,206 kPa(a)] where the vapor (cold residue gas stream 33) is separated from the condensed liquid (stream 34). Liquid stream 34 is pumped to slightly above the operating pressure of fractionation tower 13 by pump 19 before stream 34a enters heat exchanger 11 and is heated to -6°F [-21°C] and partially vaporized by heat exchange with the feed gas as described previously. The partially vaporized stream 34b is thereafter supplied as feed to fractionation tower 13 at a mid-column feed point.

[0019] Fractionation tower **13** is a conventional distillation column containing a plurality of vertically spaced trays, one or more packed beds, or some combination of trays and packing. It also includes reboilers (such as the reboiler and the side reboiler described previously) which heat and vaporize a portion of the liquids flowing down the column to provide the stripping vapors which flow up the column to strip the column bottom liquid product (stream **37**) of methane and lighter components. The trays and/or packing provide the necessary contact between the stripping vapors rising upward and cold liquid falling downward, so that the bottom product stream **37** exits the bottom of the tower at 45°F [7°C], based on reducing the methane concentration in the bottom product to 0.50% on a molar basis.

[0020] Column bottom product stream **37** is predominantly liquid carbon dioxide. A small portion (stream **38**) is subcooled in heat exchanger **16** by cool residue gas stream **33a**. The subcooled liquid (stream **38a**) at 0°F [-18°C] is expanded to lower pressure by expansion valve **20** and partially vaporized, further cooling stream **38b** to -63°F [-53°C] before it enters heat exchanger **14**. The residual liquid in stream **38b** functions as refrigerant in heat exchanger **14** to provide cooling of stream **32** as described previously, with the resulting carbon dioxide vapor leaving at -49°F [-45°C] as stream **39**. Since stream **38b** contains a small amount of heavier hydrocarbons, a small liquid purge (stream **40**) is drawn off from heat exchanger **14** to prevent an accumulation of heavier hydrocarbons in the refrigerant liquid that could elevate its boiling point and reduce the cooling efficiency of heat exchanger **14**.

[0021] The cool carbon dioxide vapor from heat exchanger **14** (stream **39**) is heated to -6°F [-21°C] in heat exchanger **11** by heat exchange with the feed gas as described previously. The warm carbon dioxide vapor (stream **39a**) at 76 psia [524 kPa(a)] is then compressed to high pressure in four stages by compressors **21**, **23**, **25**, and **27**, with cooling to

120°F [49°C] after each stage of compression by discharge coolers **22**, **24**, **26**, and **28**. The remaining portion (stream **41**) of column bottom product stream **37** is pumped to high pressure by pump **29** so that stream **41a** can combine with the dense phase fluid (stream **39i**) leaving discharge cooler **28**, forming high pressure carbon dioxide stream **42** which then flows to reinjection at 79°F [26°C] and 2015 psia [13,893 kPa(a)].

[0022] The cool residue gas (stream **33a**) leaves heat exchanger **11** at -6°F [-21°C] after heat exchange with the feed gas as described previously, and is further heated to 27°F [-3°C] in heat exchanger **16** by heat exchange with liquid carbon dioxide stream **38** as described previously. The warm residue gas stream **33b** is then re-compressed by compressor **17** and cooled to 120°F [49°C] in discharge cooler **18** before it flows to treating at 1215 psia [8,377 kPa(a)].

[0023] A summary of stream flow rates and energy consumption for the process illustrated in FIG. 1 is set forth in the following table:

Table I

(FIG. 1)

Stream Flow Summary - Lb. Moles/Hr [kg moles/Hr]

<u>Stream</u>	<u>Methane</u>	<u>Ethane</u>	<u>Propane</u>	<u>Butanes+</u>	<u>C. Dioxide</u>	<u>Total</u>
31	14,993	176	61	70	32,714	48,306
32	15,902	86	13	3	8,171	24,476
34	1,053	23	7	2	4,038	5,133
37	144	113	55	69	28,581	28,963
38	27	21	10	13	5,378	5,450
40	0	0	0	5	2	8
39	27	21	10	8	5,376	5,442
41	117	92	45	56	23,203	23,513
33	14,849	63	6	1	4,133	19,343
42	144	113	55	64	28,579	28,955

Recovery / Removal\*

Methane	99.04%	(recovered in the Residue Gas)
Carbon Dioxide	87.37%	(removed from the Residue Gas)

Carbon Dioxide Concentrations\*

Residue Gas	21.37%
Carbon Dioxide Product	98.70%

Power

Carbon Dioxide Compression	9,344	HP	[	15,361	kW]
Residue Gas Compression	6,563	HP	[	10,789	kW]
Refrigerant Compression	10,319	HP	[	16,964	kW]
Carbon Dioxide Pump	2,563	HP	[	4,214	kW]
Liquid Pump	12	HP	[	20	kW]
<b>Totals</b>	<b>28,801</b>	<b>HP</b>	<b>[</b>	<b>47,348</b>	<b>kW]</b>

\* (Based on un-rounded flow rates)

DESCRIPTION OF THE INVENTION

[0024] FIG. 2 illustrates a flow diagram of a process in accordance with the present invention. The feed gas composition and conditions considered in the process presented in FIG. 2 are the same as those in FIG. 1. Accordingly, the FIG. 2 process can be compared with that of the FIG. 1 process to illustrate the advantages of the present invention.

[0025] In the simulation of the FIG. 2 process, inlet gas enters the plant at 81°F [27°C] and 1110 psia [7,653 kPa(a)] as stream 31 and enters a heat and mass transfer means in the lower region of stripping section 113d inside processing assembly 113. This heat and

mass transfer means may be comprised of a fin and tube type heat exchanger, a plate type heat exchanger, a brazed aluminum type heat exchanger, or other type of heat transfer device, including multi-pass and/or multi-service heat exchangers. The heat and mass transfer means is configured to provide heat exchange between stream **31** flowing through one pass of the heat and mass transfer means and a distillation liquid stream flowing downward from a mass transfer means in the upper region of stripping section **113d** inside processing assembly **113**, so that stream **31** is cooled while heating the distillation liquid stream, cooling stream **31a** to 34°F [1°C] before it leaves the heat and mass transfer means. As the distillation liquid stream is heated, a portion of it is vaporized to form stripping vapors that rise upward as the remaining liquid continues flowing downward through the heat and mass transfer means. The heat and mass transfer means provides continuous contact between the stripping vapors and the distillation liquid stream so that it also functions to provide mass transfer between the vapor and liquid phases, stripping the liquid product stream **37** of methane and lighter components.

[0026] The partially cooled stream **31a** enters a heat exchange means in feed cooling section **113b** inside processing assembly **113**. This heat exchange means may also be comprised of a fin and tube type heat exchanger, a plate type heat exchanger, a brazed aluminum type heat exchanger, or other type of heat transfer device, including multi-pass and/or multi-service heat exchangers. The heat exchange means is configured to provide heat exchange between stream **31a** flowing through one pass of the heat exchange means, Freon R134a refrigerant streams, a residue gas stream arising from rectifying section **113c** inside processing assembly **113**, and cool carbon dioxide stream **38c** (which is described further below in paragraph [0028]). Stream **31a** is further cooled while heating these other streams, whereupon the further cooled stream **31b** at -14°F [-25°C] and 1076 psia [7,419 kPa(a)] is expanded to the operating pressure (approximately 618 psia [4,264 kPa(a)]) of stripping

section **113d** by expansion valve **12**, cooling stream **31c** to -35°F [-37°C] before it is supplied to processing assembly **113** above the mass transfer means in the upper region of stripping section **113d**.

[0027] Stripping section **113d** inside processing assembly **113** contains a mass transfer means consisting of a plurality of vertically spaced trays, one or more packed beds, or some combination of trays and packing. The trays and/or packing provide the necessary contact between the vapors rising upward and cold liquid falling downward. The liquid portion of expanded stream **31c** commingles with liquids falling downward from rectifying section **113c** and the combined liquid continues downward to the mass transfer means in the upper region of stripping section **113d**. The stripping vapors arising from the heat and mass transfer means in the lower region of stripping section **113d** rise upward through the mass transfer means in the upper region of stripping section **113d** to partially strip the combined liquid of methane and lighter components, with the remaining stripping vapors leaving the mass transfer means and combining with the vapor portion of expanded stream **31c** to form a distillation vapor stream that continues upward to rectifying section **113c**. The partially stripped distillation liquid leaving the mass transfer means in the upper region of stripping section **113d** continues downward into the heat and mass transfer means in the lower region of stripping section **113d** to continue the stripping of methane and lighter components from the liquid. The resulting liquid product (stream **37**) exits the lower region of stripping section **113d** and leaves processing assembly **113** at 45°F [7°C], based on reducing the methane concentration in the bottom product to 0.50% on a molar basis.

[0028] Column bottom product stream **37** is predominantly liquid carbon dioxide. A small portion (stream **38**) enters a heat exchange means in refrigerant subcooling section **113a** inside processing assembly **113**. This heat exchange means may likewise be comprised of a fin and tube type heat exchanger, a plate type heat exchanger, a brazed aluminum type

heat exchanger, or other type of heat transfer device, including multi-pass and/or multi-service heat exchangers. The heat exchange means is configured to provide heat exchange between stream **38** flowing through one pass of the heat exchange means and the residue gas stream from rectifying section **113c** inside processing assembly **113** that has been heated in the heat exchange means in feed cooling section **113b**, so that stream **38** is subcooled while further heating the residue gas stream. The resulting subcooled liquid (stream **38a**) at 0°F [-18°C] is expanded to lower pressure by expansion valve **20** and partially vaporized, further cooling stream **38b** to -64°F [-54°C] before it enters a heat and mass transfer means in rectifying section **113c** inside processing assembly **113**. This heat and mass transfer means may likewise be comprised of a fin and tube type heat exchanger, a plate type heat exchanger, a brazed aluminum type heat exchanger, or other type of heat transfer device, including multi-pass and/or multi-service heat exchangers. The heat and mass transfer means is configured to provide heat exchange between the distillation vapor stream arising from stripping section **113d** flowing upward through one pass of the heat and mass transfer means and the expanded stream **38b** flowing downward, so that the distillation vapor is cooled while heating the expanded stream. As the distillation vapor stream is cooled, a portion of it is condensed and falls downward while the remaining distillation vapor continues flowing upward through the heat and mass transfer means. The heat and mass transfer means provides continuous contact between the condensed liquid and the distillation vapor so that it also functions to provide mass transfer between the vapor and liquid phases, thereby providing rectification of the distillation vapor. The condensed liquid is collected from the bottom of the heat and mass transfer means, combined with the liquid portion of expanded stream **31c**, and directed to the mass transfer means in the upper region of stripping section **113d** as described previously. The residual liquid in stream **38b** functions as refrigerant in the heat and mass transfer means to provide the cooling and rectification of the

distillation vapor stream, with the resulting mostly vaporized carbon dioxide leaving the heat and mass transfer means in rectifying section **113c** at -48°F [-44°C] as stream **38c**.

[0029] The cool carbon dioxide from the heat and mass transfer means in rectifying section **113c** inside processing assembly **113** (stream **38c**) is heated to -10°F [-23°C] in the heat exchange means in feed cooling section **113b** inside processing assembly **113** by heat exchange with the feed gas as described previously. The warm carbon dioxide vapor (stream **38d**) at 76 psia [527 kPa(a)] is then compressed to high pressure in four stages by compressors **21**, **23**, **25**, and **27**, with cooling to 120°F [49°C] after each stage of compression by discharge coolers **22**, **24**, **26**, and **28**. The remaining portion (stream **41**) of column bottom product stream **37** is pumped to high pressure by pump **29** so that stream **41a** can combine with the dense phase fluid (stream **38l**) leaving discharge cooler **28**, forming high pressure carbon dioxide stream **42** which then flows to reinjection at 78°F [26°C] and 2015 psia [13,893 kPa(a)].

[0030] The vapor arising from rectifying section **113c** is the residue gas. It is warmed in feed cooling section **113b** as it provides cooling to the feed gas as described previously. The residue gas stream is further heated in refrigerant subcooling section **113a** as it provides cooling to stream **38** as described previously, whereupon residue gas stream **33** leaves processing assembly **113** at 20°F [-7°C]. The residue gas stream is then re-compressed by compressor **17** and cooled to 120°F [49°C] in discharge cooler **18** before it flows to treating at 1215 psia [8,377 kPa(a)].

[0031] A summary of stream flow rates and energy consumption for the process illustrated in FIG. 2 is set forth in the following table:

Table II

(FIG. 2)

Stream Flow Summary - Lb. Moles/Hr [kg moles/Hr]

<u>Stream</u>	<u>Methane</u>	<u>Ethane</u>	<u>Propane</u>	<u>Butanes+</u>	<u>C. Dioxide</u>	<u>Total</u>
31	14,993	176	61	70	32,714	48,306
37	144	109	55	70	28,580	28,959
38	24	19	9	12	4,887	4,951
41	120	90	46	58	23,693	24,008
33	14,849	67	6	0	4,134	19,347
42	144	109	55	70	28,580	28,959

Recovery / Removal\*

Methane	99.03%	(recovered in the Residue Gas)
Carbon Dioxide	87.36%	(removed from the Residue Gas)

Carbon Dioxide Concentrations\*

Residue Gas	21.37%
Carbon Dioxide Product	98.69%

Power

Carbon Dioxide Compression	8,449 HP	[ 13,890 kW]
Residue Gas Compression	6,119 HP	[ 10,060 kW]
Refrigerant Compression	11,498 HP	[ 18,902 kW]
Carbon Dioxide Pump	2,617 HP	[ 4,302 kW]
<hr/>		
Totals	28,683 HP	[ 47,154 kW]

\* (Based on un-rounded flow rates)

[0032] A comparison of Tables I and II shows that, compared to the prior art, the present invention maintains essentially the same methane recovery (99.03% versus 99.04% for the prior art), carbon dioxide removal (87.36% versus 87.37% for the prior art), carbon dioxide concentration in the residue gas (21.37% versus 21.37% for the prior art), and carbon dioxide purity (98.69% versus 98.70% for the prior art). However, further comparison of Tables I and II shows that the process performance was achieved using less power than the prior art. In terms of the total power consumption, the present invention represents a 0.4% improvement over the prior art of the FIG. 1 process.

[0033] The improvement in energy efficiency provided by the present invention over that of the prior art of the FIG. 1 process is primarily due to three factors. First, the compact arrangement of the heat exchange means in refrigerant subcooling section **113a** and feed cooling section **113b** inside processing assembly **113** eliminates the pressure drop imposed by the interconnecting piping found in conventional processing plants. The result is that the residue gas flowing to compressor **17** is at higher pressure for the present invention compared to the prior art, thereby reducing the power required by the present invention to restore the residue gas to the pressure needed for subsequent treating.

[0034] Second, using the heat and mass transfer means in the lower region of stripping section **113d** to simultaneously heat the distillation liquid leaving the mass transfer means in the upper region of stripping section **113d** while allowing the resulting vapors to contact the liquid and strip its volatile components is more efficient than using a conventional distillation column with external reboilers. The volatile components are stripped out of the liquid continuously, reducing the concentration of the volatile components in the stripping vapors more quickly and thereby improving the stripping efficiency for the present invention.

[0035] Third, using the heat and mass transfer means in rectifying section **113c** to simultaneously cool the distillation vapor stream arising from stripping section **113d** while condensing the carbon dioxide and the heavier hydrocarbon components from the distillation vapor stream provides more efficient rectification than using reflux in a conventional distillation column. As a result, less of the liquid carbon dioxide in bottom product stream **37** is needed to provide low level refrigeration to the process compared to the prior art of the FIG. 1 process. This allows more of the carbon dioxide to be pumped to high pressure rather than being compressed, reducing the power needed to deliver the carbon dioxide to reinjection (due to the inherent power advantage of pumping liquids compared to compressing gases).

[0036] The present invention offers two other advantages over the prior art in addition to the increase in processing efficiency. First, the compact arrangement of processing assembly **113** of the present invention replaces seven separate equipment items in the prior art (heat exchangers **10**, **11**, **14**, and **16**, separator **15**, pump **19**, and fractionation tower **13** in FIG. 1) with a single equipment item (processing assembly **113** in FIG. 2). This reduces the plot space requirements, eliminates the interconnecting piping, and eliminates the power consumed by the reflux pump, reducing the capital cost and operating cost of a process plant utilizing the present invention over that of the prior art. Second, elimination of the interconnecting piping means that a processing plant utilizing the present invention has far fewer flanged connections compared to the prior art, reducing the number of potential leak sources in the plant. Hydrocarbons are volatile organic compounds (VOCs), some of which are classified as greenhouse gases and some of which may be precursors to atmospheric ozone formation. Since carbon dioxide is also a greenhouse gas, the present invention significantly reduces the potential for atmospheric releases that can damage the environment.

#### Other Embodiments

[0037] As described earlier for the embodiment of the present invention shown in FIG. 2, feed stream **31** is totally condensed as it is cooled in the heat and mass transfer means in stripping section **113d** and the heat exchange means in feed cooling section **113b**, and the resulting liquid stream **31b** is then expanded by expansion valve **12** to the operating pressure of stripping section **113d** inside processing assembly **118**. However, the present invention is not limited to this embodiment. Depending on the quantity of heavier hydrocarbons in the feed gas and the feed gas pressure, the cooled feed stream **31b** expanded by expansion valve **12** in FIG. 2 may not contain any liquid (because it is above its dewpoint, or because it is above its cricondenbar). Some circumstances may favor partial condensation of the feed gas,

separating it into its respective vapor and liquid phases, and then expanding the vapor and liquid phases separately. Such an embodiment is shown in FIG. 3, where processing assembly **113** includes separator section **113e** to separate the partially condensed feed gas (stream **31b**) into vapor stream **43** and liquid stream **44**. (Separator section **113e** has an internal head or other means to divide it from stripping section **113d**, so that the two sections inside processing assembly **113** can operate at different pressures.) The vapor stream **43** enters a work expansion machine **50** in which mechanical energy is extracted from this portion of the high pressure feed as the vapor is expanded substantially isentropically to the operating pressure of rectifying section **113c**, whereupon the expanded stream **43a** is supplied to processing assembly **113** below the heat and mass transfer means in rectifying section **113c**. The work recovered is often used to drive a centrifugal compressor (such as item **51**) that can be used to re-compress the residue gas stream (stream **33**), for example. Liquid stream **44** is expanded to the operating pressure of stripping section **113d** by expansion valve **12** before the expanded stream **44a** is supplied to processing assembly **113** above the mass transfer means in the upper region of stripping section **113d**. In some cases, it may be advantageous to include a mass transfer means in rectifying section **113c** below the point at which expanded stream **43a** enters rectifying section **113c**.

[0038] In some circumstances, it may be advantageous to use an external separator vessel to separate cooled feed stream **31b**, rather than including separator section **113e** in processing assembly **113**. As shown in FIG. 4, separator **52** can be used to separate cooled feed stream **31b** into vapor stream **43** and liquid stream **44**.

[0039] Feed gas conditions, plant size, available equipment, or other factors may indicate that elimination of work expansion machine **50**, or replacement with an alternate expansion device (such as an expansion valve), is feasible in the FIGS. 3 and 4 embodiments. Although individual stream expansion is depicted in particular expansion devices, alternative

expansion means may be employed where appropriate. For example, conditions may warrant work expansion of cooled feed stream **31b** in FIG. 2 or liquid stream **44** in FIGS. 3 and 4.

[0040] In accordance with the present invention, the use of external refrigeration to supplement the cooling available to the inlet gas from the distillation vapor and liquid streams may be employed. In such cases, the heat exchange means in feed cooling section **113b** may include one or more passes for the refrigerant as shown by the dashed lines in FIGS. 2 through 4. Alternatively, conventional gas chiller(s) could be used to cool stream **31a** with refrigerant before stream **31a** enters the heat exchange means in feed cooling section **113b** and/or cool stream **31b** with refrigerant before stream **31b** enters expansion valve **12** (FIG. 2), separator section **113e** (FIG. 3) or separator **52** (FIG. 4).

[0041] Depending on the temperature and richness of the feed gas and the amount of methane allowed in liquid product stream **37**, there may not be sufficient heating available from feed stream **31** to cause the liquid leaving stripping section **113d** to meet the product specifications. In such cases, the heat and mass transfer means in stripping section **113d** may include provisions for providing supplemental heating with heating medium as shown by the dashed lines in FIGS. 2 through 4. Alternatively, another heat and mass transfer means can be included in the lower region of stripping section **113d** for providing supplemental heating, or stream **31** can be heated with heating medium before it is supplied to the heat and mass transfer means in stripping section **113d**.

[0042] Depending on the type of heat transfer devices selected for the heat exchange means in refrigerant subcooling section **113a** and feed cooling section **113b**, it may be possible to combine these heat exchange means in a single multi-pass and/or multi-service heat transfer device. In such cases, the multi-pass and/or multi-service heat transfer device will include appropriate means for distributing, segregating, and collecting stream **31a**, stream **38**, stream **38c**, and stream **33** in order to accomplish the desired cooling and heating.

Likewise, the type of heat and mass transfer device selected for the heat and mass transfer means in rectifying section 113c may allow combining it with the heat exchange means in feed cooling section 113b (and possibly with the heat exchange means in refrigerant subcooling section 113a as well) in a single multi-pass and/or multi-service heat and mass transfer device. In such cases, the multi-pass and/or multi-service heat and mass transfer device will include appropriate means for distributing, segregating, and collecting stream 31a, stream 38, stream 38b, stream 38c, and stream 33 in order to accomplish the desired cooling and heating.

[0043] In some circumstances, the portion (stream 38) of column bottom product stream 37 that is used to provide refrigeration in FIGS. 2 through 4 may not need to be restored to high pressure after it has been heated (stream 38d). In such cases, the compression and cooling shown (compressors 21, 23, 25, and 27 and discharge coolers 22, 24, 26, and 28) may not be needed, and only stream 41a flows to stream 42.

[0044] The present invention provides improved separation of carbon dioxide from hydrocarbon gas streams per amount of utility consumption required to operate the process. An improvement in utility consumption required for operating the process may appear in the form of reduced power requirements for compression or re-compression, reduced power requirements for pumping, reduced power requirements for external refrigeration, reduced energy requirements for supplemental heating, reduced energy requirements for tower reboiling, or a combination thereof.

[0045] While there have been described what are believed to be preferred embodiments of the invention, those skilled in the art will recognize that other and further modifications may be made thereto, e.g. to adapt the invention to various conditions, types of

feed, or other requirements without departing from the spirit of the present invention as defined by the following claims.

WE CLAIM:

1. A process for the separation of a gas stream containing at least methane and carbon dioxide into a volatile residue gas fraction containing a major portion of said methane and a relatively less volatile fraction containing a major portion of said carbon dioxide wherein

- (1) said gas stream is cooled, thereby forming a cooled stream;
- (2) said cooled stream is expanded to intermediate pressure

whereby it is further cooled;

(3) said expanded cooled stream is supplied as a top feed to a mass transfer means housed in a processing assembly, said mass transfer means having a lower region and an upper region;

(4) a distillation liquid stream is collected from the lower region of said mass transfer means and heated in a first heat and mass transfer means housed in said processing assembly, thereby to supply at least a portion of the cooling of step (1) while simultaneously stripping the more volatile components from said distillation liquid stream, and thereafter discharging said heated and stripped distillation liquid stream from said processing assembly as said relatively less volatile fraction;

(5) a first distillation vapor stream is collected from the upper region of said mass transfer means and combined with any vapor portion of said expanded cooled stream to form a second distillation vapor stream;

(6) said second distillation vapor stream is cooled in a second heat and mass transfer means housed in said processing assembly, thereby condensing the less volatile components from said second distillation vapor stream and forming a condensed stream and a third distillation vapor stream;

- (7) at least a portion of said condensed stream is supplied as another top feed to said mass transfer means;
- (8) said relatively less volatile fraction is divided into at least first and second streams;
- (9) said second stream is cooled and thereafter expanded to lower pressure whereby it is further cooled;
- (10) said expanded cooled second stream is heated, thereby to supply at least a portion of the cooling of steps (1) and (6);
- (11) said third distillation vapor stream is heated in one or more heat exchange means housed in said processing assembly, thereby to supply at least a portion of the cooling of steps (1) and (9), and thereafter discharging said heated third distillation vapor stream from said processing assembly as said volatile residue gas fraction; and
- (12) the quantities and temperatures of said feed streams to said mass transfer means are effective to maintain the temperature of said upper region of said mass transfer means at a temperature whereby the major portion of said carbon dioxide is recovered in said relatively less volatile fraction.

2. A process for the separation of a gas stream containing at least methane and carbon dioxide into a volatile residue gas fraction containing a major portion of said methane and a relatively less volatile fraction containing a major portion of said carbon dioxide wherein
  - (1) said gas stream is cooled sufficiently to partially condense it;
  - (2) said partially condensed gas stream is supplied to a separating means and is separated therein to provide a vapor stream and at least one liquid stream;
  - (3) said at least one liquid stream is expanded to intermediate pressure;

(4) said expanded liquid stream is supplied as a top feed to a mass transfer means housed in a processing assembly, said mass transfer means having a lower region and an upper region;

(5) said vapor stream is expanded to said intermediate pressure and is supplied as a feed to said processing assembly above said mass transfer means;

(6) a distillation liquid stream is collected from the lower region of said mass transfer means and heated in a first heat and mass transfer means housed in said processing assembly, thereby to supply at least a portion of the cooling of step (1) while simultaneously stripping the more volatile components from said distillation liquid stream, and thereafter discharging said heated and stripped distillation liquid stream from said processing assembly as said relatively less volatile fraction;

(7) a first distillation vapor stream is collected from the upper region of said mass transfer means and combined with the vapor portion of said expanded vapor stream to form a second distillation vapor stream;

(8) said second distillation vapor stream is cooled in a second heat and mass transfer means housed in said processing assembly, thereby condensing the less volatile components from said second distillation vapor stream and forming a condensed stream and a third distillation vapor stream;

(9) at least a portion of said condensed stream is supplied as another top feed to said mass transfer means;

(10) said relatively less volatile fraction is divided into at least first and second streams;

(11) said second stream is cooled and thereafter expanded to lower pressure whereby it is further cooled;

(12) said expanded cooled second stream is heated, thereby to supply at least a portion of the cooling of steps (1) and (8);

(13) said third distillation vapor stream is heated in one or more heat exchange means housed in said processing assembly, thereby to supply at least a portion of the cooling of steps (1) and (11), and thereafter discharging said heated third distillation vapor stream from said processing assembly as said volatile residue gas fraction; and

(14) the quantities and temperatures of said feed streams to said mass transfer means are effective to maintain the temperature of said upper region of said mass transfer means at a temperature whereby the major portion of said carbon dioxide is recovered in said relatively less volatile fraction.

3. The process according to claim 2 wherein said separating means is housed in said processing assembly.

4. The process according to claim 2 or 3 wherein

(1) an additional mass transfer means is housed in said processing assembly below said second heat and mass transfer means, said additional mass transfer means having a lower region and an upper region;

(2) said expanded vapor stream is supplied as a top feed to said additional mass transfer means;

(3) said first distillation vapor stream is directed to said additional mass transfer means as a bottom feed thereto;

(4) a fourth distillation vapor stream is collected from the upper region of said additional mass transfer means and combined with said vapor portion of said expanded vapor stream to form said second distillation vapor stream;

(5) at least a portion of said condensed stream is supplied as another top feed to said additional mass transfer means; and

(6) the quantities and temperatures of said feed streams to said additional mass transfer means are effective to maintain the temperature of said upper region of said additional mass transfer means at a temperature whereby the major portion of said carbon dioxide is recovered in said relatively less volatile fraction.

5. The process according to claim 1, 2, or 3 wherein said first heat and mass transfer means includes one or more passes for an external heating medium to supplement the heating supplied by said gas stream for said stripping of said more volatile components from said distillation liquid stream.

6. The process according to claim 4 wherein said first heat and mass transfer means includes one or more passes for an external heating medium to supplement the heating supplied by said gas stream for said stripping of said more volatile components from said distillation liquid stream.

7. An apparatus for the separation of a gas stream containing at least methane and carbon dioxide into a volatile residue gas fraction containing a major portion of said methane and a relatively less volatile fraction containing a major portion of said carbon dioxide comprising

(1) first heat and mass transfer means housed in a processing assembly to cool said gas stream;

(2) first heat exchange means housed in said processing assembly and connected to said first heat and mass transfer means to receive said cooled gas stream and further cool it, thereby forming a cooled stream;

(3) first expansion means connected to said first heat exchange means to receive said cooled stream and expand it to intermediate pressure;

(4) mass transfer means housed in said processing assembly and connected to said first expansion means to receive said expanded cooled stream as a top feed thereto, said mass transfer means having a lower region and an upper region;

(5) liquid collecting means housed in said processing assembly and connected to said mass transfer means to receive a distillation liquid stream from the lower region of said mass transfer means;

(6) said first heat and mass transfer means being further connected to said liquid collecting means to receive said distillation liquid stream and heat it, thereby to supply at least a portion of the cooling of step (1) while simultaneously stripping the more volatile components from said distillation liquid stream, and thereafter discharging said heated and stripped distillation liquid stream from said processing assembly as said relatively less volatile fraction;

(7) vapor collecting means housed in said processing assembly and connected to said mass transfer means to receive a first distillation vapor stream from the upper region of said mass transfer means;

(8) combining means connected to said vapor collecting means to receive said first distillation vapor stream and combine it with any vapor portion of said expanded cooled stream and form thereby a second distillation vapor stream;

(9) second heat and mass transfer means housed in said processing assembly and connected to said combining means to receive said second distillation vapor stream and cool it, thereby forming a condensed stream and a third distillation vapor stream;

(10) said mass transfer means being further connected to said second heat and mass transfer means to receive at least a portion of said condensed stream as another top feed thereto;

(11) dividing means connected to said processing assembly to receive said relatively less volatile fraction and divide it into at least first and second streams;

(12) second heat exchange means housed in said processing assembly and connected to said dividing means to receive said second stream and cool it;

(13) second expansion means connected to said second heat exchange means to receive said cooled second stream and expand it to lower pressure;

(14) said second heat and mass transfer means being further connected to said second expansion means to receive said expanded cooled second stream and heat it, thereby to supply at least a portion of the cooling of step (9);

(15) said first heat exchange means being further connected to said second heat and mass transfer means to receive said heated expanded second stream and further heat it, thereby to supply at least a portion of the cooling of step (2);

(16) said first heat exchange means being further connected to said second heat and mass transfer means to receive said third distillation vapor stream and heat it, thereby to supply at least a portion of the cooling of step (2);

(17) said second heat exchange means being further connected to said first heat exchange means to receive said heated third distillation vapor stream and further heat it, thereby to supply at least a portion of the cooling of step (12), and thereafter discharging said further heated third distillation vapor stream from said processing assembly as said volatile residue gas fraction; and

(18) control means adapted to regulate the quantities and temperatures of said feed streams to said upper region of said mass transfer means to maintain the temperature of said upper region of said mass transfer means at a temperature whereby the major portion of said carbon dioxide is recovered in said relatively less volatile fraction.

8. An apparatus for the separation of a gas stream containing at least methane and carbon dioxide into a volatile residue gas fraction containing a major portion of said methane and a relatively less volatile fraction containing a major portion of said carbon dioxide comprising

- (1) first heat and mass transfer means housed in a processing assembly to cool said gas stream;
- (2) first heat exchange means housed in said processing assembly and connected to said first heat and mass transfer means to receive said cooled gas stream and further cool it, thereby forming a partially condensed gas stream;
- (3) separating means connected to said first heat exchange means to receive said partially condensed gas stream and separate it into a vapor stream and at least one liquid stream;
- (4) first expansion means connected to said separating means to receive said at least one liquid stream and expand it to intermediate pressure;
- (5) mass transfer means housed in said processing assembly and connected to said first expansion means to receive said expanded liquid stream as a top feed thereto, said mass transfer means having a lower region and an upper region;
- (6) second expansion means connected to said separating means to receive said vapor stream and expand it to said intermediate pressure, said second expansion means being further connected to said processing assembly to supply said expanded vapor stream as a feed thereto above said mass transfer means;
- (7) liquid collecting means housed in said processing assembly and connected to said mass transfer means to receive a distillation liquid stream from the lower region of said mass transfer means;

(8) said first heat and mass transfer means being further connected to said liquid collecting means to receive said distillation liquid stream and heat it, thereby to supply at least a portion of the cooling of step (1) while simultaneously stripping the more volatile components from said distillation liquid stream, and thereafter discharging said heated and stripped distillation liquid stream from said processing assembly as said relatively less volatile fraction;

(9) vapor collecting means housed in said processing assembly and connected to said mass transfer means to receive a first distillation vapor stream from the upper region of said mass transfer means;

(10) combining means connected to said vapor collecting means to receive said first distillation vapor stream and combine it with the vapor portion of said expanded vapor stream and form thereby a second distillation vapor stream;

(11) second heat and mass transfer means housed in said processing assembly and connected to said combining means to receive said second distillation vapor stream and cool it, thereby forming a condensed stream and a third distillation vapor stream;

(12) said mass transfer means being further connected to said second heat and mass transfer means to receive at least a portion of said condensed stream as another top feed thereto;

(13) dividing means connected to said processing assembly to receive said relatively less volatile fraction and divide it into at least first and second streams;

(14) second heat exchange means housed in said processing assembly and connected to said dividing means to receive said second stream and cool it;

(15) third expansion means connected to said second heat exchange means to receive said cooled second stream and expand it to lower pressure;

(16) said second heat and mass transfer means being further connected to said third expansion means to receive said expanded cooled second stream and heat it, thereby to supply at least a portion of the cooling of step (11);

(17) said first heat exchange means being further connected to said second heat and mass transfer means to receive said heated expanded second stream and further heat it, thereby to supply at least a portion of the cooling of step (2);

(18) said first heat exchange means being further connected to said second heat and mass transfer means to receive said third distillation vapor stream and heat it, thereby to supply at least a portion of the cooling of step (2);

(19) said second heat exchange means being further connected to said first heat exchange means to receive said heated third distillation vapor stream and further heat it, thereby to supply at least a portion of the cooling of step (14), and thereafter discharging said further heated third distillation vapor stream from said processing assembly as said volatile residue gas fraction; and

(20) control means adapted to regulate the quantities and temperatures of said feed streams to said upper region of said mass transfer means to maintain the temperature of said upper region of said mass transfer means at a temperature whereby the major portion of said carbon dioxide is recovered in said relatively less volatile fraction.

9. The apparatus according to claim 8 wherein said separating means is housed in said processing assembly.

10. The apparatus according to claim 8 or 9 wherein

(1) an additional mass transfer means is housed in said processing assembly below said second heat and mass transfer means, said additional mass transfer means having a lower region and an upper region;

(2) said additional mass transfer means is connected to said second expansion means to receive said expanded vapor stream as a top feed thereto;

(3) said additional mass transfer means is further connected to said vapor collecting means to receive said first distillation vapor stream as a bottom feed thereto;

(4) an additional vapor collecting means is housed in said processing assembly and connected to said additional mass transfer means to receive a fourth distillation vapor stream from the upper region of said additional mass transfer means;

(5) said combining means is adapted to be connected to said additional vapor collecting means to receive said fourth distillation vapor stream and combine it with said vapor portion of said expanded vapor stream, thereby forming said second distillation vapor stream;

(6) said additional mass transfer means is further connected to said second heat and mass transfer means to receive at least a portion of said condensed stream as another top feed thereto; and

(7) said control means is adapted to regulate the quantities and temperatures of said feed streams to said upper region of said additional mass transfer means to maintain the temperature of said upper region of said additional mass transfer means at a temperature whereby the major portion of said carbon dioxide is recovered in said relatively less volatile fraction.

11. The apparatus according to claim 7, 8, or 9 wherein said first heat and mass transfer means includes one or more passes for an external heating medium to supplement the heating supplied by said gas stream for said stripping of said more volatile components from said distillation liquid stream.

12. The apparatus according to claim 10 wherein said first heat and mass transfer means includes one or more passes for an external heating medium to supplement the

heating supplied by said gas stream for said stripping of said more volatile components from said distillation liquid stream.

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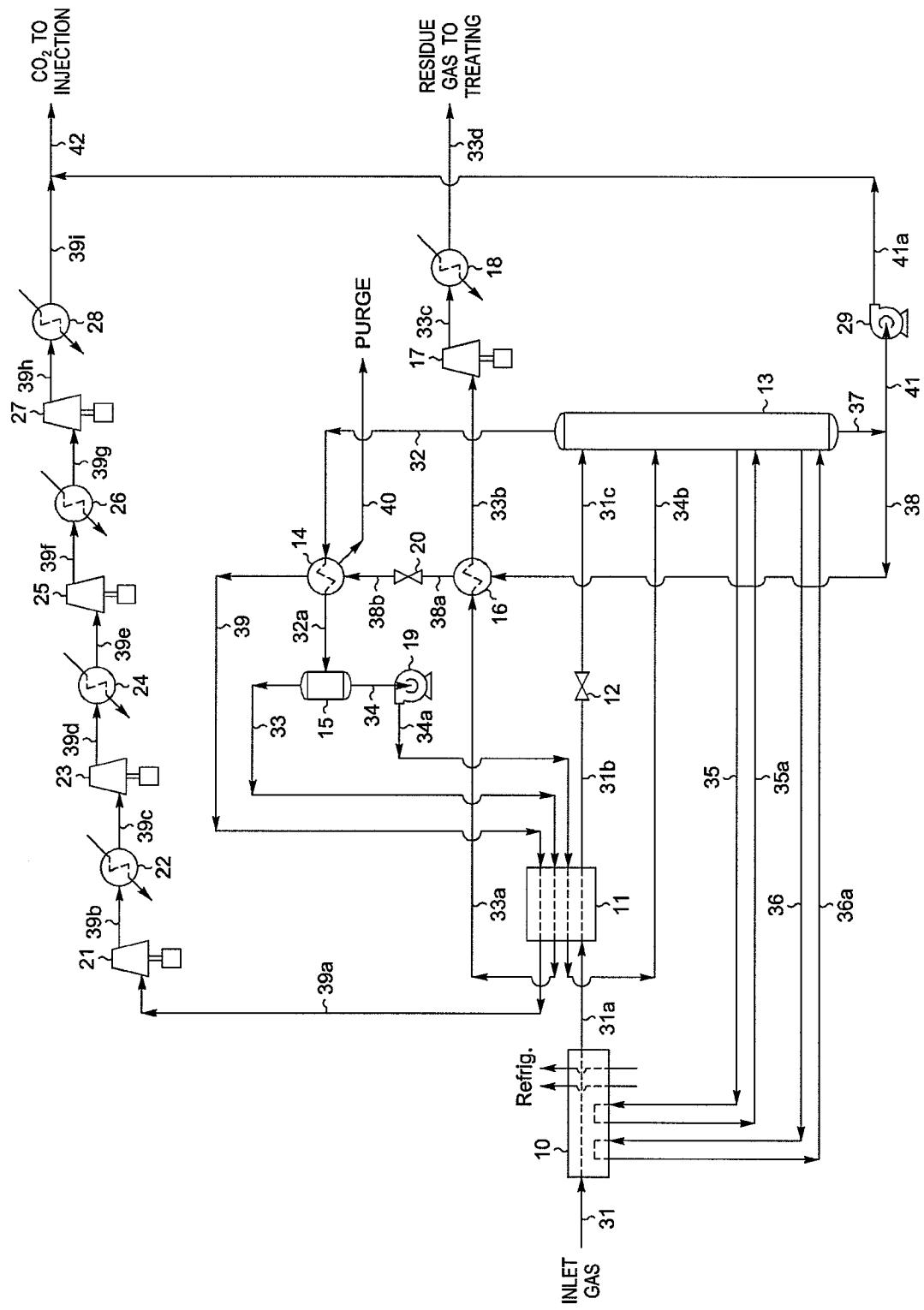


FIG. 1  
(PRIOR ART)

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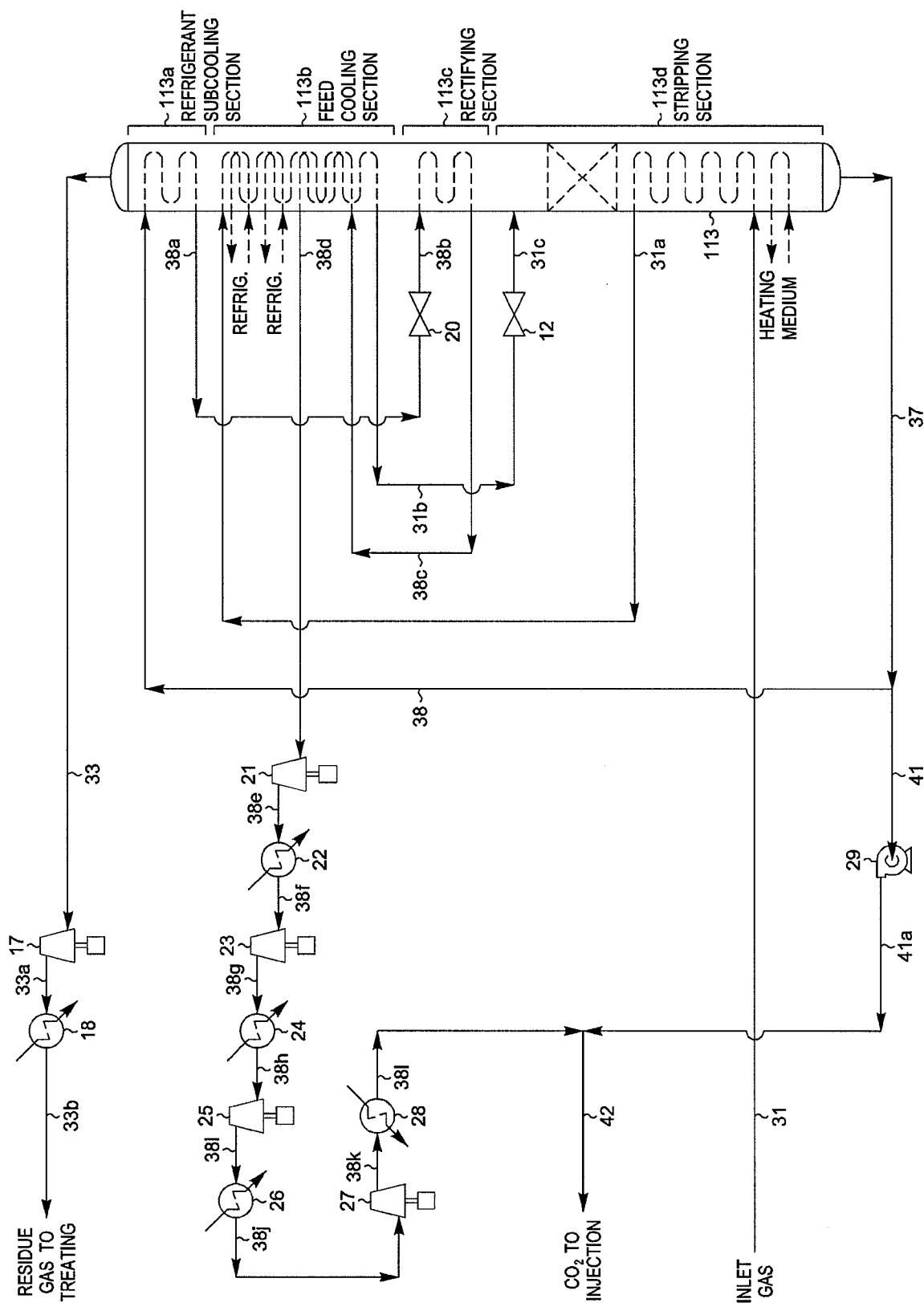


FIG. 2

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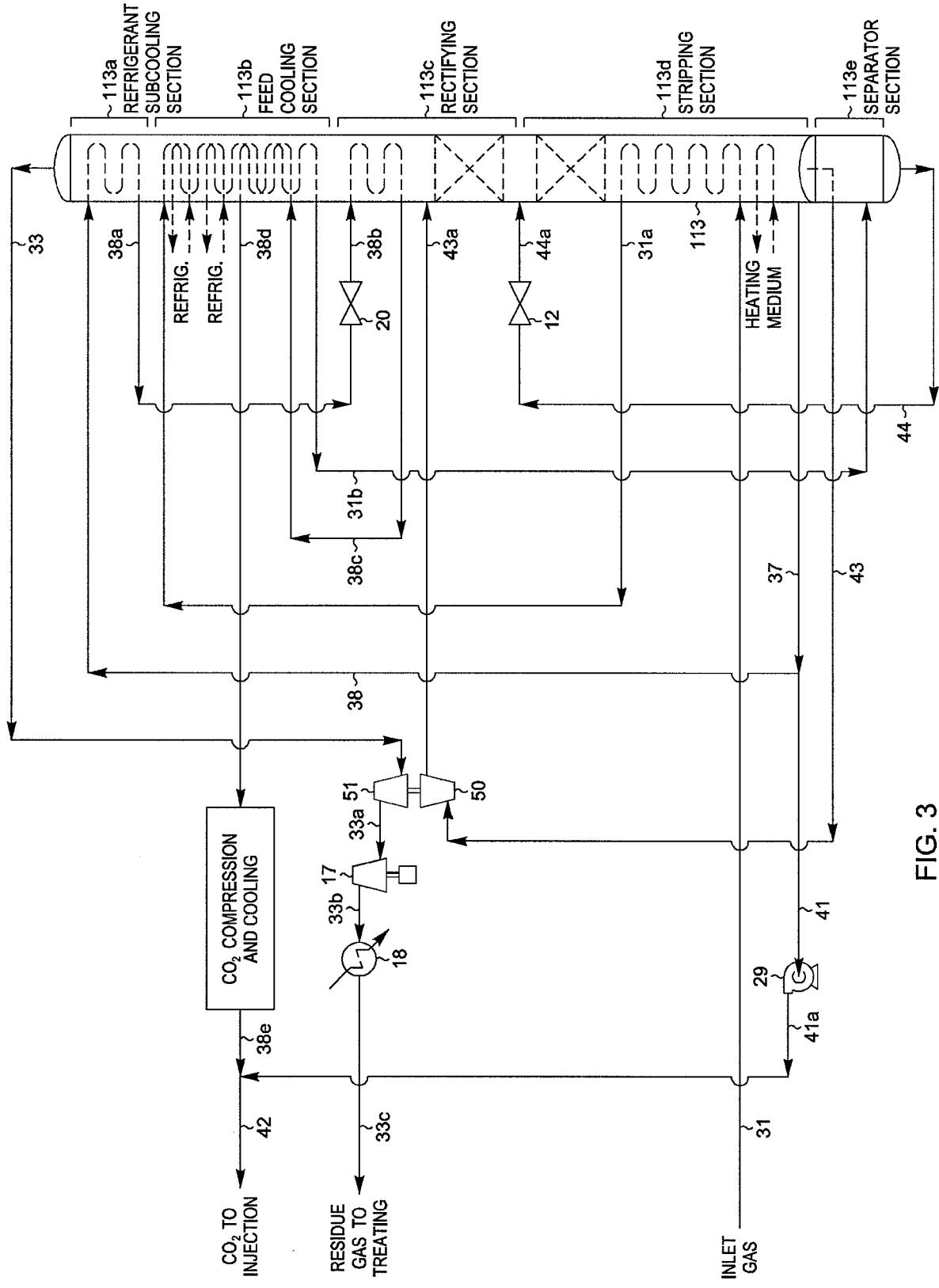


FIG. 3

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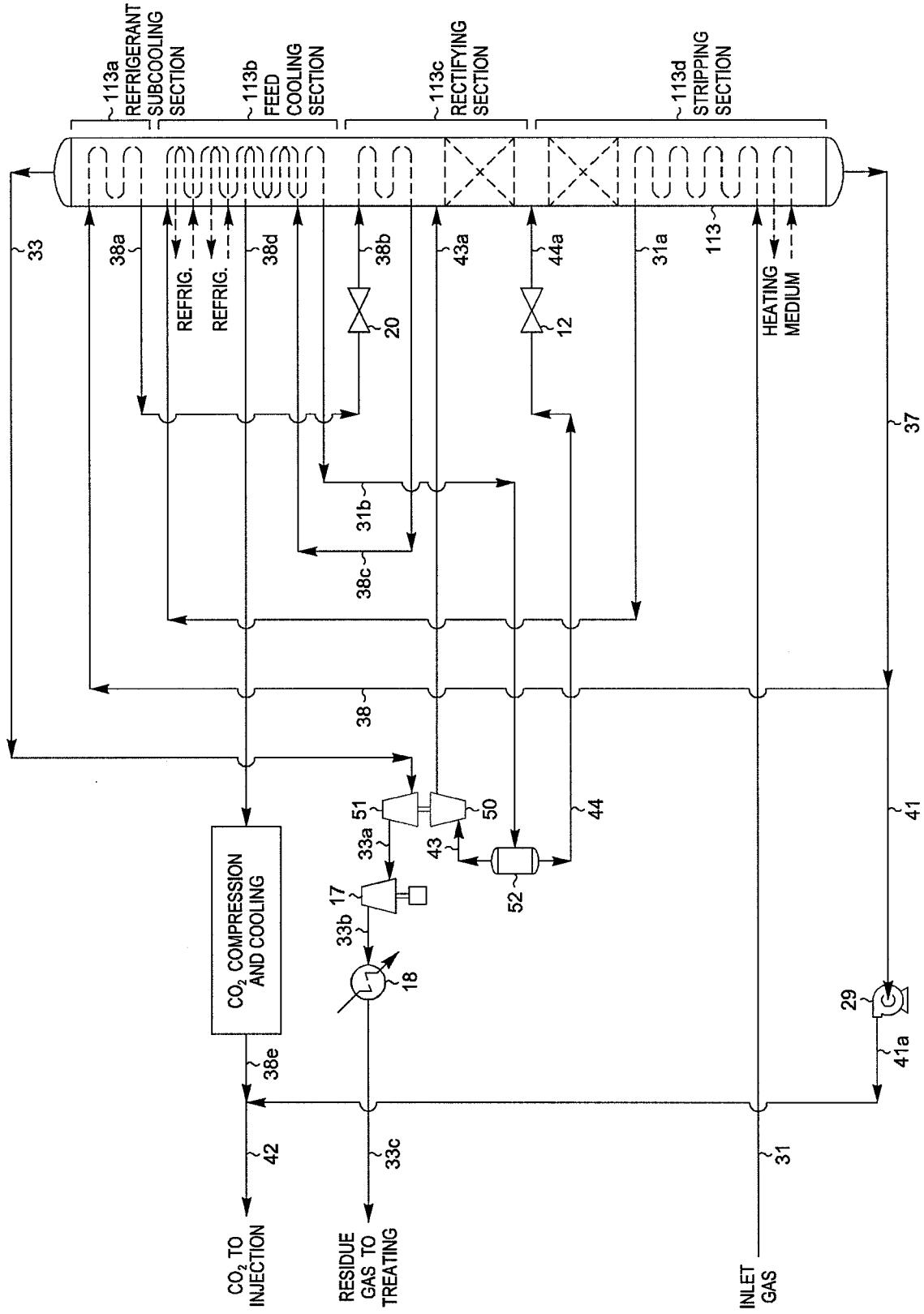


FIG. 4

**INTERNATIONAL SEARCH REPORT**

International application No.

PCT/US 10/37098

**A. CLASSIFICATION OF SUBJECT MATTER**

IPC(8) - F25J 3/00 (2010.01)  
USPC - 423/220; 437.1; 62/620

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 3/00 (2010.01)

USPC: 423/220; 437.1; 62/620

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched  
USPC: 423/220; 437.1; 62/620 (keyword limited; terms below)

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

PubWest(PGPB,USPT,USOC,EPAB,JPAB); esp@cenet; Google Scholar; Google Patents

Search Terms used: bottom\$1, carbon, combin\$3, condens\$3, control\$4, CO2, dioxide\$1, distill\$5, divid\$3, exchang\$3, expand\$5, fluid\$1, gas\$2, heat\$3, liquid\$1, methane, remov\$3, rich, second, separa\$3, split\$4, stream\$1, strip\$4, supplement\$3, vapor\$1, volatile\$1

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US 5,335,504 A (DURR et al.) 09 August 1994 (09.08.1994), entire document, especially FIG. 2, FIG. 7, col. 2, ln. 64-col. 3, ln. 3, col. 6, ln. 5-13, col. 6, ln. 31-38	1-12
Y	US 2005/0268649 A1 (WILKINSON et al.) 08 December 2005 (08.12.2005), para [0002], [0009], [0035], [0077]	1-12
A	US 3,983,711 A (SOLOMON) 05 October 1976 (05.10.1976), FIG. 2, col. 5, ln. 19-col. 6, ln. 24	1-12

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	Date of mailing of the international search report
20 July 2010 (20.07.2010)	<b>17 AUG 2010</b>
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:  Lee W. Young  PCT Helpdesk: 571-272-4300 PCT OSP: 571-272-7774