

separation stage (2) and the third retentate stream (10a + 10b) of the permeate separation stage (3) are recycled and mixed with the raw gas stream. By controlling the flow rates of the two streams (9a) and (10a) and hence the permeate pressure of the retentate separation stage (2) and/or the retentate pressure of the permeate separation stage (3), the inventors surprisingly succeeded in achieving the stated objects.

The inventors thereby succeeded in providing a method and a plant in each of which the purities and yields of the product streams of retentate separation stage (2) and of the permeate separation stage (3) can be controlled independently of each other. Very high yields coupled with very good purities are obtainable even in the event of fluctuations in the raw gas stream.

In a first aspect, there is provided an apparatus for separating gases, comprising as membrane separation stages at least a feed stream separation stage (1), a retentate separation stage (2) and a permeate separation stage (3) and also at least one means selected from the group consisting of a compressor (4), arranged on the feed side of said feed stream separation stage (1), ~~and/or at least one, preferably one or two, a vacuum pump(s) arranged downstream of said feed stream separation stage (1), preferably on the permeate side of said retentate separation stage (2) in a second permeate stream (9a + 9b) and/or on the permeate side of said permeate separation stage (3) in a third permeate stream (11), wherein~~

said feed stream separation stage (1) separates a feed stream (5), consisting of two or more components, into a first permeate stream (6) and a first retentate stream (7),

said retentate separation stage (2) divides said first retentate stream (7) into a second permeate stream (9a + 9b), wherein (9a) characterizes the part-stream upstream of a permeate control means (18) and downstream of said retentate separation stage (2) and (9b) characterizes the part-stream downstream of said permeate control means (18), and said part-stream (9b) is supplied to said feed stream (5), and a second retentate stream (8) which is removed as product or further processed,

said permeate separation stage (3) divides said first permeate stream (6) into a third retentate stream (10a + 10b), wherein (10a) characterizes the part-stream upstream of a retentate control means (19) and downstream of said permeate separation stage (3) and (10b) characterizes the part-stream downstream of said retentate control means (19), and said part-stream (10b) is supplied to said feed stream (5), and a third permeate stream (11), which is removed as product or further processed or discarded,

characterized in that

said second permeate stream (9a + 9b) comprises at least one permeate control means (18) with which the permeate pressure of said retentate separation stage (2) can be raised or lowered and which is controlled on the basis of measured values from one or more

measuring means (20a) in said first retentate stream (7) ~~and/or~~ one or more measuring means (20b) in said second retentate stream (8) or from both, wherein said measuring means are preferably different,

and/or

said third retentate stream (10a + 10b) comprises at least one retentate control means (19) with which the retentate pressure of said permeate separation stage (3) can be raised or lowered and which is controlled on the basis of measured values from one or more

measuring means (21a) in said first permeate stream (6) ~~and/or~~ one or more measuring means (21b) in said third permeate stream (11) or both, wherein the measuring means are preferably different.

In a second aspect, there is provided a method of controlling a gas separation plant, the separation plant comprising an apparatus as disclosed above characterized in that

- i. when the concentration of a less readily permeating component B, optionally also determined through a parameter correlating therewith, of said second retentate stream (8) falls below a predetermined setpoint value, the pressure of said second permeate stream (9a) is lowered by said permeate control means (18) until said concentration/parameter is back in the setpoint range,
- ii. when the concentration of a less readily permeating component B, optionally also determined through a parameter correlating therewith, of said second retentate stream (8) rises above a predetermined setpoint value, the pressure of said second permeate stream (9a) is raised by said permeate control means (18) until said concentration/parameter is back in the setpoint range,

and/or in that

- iii. when the concentration of a less readily permeating component B, optionally also determined through a parameter correlating therewith, of said third permeate stream (11) falls below a predetermined setpoint value, the pressure of said third retentate stream (10a) is raised by said retentate control means (19) until said concentration/parameter is back in the setpoint range,
- iv. when the concentration of a less readily permeating component B, optionally also determined through a parameter correlating therewith, of said third permeate stream (11) rises above a predetermined setpoint value, the pressure of said third retentate stream (10a) is

lowered by said retentate control means (19) until said concentration/parameter is back in the setpoint range.

In a third aspect, there is provided a method of controlling a gas separation plant, the separation plant comprising an apparatus as disclosed above characterized in that

- v. when the volume flow of said first retentate stream (7) increases, the pressure of said second permeate stream (9a) is lowered by said permeate control means (18) until a property of said second retentate stream (8) which is correlated by a calibration curve with said volume flow of said first retentate stream (7), preferably the composition of said second retentate stream (8), is back in the setpoint range,
- vi. when the volume flow of said first retentate stream (7) decreases, the pressure of said second permeate stream (9a) is raised by said permeate control means (18) until a property of said second retentate stream (8) which is correlated by a calibration curve with said volume flow of said first retentate stream (7), preferably the composition of said second retentate stream (8), is back in the setpoint range,

and/or

- vii. when the volume flow of said first permeate stream (6) increases, the pressure of said third retentate stream (10a) is raised by said retentate control means (19) until a property of said third permeate stream (11) which is correlated by a calibration curve with said volume flow of said first permeate stream (6), preferably the composition of said third permeate stream (11), is back in the setpoint range,
- viii. when the volume flow of said first permeate stream (6) decreases, the pressure of said third retentate stream (10a) is lowered by said retentate control means (19) until a property of said third permeate stream (11) which is correlated by a calibration curve with said volume flow of said first permeate stream (6), preferably the composition of said third permeate stream (11), is back in the setpoint range.

Claims

1. An apparatus for separating gases, comprising as membrane separation stages at least a feed stream separation stage (1), a retentate separation stage (2) and a permeate separation stage (3) and also at least one means selected from the group consisting of a compressor (4), arranged on the feed side of said feed stream separation stage (1), ~~and /or at least one, preferably one or two, a vacuum pump(s)~~ arranged downstream of ~~said feed stream separation stage (1),~~ preferably on the permeate side of said retentate separation stage (2) in a second permeate stream (9a + 9b) ~~and/or~~ on the permeate side of said permeate separation stage (3) in a third permeate stream (11),

wherein

said feed stream separation stage (1) separates a feed stream (5), consisting of two or more components, into a first permeate stream (6) and a first retentate stream (7),

said retentate separation stage (2) divides said first retentate stream (7) into a second permeate stream (9a + 9b), wherein (9a) characterizes the part-stream upstream of a permeate control means (18) and downstream of said retentate separation stage (2) and (9b) characterizes the part-stream downstream of said permeate control means (18), and said part-stream (9b) is supplied to said feed stream (5), and a second retentate stream (8) which is removed as product or further processed,

said permeate separation stage (3) divides said first permeate stream (6) into a third retentate stream (10a + 10b), wherein (10a) characterizes the part-stream upstream of a retentate control means (19) and downstream of said permeate separation stage (3) and (10b) characterizes the part-stream downstream of said retentate control means (19), and said part-stream (10b) is supplied to said feed stream (5), and a third permeate stream (11), which is removed as product or further processed or discarded,

characterized in that

- said second permeate stream (9a + 9b) comprises at least one permeate control means (18) with which the permeate pressure of said retentate separation stage (2) can be raised or lowered and which is controlled on the basis of measured values from one or more measuring means (20a) in said first retentate stream (7) ~~and/or~~ one or more measuring means (20b) in said second retentate stream (8) or from both, wherein said measuring means are preferably different, and/or

- said third retentate stream (10a + 10b) comprises at least one retentate control means (19) with which the retentate pressure of said permeate separation stage (3) can be raised or lowered and which is controlled on the basis of measured values from one or more measuring means (21a) in said first permeate stream (6) ~~and/or~~ one or more measuring means (21b) in said third permeate stream (11) or both, wherein the measuring means are preferably different.

2. An apparatus according to Claim 1,

characterized ~~in that~~ by one or more of the following features

- ~~the~~ said first permeate stream (6) is not subjected to recompression,

~~and/or~~

- gas separation membrane modules having a mixed gas selectivity CO_2/CH_4 of not less than 30, preferably not less than 35, more preferably not less than 40 and most preferably not less than 45 are used at least in said feed stream separation stage (1), but preferably in all three membrane separation stages (1) to (3),

~~and/or~~

- at least one of said membrane separation stages (1) to (3) comprises more than one gas separation membrane module interconnected in parallel and/or series,

~~and/or~~

- the gas separation membrane module(s) consist(s) of hollow fibre membranes and/or flat membranes,

~~and/or~~

- said apparatus is configured such that the gas volume recycled in said second permeate stream (9b) and in said third retentate stream (10b) amounts in total to less than 60% by volume of the volume of ~~the a~~ raw gas stream (17),

~~and/or~~

- said apparatus is configured such that the concentration of at least one permeate gas of said feed stream separation stage (1), after returning said second permeate stream (9b) and said third retentate stream (10b), is raised in said feed stream (5), preferably by not less than 2%, more preferably by not less than 3% and even more preferably by 3 to 40%, all compared with the concentration in said raw gas stream (17).

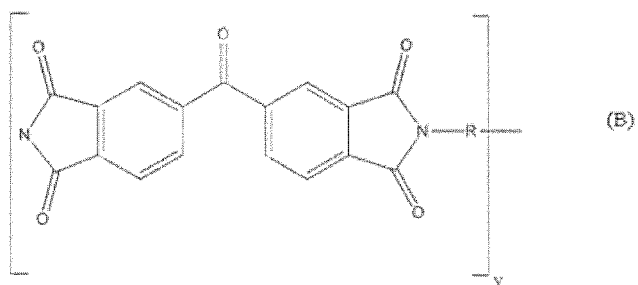
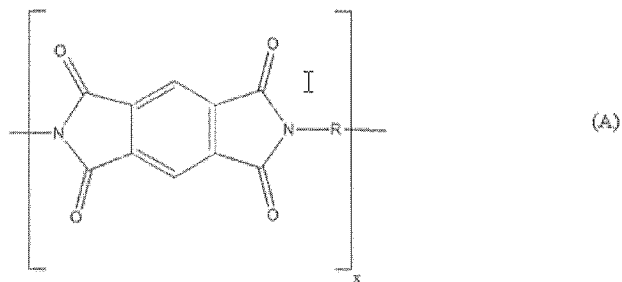
3. An apparatus according to Claim 1 or 2,

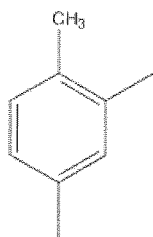
characterized by one or more of the following features ~~in that~~

- said second permeate stream (9b) and said third retentate stream (10b) are led to the suction side of said compressor (4),
and/or in that
 - a multi-stage compressor (4) is used, wherein preferably said second permeate stream (9b) and/or said third retentate stream (10b) is/are introduced into said compressor (4) between two compression stages,
 - and/or in that said compressor (4) is arranged in said apparatus such that it generates a pressure gradient in said feed stream separation stage (1).
- 4. An apparatus according to any of Claims 1 to 3,
characterized in that
 - said apparatus comprises a controller means (24) which adapts the power requirements of said compressor (4), preferably its rotary speed, to changes in at least one stream selected from the group consisting of the said second permeate stream (9b), the and/or said third retentate stream (10b) and the /or said raw gas stream (17),
and/or in that
 - said apparatus is configured such that changing amounts of recycled gas from said second permeate stream (9b) and/or said third retentate stream (10b) or from both are equalized, preferably automatically, by a regulation of the supplied amount of raw gas, preferably via a raw gas control means (25), preferably without changing the rotary speed of said compressor (4).
- 5. An apparatus according to any of Claims 1 to 4,
characterized in that
 - flow meters are used as measuring means (20a) and/or (21a) or in both means in said first retentate stream (7) and/or in said first permeate stream (6),
or in that
 - an online or offline measuring means (20b) and/or (21b) is used in said second retentate stream (8) and/or in said third permeate stream (11) to determine the composition of the particular gas mixture.
- 6. An apparatus according to any of Claims 1 to 5,
characterized in that
 - the materials used for the separation-active layer of the membranes are amorphous or partly crystalline plastics such as, for example but not exclusively, polyimides, polyamides, polysulphones, cellulose acetates and derivatives, polyphenylene

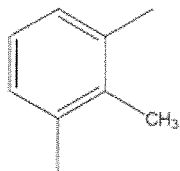
oxides, polysiloxanes, polymers having intrinsic microporosity, mixed matrix membranes, facilitated transport membranes, polyethylene oxides, polypropylene oxides or mixtures thereof.

7. An apparatus according to Claim 6, characterized in that the material used for the separation-active layer of the membranes is a polyimide of the general formula A_xB_y .

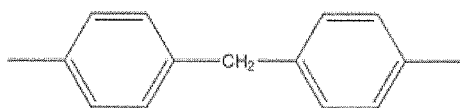




(L2)



(L3)



(L4)

where $0 \leq x \leq 0.5$ and $1 \geq y \geq 0.5$ and R corresponds to one or more, identical or different radicals R selected from the group consisting of the radicals L1, L2, L3 and L4, preferably a polyimide of CAS number 9046-51-9 and/or a polyimide of CAS number 134119-41-8.

8. An apparatus according to any of Claims 1 to 7, characterized in that
 - not less than 95%, preferably not less than 97%, more preferably not less than 99% and most preferably not less than 99.5% of the feed stream separation stage (1) retentate component led into said apparatus with said raw gas stream (17) is removed via said second retentate stream (8), and/or 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 feed stream separation stage (1) retentate component led into said apparatus with said raw gas stream (17) is removed via said third permeate stream (11).
9. A method of controlling a gas separation plant, the separation plant comprising an apparatus according to any of Claims 1 to 8, characterized in that
 - i. when the concentration of a less readily permeating component B, optionally also determined through a parameter correlating therewith,

of said second retentate stream (8) falls below a predetermined setpoint value, the pressure of said second permeate stream (9a) is lowered by said permeate control means (18) until said concentration/parameter is back in the setpoint range,

- ii. when the concentration of a less readily permeating component B, optionally also determined through a parameter correlating therewith, of said second retentate stream (8) rises above a predetermined setpoint value, the pressure of said second permeate stream (9a) is raised by said permeate control means (18) until said concentration/parameter is back in the setpoint range,

and/or in that

- iii. when the concentration of a less readily permeating component B, optionally also determined through a parameter correlating therewith, of said third permeate stream (11) falls below a predetermined setpoint value, the pressure of said third retentate stream (10a) is raised by said retentate control means (19) until said concentration/parameter is back in the setpoint range,
- iv. when the concentration of a less readily permeating component B, optionally also determined through a parameter correlating therewith, of said third permeate stream (11) rises above a predetermined setpoint value, the pressure of said third retentate stream (10a) is lowered by said retentate control means (19) until said concentration/parameter is back in the setpoint range.

10. A method according to Claim 9,

characterized in that

the concentration is determined online and/or offline.

11. A method of controlling a gas separation plant, the separation plant comprising an apparatus according to any of Claims 1 to 8,

characterized in that

- v. when the volume flow of said first retentate stream (7) increases, the pressure of said second permeate stream (9a) is lowered by said permeate control means (18) until a property of said second retentate stream (8) which is correlated by a calibration curve with said volume

14. A method according to any of Claims 89 to 13,
characterized in that

the driving force used for the separation task is a partial pressure difference between the retentate side and the permeate side in the particular membrane separation stages, wherein said partial pressure difference is generated by at least one means selected from the group consisting of

- a compressor (4), which is arranged on the feed side of said feed stream separation stage (1),
- and optionally by at least one, preferably one or two, vacuum pump(s) arranged in said second permeate stream and/or third permeate stream (9a + 9b) and/or said third permeate stream (11), and/or by
- a permeate-side purge gas stream,

and/or in that

the pressure of the permeate of said feed stream separation stage (1) is in an equal or elevated state relative to the ambient pressure, so there is still a partial pressure difference between the retentate and the permeate of said permeate separation stage (3) and hence there is a driving force in the event that said permeate of said permeate separation stage (3) is at ambient pressure or negative pressure is applied.

15. A method according to any of Claims 9 to 14,
characterized by at least one of the following features ~~in that~~

- a controller means (24) adapts the power requirements of said compressor (4), preferably its rotary speed, to changes in at least one stream selected from the group consisting of the second permeate stream (9b), and/or said the third retentate stream (10b) and/or said the raw gas stream (17),

~~and/or in that~~

- changing amounts of recycled gas from said second permeate stream (9b) and/or said third retentate stream (10b) or both are equalized, preferably automatically, by a regulation of the supplied amount of raw gas, preferably via said raw gas control means (25), preferably without changing the rotary speed of said compressor, or in that

16. A method according to any of Claims 9 to 15, characterized in that

the performance of the plant ~~of the present invention~~ is raised or lowered by changing the volume throughput of said compressor (4), wherein one or more of the following results are applied

- a resultant change in the concentration of said less readily permeating component B in said second retentate stream (8) is counteracted as per method alternatives i/ii, and/or

- a resultant change in the concentration of said less readily permeating component B in said third permeate stream (11) is counteracted as per method alternatives iii/iv,
and/or
- a resultant change in the flow rate of said first retentate stream (7) is counteracted according to method alternatives v/vi,
and/or
- a resultant change in the flow rate of said first permeate stream (6) is counteracted according to method alternatives vii/viii.

1746. A method according to any of Claims 9 to ~~15~~16, characterized in that said method is practised in the context of operating a biogas plant and in that the rotary speed of the compressor and hence the volume throughput of said compressor (4) is controlled according to said biogas plant fill level, preferably determined via fermenter pressure or intermediate store fill level, in order that the fill level in the fermenter and/or intermediate store may be changed or kept constant, and/or 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 methane or predominantly but not exclusively carbon monoxide and hydrogen or raw biogas or raw natural gas.

1847. A biogas plant comprising an apparatus according to any of Claims 1 to 8.