

Claims

1. An apparatus for separating gases, comprising membrane separation stages (1) to (3), at least one compressor (4) and/or at least one, preferably one or two, vacuum pump(s) (15), the feed stream separation stage (1) separating a feed stream (5) consisting of at least two components into a first permeate stream (6) and a first retentate stream (7), the retentate separation stage (2) dividing the first retentate stream (7) into a second permeate stream (9) which is supplied to the feed stream (5), and a second retentate stream (8) which is withdrawn as a product, and the permeate separation stage (3) dividing the first permeate stream (6) into a third retentate stream (10) which is supplied to the feed stream (5), and a third permeate stream (11) which is withdrawn as a product, characterized in that
- the first permeate stream (6) is not subjected to any recompression,
 - at least in the feed stream separation stage (1), but preferably in all three membrane separation stages (1) to (3), gas separation membrane modules having a mixed gas selectivity of at least 30 are used,
 - the apparatus is configured such that the gas volume recycled in the second permeate stream (9) and in the third retentate stream (10) totals less than 60% by volume of the volume of the crude gas stream (17),
 - the concentration of at least one permeate gas of the feed stream separation stage (1), after recycling of the second permeate stream (9) and of the third retentate stream (10), is increased in the feed stream (5), preferably increased by at least 2%, more preferably by at least 3% and more preferably by 3 to 40%, in each case

compared to the concentration in the crude gas stream (17).

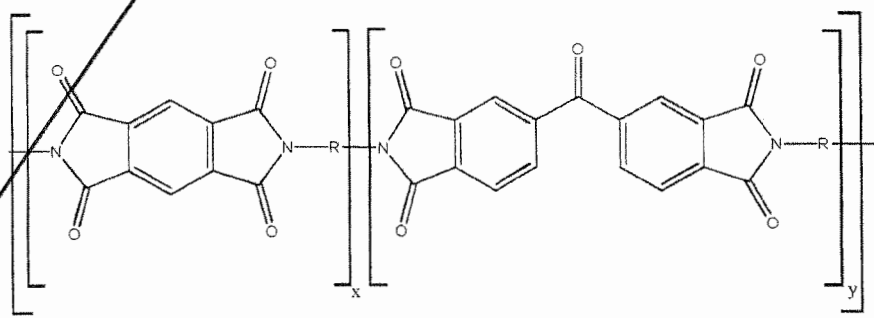
2. The apparatus as claimed in claim 1,
5 characterized in that
the second permeate stream (9) and the third retentate stream (10) are passed along the suction side of the compressor (4) for reprocessing.
- 10 3. The apparatus as claimed in claim 1 or 2,
characterized in that
a multistage compressor (4) is used.
- 15 4. The apparatus as claimed in claim 3,
characterized in that
the second permeate stream (9) and/or the third retentate stream (10) is/are introduced into the compressor (4) between two compression stages.
- 20 5. The apparatus as claimed in any of claims 1 to 4,
characterized in that
the first retentate stream (7) and/or the second retentate stream (8) and/or the third retentate stream (10) is/are passed through a pressure-
25 reducing valve.
6. The apparatus as claimed in any of claims 1 to 5,
characterized in that
a gas separation membrane module having a mixed
30 gas selectivity of at least 35, preferably at least 40 and more preferably at least 45 is used
in at least one of the membrane separation stages
(1) to (3).
- 35 7. The apparatus as claimed in any of claims 1 to 6,
characterized in that
at least one of the membrane separation stages (1)
to (3) comprises more than one gas separation

membrane module, which are connected in parallel and/or in series.

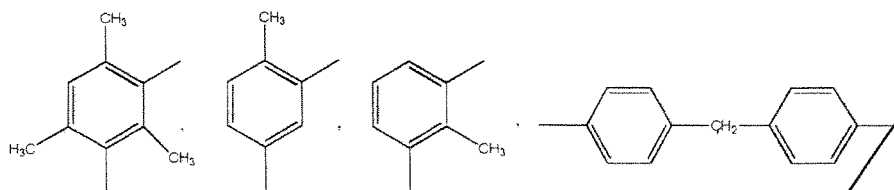
8. The apparatus as claimed in any of claims 1 to 7,
5 characterized in that
the gas separation membrane module(s) consist(s)
of hollow fiber membranes and/or flat membranes.

9. The apparatus as claimed in any of claims 1 to 8,
10 characterized in that
the materials used for the separation-active layer
of the membranes are amorphous or semicrystalline
polymers, for example but not exclusively
polyimides, polyamides, polysulfones, cellulose
15 acetates and derivatives, polyphenylene oxides,
polysiloxanes, polymers with intrinsic
microporosity, mixed matrix membranes, facilitated
transport membranes, polyethylene oxides,
polypropylene oxides or mixtures thereof.

20 10. The apparatus as claimed in claim 9,
characterized in that
the material used for the separation-active layer
of the membranes is a polyimide of the general
25 formula



30 R is selected from the group consisting of



x,y: mol fraction where $0 < x < 0.5$ and $1 > y > 0.5$.

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11. The apparatus as claimed in any of claims 1 to 10, characterized in that at least 95%, preferably at least 97%, more preferably at least 99% and most preferably at least 99.5% of the retentate component of the feed stream separation stage (1) which has been introduced into the apparatus with the crude gas stream (17) is discharged via the second retentate stream (8).

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12. The apparatus as claimed in any of claims 1 to 11, characterized in that not more than 5%, preferably not more than 3%, more preferably not more than 1% and most preferably not more than 0.5% of the retentate component of the feed stream separation stage (1) introduced into the apparatus with the crude gas stream (17) is discharged via the third permeate stream (11).

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13. A process for separating gas mixtures, characterized in that an apparatus according to any of claims 1 to 12 is used.

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14. The process as claimed in claim 13, characterized in that the gas mixture used is a mixture of predominantly but not exclusively carbon dioxide and methane or predominantly but not exclusively hydrogen and

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methane or predominantly but not exclusively carbon monoxide and hydrogen or crude biogas or crude natural gas.

- 5 15. The process as claimed in either of claims 13 and 14,
characterized in that
the pressure on the permeate side (6) of the feed
stream separation stage (1) is adjusted,
10 preferably by means of a pressure-reducing valve
(14) on the retentate side of the permeate
separation stage (3), to 1 to 30 bar, preferably
to 2 to 20 bar and more preferably to 3 and
10 bar.
- 15 16. The process as claimed in any of claims 13 to 15,
characterized in that
the pressure of the first and second retentate
stream (7) and (8) is adjusted, preferably by
20 means of a pressure-reducing valve (13) in the
second retentate stream (8), to 1 to 100 bar,
preferably to 5 to 80 bar and more preferably to
10 to 70 bar.
- 25 17. The process as claimed in any of claims 13 to 16,
characterized in that
the driving force used for the separation task is
a partial pressure differential between the
retentate side and the permeate side in the
30 respective membrane separation stages, the partial
pressure differential being generated by a
compressor in the feed stream (5) and/or by a
vacuum pump (15) in the second and/or third
permeate stream (9) and/or (11) and/or by a
35 permeate-side purge gas stream.
18. The process as claimed in claim 16 or 17,
characterized in that

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the pressure of the permeate of the feed stream separation stage (1) is the same or increased relative to the ambient pressure, such that a partial pressure differential still exists between retentate and permeate of the permeate separation stage (3), and hence there is a driving force in the case that the permeate of the permeate separation stage (3) is at ambient pressure or reduced pressure is applied.

CURRENT CLAIMS

Claim 1: An apparatus for separating gases, comprising: membrane separation stages;
and,

a compressor, a vacuum pump, or both,

wherein the membrane separation stages comprise a feed stream separation stage, a retentate separation stage, and a permeate separation stage:

the feed stream separation stage is configured to separate a feed stream comprising at least two components into a first permeate stream and a first retentate stream,

the retentate separation stage is configured to divide the first retentate stream into a second permeate stream and a second retentate stream,

wherein

the second permeate stream is supplied to the feed stream, and the second retentate stream is withdrawn as a product, and

the permeate separation stage is configured to divide the first permeate stream into a third retentate stream and a third permeate stream,

wherein

the third retentate stream is supplied to the feed stream, and the third permeate stream is withdrawn as a product,

the apparatus is configured not to subject the first permeate stream to any recompression,

the feed stream separation stage comprises a gas separation membrane module having a mixed gas selectivity of at least 30,

the apparatus is configured such that a gas volume recycled in the second permeate stream and in the third retentate stream totals less than 60% by volume of a crude gas stream,

the apparatus is configured to increase a concentration of at least one permeate gas of the feed stream separation stage, after recycling of the second permeate stream and of the third retentate stream, in the feed stream, compared to a concentration in the crude gas stream.

Claim 2: The apparatus of claim 1,

wherein

the apparatus is configured to pass the second permeate stream and the third retentate stream along a suction side of the compressor.

Claim 3: The apparatus of claim 1, comprising:

a multistage compressor as a compressor.

Claim 4: The apparatus of claim 3, wherein the apparatus is configured to introduce the second permeate stream, the third retentate stream, or both into the compressor between two compression stages.

Claim 5: The apparatus of claim 1, wherein the apparatus is configured to pass at least one selected from the group consisting of the first retentate stream, the second retentate stream, and the third retentate stream through a pressure-reducing valve.

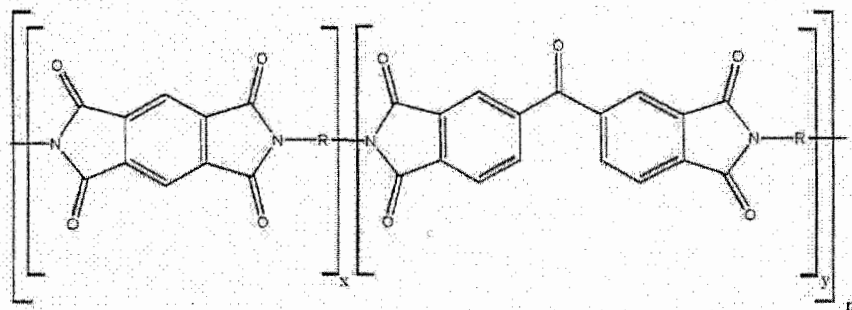
Claim 6: The apparatus of claim 1, wherein the gas separation membrane module in at least one of the membrane separation stages has a mixed gas selectivity of at least 35.

Claim 7: The apparatus of claim 1, wherein at least one of the membrane separation stages comprises more than one gas separation membrane module, wherein the modules are connected in parallel, in series, or in parallel and in series.

Claim 8: The apparatus of claim 1, wherein at least one of the membrane separation stages comprises a gas separation membrane module comprising a hollow fiber membrane, a flat membrane, or both.

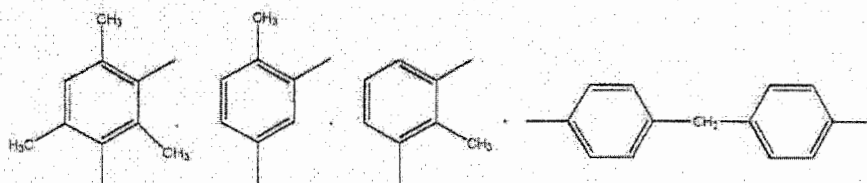
Claim 9: The apparatus of claim 1, wherein a separation-active layer of a membrane comprises a material that is an amorphous or semicrystalline polymer.

Claim 10: The process of claim 15, wherein the material is a polyimide of formula



wherein

each R is independently selected from the group consisting of



x and y are mol fractions,

$0 < x < 0.5$, and

$1 > y > 0.5$.

Claim 11: The apparatus of claim 1, wherein the apparatus is configured to discharge at least 95% of a retentate component of the feed stream separation stage which has been introduced into the apparatus with the crude gas stream via the second retentate stream.

Claim 12: The apparatus of claim 1, wherein the apparatus is configured to discharge not more than 5% of a retentate component of the feed stream separation stage introduced into the apparatus with the crude gas stream via the third permeate stream.

Claim 13: A process for separating a gas mixture stream comprising:
separating the gas mixture with an apparatus comprising:
membrane separation stages; and
a compressor, a vacuum pump, or both,
wherein the membrane separation stages comprise a feed stream separation stage, a retentate separation stage, and a permeate separation stage:
the feed stream separation stage is configured to separate the gas mixture stream into a first permeate stream and a first retentate stream,
the retentate separation stage is configured to divide the first retentate stream into a second permeate stream and a second retentate stream,

wherein the second permeate stream is supplied to the gas mixture stream, and the second retentate stream is withdrawn as a product, and the permeate separation stage is configured to divide the first permeate stream into a third retentate stream and a third permeate stream,

wherein the third retentate stream is supplied to the gas mixture stream, and the third permeate stream is withdrawn as a product,

the apparatus is configured not to subject the first permeate stream to any recompression,

the feed stream separation stage comprises a gas separation membrane module having a mixed gas selectivity of at least 30,

the apparatus is configured such that a gas volume recycled in the second permeate stream and in the third retentate stream totals less than 60% by volume of a crude gas stream,

the apparatus is configured to increase a concentration of at least one permeate gas of the feed stream separation stage, after recycling of the second permeate stream and of the third retentate stream, in the feed stream, compared to a concentration in the crude gas stream, and

wherein the third permeate stream comprises more than 95 vol% of a retentate gas of the feed stream separation stage in the gas mixture stream, and the second retentate system comprises more than 95 vol% of a permeate gas of the feed stream separation stage in the gas mixture stream.

Claim 14: The process of claim 13, wherein

the crude gas stream comprises:

at least one of

a crude biogas and

a crude natural gas.

Claim 15: The process of claim 13, wherein the separating comprises adjusting a pressure on a permeate side of the feed stream separation stage with a pressure-reducing valve on a retentate side of the permeate separation stage, to from 1 to 30 bar.

Claim 16: The process of claim 13, wherein the separating comprises adjusting a pressure of the first and second retentate stream with a pressure-reducing valve in the second retentate stream, to from 1 to 100 bar.

Claim 17: The process of claim 13, wherein a driving force is a partial pressure differential between a retentate side and a permeate side in the respective membrane separation stages,

wherein the partial pressure differential is generated by at least one selected from the group consisting of a compressor in the feed stream, a vacuum pump in the second permeate stream, a vacuum pump in the third permeate stream, and a permeate-side purge gas stream.

Claim 18: The process of claim 17, wherein a pressure of the permeate side of the feed stream separation stage is greater than or equal to an ambient pressure,

the driving force comprises the ambient pressure or a reduce pressure at the permeate side of the permeate separation stage.

Claim 19: The apparatus of claim 1, wherein the material is at least one selected from the group consisting of a polyimide, a polyamide, a polysulfone, a cellulose acetate, a cellulose acetate derivative, a polyphenylene oxide, a polysiloxane, a polymer with an intrinsic microporosity, a mixed matrix membrane, a facilitated transport membrane, a polyethylene oxide, and a polypropylene oxide.

Claim 20: The process of claim 14, wherein the crude gas mixture consists essentially of carbon dioxide and methane.

Claim 21: The process of claim 13, wherein the third permeate stream comprises more than 97 vol% of the carbon dioxide in the gas mixture stream.

Claim 22: The process of claim 13, wherein the second retentate stream comprises more than 97 vol% of the methane in the gas mixture stream.

Claim 23: The process of claim 13, wherein the crude gas stream comprises carbon dioxide and methane.

Claim 24: The process of claim 13, wherein the crude gas stream comprises hydrogen and methane.

Claim 25: The process of claim 13, wherein the crude gas stream comprises carbon monoxide and hydrogen.

Claim 26: The process of claim 13, wherein the apparatus is configured to pass the second permeate stream and the third retentate stream along a suction side of the compressor.

Claim 27: The process of claim 13, wherein the compressor is a multistage compressor.

Claim 28: The process of claim 13, wherein the apparatus is configured to introduce the second permeate stream, the third retentate stream, or both into the multistage compressor between two compression stages.

Claim 29: The process of claim 13, wherein the apparatus is configured to pass at least one selected from the group consisting of the first retentate stream, the second retentate stream, and the third retentate stream through a pressure-reducing valve.

Claim 30: The process of claim 13, wherein the gas separation membrane module in at least one of the membrane separation stages having a mixed gas selectivity for carbon dioxide and methane of at least 35.

Claim 31: The process of claim 13, wherein at least one of the membrane separation stages comprises more than one gas separation membrane module, wherein the modules are connected in parallel, in series, or in parallel and in series.

Claim 32: The process of claim 13, wherein at least one of the membrane separation stages comprises a gas separation membrane module comprising a hollow fiber membrane, a flat membrane, or both.

Claim 33: The process of claim 13, wherein a separation-active layer of a membrane comprises a material that is an amorphous or semicrystalline polymer.

Claim 34: A process for separating a gas mixture stream comprising methane and carbon dioxide, comprising:

separating the gas mixture with an apparatus comprising:

membrane separation stages; and,

a compressor, a vacuum pump, or both,

wherein the membrane separation stages comprise a feed stream separation stage, a retentate separation stage, and a permeate separation stage:

the feed stream separation stage is configured to separate a feed stream comprising at least two components into a first permeate stream and a first retentate stream,

the retentate separation stage is configured to divide the first retentate stream into a second permeate stream and a second retentate stream,

wherein the second permeate stream is supplied to the feed stream, and the second retentate stream is withdrawn as a product, and

the permeate separation stage is configured to divide the first permeate stream into a third retentate stream and a third permeate stream,

wherein the third retentate stream is supplied to the feed stream, and the third permeate stream is withdrawn as a product,

the apparatus is configured not to subject the first permeate stream to any recompression,

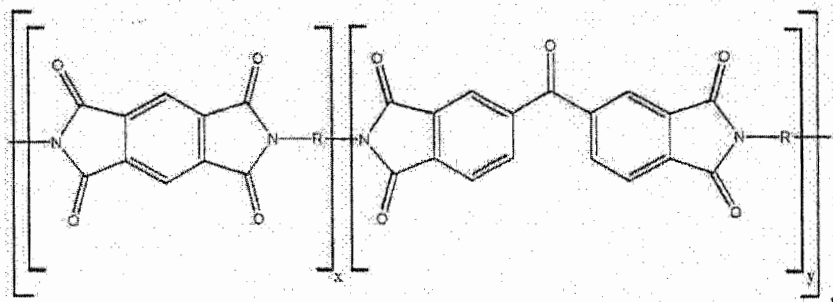
the feed stream separation stage comprises a gas separation membrane module having a mixed gas selectivity of at least 30,

the apparatus is configured such that a gas volume recycled in the second permeate stream and in the third retentate stream totals less than 60% by volume of a crude gas stream,

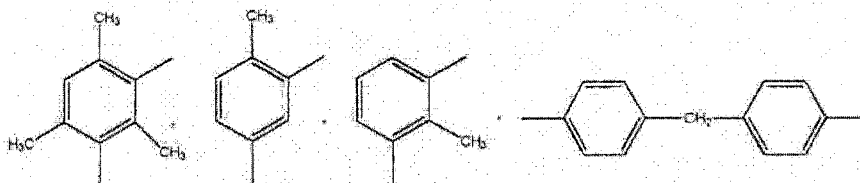
the apparatus is configured to increase a concentration of at least one permeate gas of the feed stream separation stage, after recycling of the second permeate stream and of the third retentate stream, in the feed stream, compared to a concentration in the crude gas stream.

Claim 35: The process of claim 34, wherein the material is at least one selected from the group consisting of a polyimide, a polyamide, a polysulfone, a cellulose acetate, a cellulose acetate derivative, a polyphenylene oxide, a polysiloxane, a polymer with an intrinsic microporosity, a mixed matrix membrane, a facilitated transport membrane, a polyethylene oxide, and a polypropylene oxide.

Claim 36: The process of claim 35, wherein the material is a polyimide of formula



wherein each R is independently selected from the group consisting of



x and y are mol fractions,

$0 < x < 0.5$, and

$1 > y > 0.5$.

Claim 37: The process of claim 34, wherein the crude gas stream comprises at least one gas pair selected from the group consisting of:

- carbon dioxide and methane;
- hydrogen and methane;
- carbon monoxide and hydrogen;
- a crude biogas; and
- a crude natural gas.

Claim 38: The process of claim 34, wherein the crude gas stream consists essentially of at least one gas pair selected from the group consisting of:

- carbon dioxide and methane;
- hydrogen and methane;
- carbon monoxide and hydrogen;
- a crude biogas; and

a crude natural gas.

Claim 39: The process of claim 34, wherein the third permeate stream comprises less than 5 vol. % of the retentate gas compared to the amount of said retentate gas originally introduced into the process with crude gas stream and that the second retentate stream comprises more than 95 vol. % of the retentate gas of the feed stream separation stage compared to the amount of said retentate gas originally introduced into the process with the crude gas stream.

Claim 40: The process of claim 39, wherein a driving force is a partial pressure differential between a retentate side and a permeate side in the respective membrane separation stages, and

wherein the partial pressure differential is generated by at least one selected from the group consisting of a compressor in the feed stream, a vacuum pump in the second permeate stream, a vacuum pump in the third permeate stream, and a permeate-side purge gas stream.

Claim 41: The process of claim 39, wherein a pressure of the permeate side of the feed stream separation stage is greater than or equal to an ambient pressure,

the driving force comprises the ambient pressure or a reduce pressure at the permeate side of the permeate separation stage.

Claim 42: The process of claim 39, wherein the material is at least one selected from the group consisting of a polyimide, a polyamide, a polysulfone, a cellulose acetate, a cellulose acetate derivative, a polyphenylene oxide, a polysiloxane, a polymer with an intrinsic

microporosity, a mixed matrix membrane, a facilitated transport membrane, a polyethylene oxide, and a polypropylene oxide.

Claim 43: The process of claim 34, wherein the separating comprises adjusting a pressure of the first and second retentate stream with a pressure-reducing valve in the second retentate stream, to from 1 to 100 bar.

Claim 44: The process of claim 34, wherein the separating comprises adjusting a pressure on a permeate side of the feed stream separation stage with a pressure-reducing valve on a retentate side of the permeate separation stage, to from 1 to 30 bar.

Claim 45: The process of claim 34, wherein the gas separation membrane module in at least one of the membrane separation stages has a mixed gas selectivity of at least 35.

Claim 46: The process of claim 34, wherein the gas separation membrane module in at least one of the membrane separation stages has a mixed gas selectivity of at least 45.

Claim 47: The process of claim 34, wherein the apparatus comprises at least one gas separation membrane module comprising hollow polyimide fibers having a separation area of about 5m^2 .

Claim 48: The process of claim 34, wherein the apparatus is configured to pass the second permeate stream and the third retentate stream along a suction side of the compressor.

Claim 49: The process of claim 34, wherein the compressor is a multistage compressor.

Claim 50: The process of claim 34, wherein the apparatus is configured to introduce the second permeate stream, the third retentate stream, or both into the multistage compressor between two compression stages.

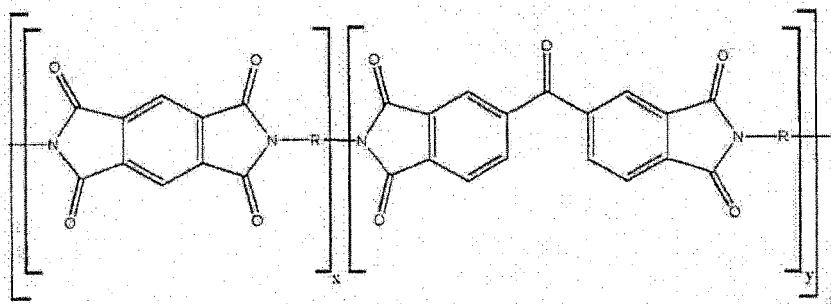
Claim 51: The process of claim 34, wherein the apparatus is configured to pass at least one selected from the group consisting of the first retentate stream, the second retentate stream, and the third retentate stream through a pressure-reducing valve.

Claim 52: The process of claim 34, wherein at least one of the membrane separation stages comprises more than one gas separation membrane module, wherein the modules are connected in parallel, in series, or in parallel and in series.

Claim 53: The process of claim 34, wherein at least one of the membrane separation stages comprises a gas separation membrane module comprising a hollow fiber membrane, a flat membrane, or both.

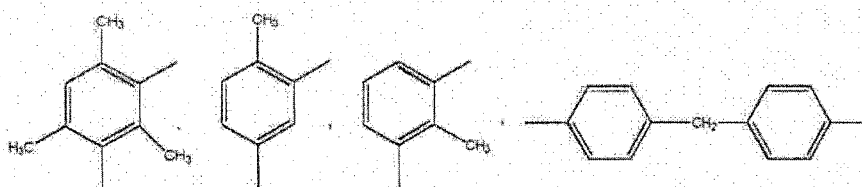
Claim 54: The process of claim 39, wherein a separation-active layer of a membrane comprises a material that is an amorphous or semicrystalline polymer.

Claim 55: The process of claim 39, wherein the material is a polyimide of formula



wherein

each R is independently selected from the group consisting of



x and y are mol fractions,

$0 < x < 0.5$, and

$1 > y > 0.5$.

Claim 56: The apparatus of claim 1, wherein the second retentate stream comprises more than 97 vol% of the methane in the gas mixture stream.

Claim 57: The apparatus of claim 1, wherein the apparatus comprises at least one gas separation membrane module having a gas selectivity for methane and carbon dioxide of at least 45.

Claim 58: The apparatus of claim 1, wherein the apparatus comprises at least one gas separation membrane module comprising hollow polyimide fibers having a separation area of about 5m².