HU000033254T2

(19) **HU** (11) Lajstromszám: **E 033 254** (13) **T2**

MAGYARORSZÁG
Szellemi Tulajdon Nemzeti Hivatala

EURÓPAI SZABADALOM
SZÖVEGÉNEK FORDÍTÁSA

(21) Magyar ügyszám: **E 11 722404**
(22) A bejelentés napja: **2011. 05. 26.**

(51) Int. Cl.: ***B01D 53/22*** *(2006.01)*
C10L 3/10 *(2006.01)*

(96) Az európai bejelentés bejelentési száma:
EP 20110722404
(97) Az európai bejelentés közzétételi adatai:
EP 2588217 A1 **2012. 01. 05.**
(97) Az európai szabadalom megadásának meghirdetési adatai:
EP 2588217 B1 **2017. 02. 22.**

(86) A nemzetközi (PCT) bejelentési szám:
PCT/EP 11/058636
(87) A nemzetközi közzétételi szám:
WO 12000727

(30) Elsőbbségi adatok:
10168121 **2010. 07. 01.** **EP**

(72) Feltaláló(k):
UNGERANK, Markus, A-4320 Perg (AT)
BAUMGARTEN, Goetz, 45721 Haltern am See (DE)
PRISKE, Markus, 40472 Düsseldorf (DE)
ROEGL, Harald, A-4702 Wallern an der Trattnach (AT)

(73) Jogosult(ak):
Evonik Fibres GmbH, 4861 Schörfling am Attersee (AT)

(74) Képviselő:
Danubia Szabadalmi és Jogi Iroda Kft., Budapest

(54) **Eljárás gázok szétválasztására**

Az európai szabadalom ellen, megadásának az Európai Szabadalmi Közlönyben való meghirdetésétől számított kilenc hónapon belül, felszólalást lehet benyújtani az Európai Szabadalmi Hivatalnál. (Európai Szabadalmi Egyezmény 99. cikk(1))

A fordítást a szabadalmas az 1995. évi XXXIII. törvény 84/H. §-a szerint nyújtotta be. A fordítás tartalmi helyességét a Szellemi Tulajdon Nemzeti Hivatala nem vizsgálta.

Process for separation of gases

The invention relates to a process using an apparatus having a chain of gas separation membrane modules, for separation of gas mixtures into two fractions each of increased purity.

By means of a gas separation membrane, it is possible to separate gas mixtures on the basis of different permeabilities (= flow rate per unit time, unit area, pressure differential and layer thickness) of the individual gases in a polymer. The polymers are generally processed to give hollow fibers or flat membranes. The membranes are notable for a very thin separation layer at the surface of the membrane, such that the permeance (flow rate per unit time, unit area and pressure differential) of the membrane is at a maximum.

The separation outcome achievable with a membrane in a single pass depends not only on the selectivity of the membrane but also on the pressure ratio between high-pressure and low-pressure side of the membrane. The greater the pressure ratio, the better the maximum separation outcome achievable.

In the range of low pressure ratios, the curves for the different selectivities run close to one another (see figure 1). The separation outcome in this range is determined by the pressure ratio. This range is therefore referred to as "pressure-limited". In the range of high pressure ratios, the separation outcome can barely be influenced by the pressure ratio. This range is referred to as "selectivity-limited".

A series of membrane connection arrangements for the separation of gases is known from the literature. In Baker, IndEngChemRes, Natural Gas Processing with Membranes, 47 (2008), there is a comprehensive list of various known connection arrangements. For instance, using the example of the separation of methane (retentate gas) and CO_2 (permeate gas) with a simple one-stage membrane separation step according to fig. 2, high purities can be achieved in the product stream (98% CH_4). However, the quality of the permeate-side offgas stream cannot be influenced and is of low purity (44% CO_2). There is correspondingly large slippage, i.e. loss of methane.

In the case of a one-stage connection arrangement with recycling (fig. 3), the purities can be increased slightly. However, it is not possible thereby to solve the fundamental problem of the low retentate gas yield and of the permeate gas quality.

For higher permeate purities and higher yields of the component retained, a series of multistage connection arrangements are known. In a connection arrangement with a single permeate stage, with recompression and recycling of the first permeate (fig. 4), it is possible to improve permeate purity (86%) and methane yield.

If the better-permeating component occurs in increased concentrations (30%), a connection arrangement according to fig. 5 may be advantageous according to the literature. Two permeate gas-rich streams (82% CO_2, 93% CO_2) and one retentate gas-rich stream (82% CH_4) are produced.

Bhide (MemSci, Hybrid processes for the removal of acid gases from natural gas, 1998) discloses a three-stage operation for the removal of acid gases from natural gas (fig. 6). The gas stream to be treated is subjected to a coarse purification in an upstream membrane. The prepurified natural gas which remains on the pressure side is conducted through a further membrane. On the retentate side, the target product stream forms, the natural gas depleted of the acid gases. The acid gas-enriched stream on the permeate side is compressed and supplied to a further membrane stage. The retentate of this stage is compressed to the pressure level of the upstream stage and recirculated upstream thereof. This additional energy expenditure and financial investment is intended to increase the yield of methane. Disadvantages of this connection arrangement are the expenditures for the compressors and the inadequate purities of the acid gas-enriched stream.

Chenar (MemSci Application of Cardo-type polyimide (PI) and polyphenylene oxide (PPO) hollow, 2008) describes an operation with staged arrangement for permeate with recompression (fig. 7). Disadvantages of this connection arrangement are the severely limited purities of the retentate gas and the additional expenditure for the recompression.

EP0 799 634 discloses a connection arrangement according to fig. 8. However, disadvantages are an additional potential introduction of oil or water as a sealant and lubricant, additional high capital costs, increased energy consumption resulting from additional compression, and increased likelihood of failure as a result of the moving parts.

Disadvantages of the abovementioned processes are the additional recompression and the purities, which are still much too low, of retentate gas in the overall retentate stream, and the insufficient yields of the retentate gas in the retentate stream.

Fig. 9 shows a technology which is frequently proposed and has also been implemented particularly for the processing of biogas (Air Liquide and Harasek). What is disclosed is a connection arrangement with staged arrangement for retentate with permeate recycling from the second stage.

Eindhoven University of Technology has developed a connection arrangement with an internal staged arrangement, which enables methane purities of more than 90% (see fig. 10). A disadvantage of this connection arrangement is the inadequate purity of the retentate gas.

EP 0 603 798 discloses a multistage connection arrangement for production of nitrogen. The disadvantage of this process is the insufficient purity of the permeating component and the use of at least two compressors.

EP0695574 discloses a connection arrangement with partial utilization of a permeate stream as a sweep stream for the production of a retentate of maximum purity. A disadvantage of this process is the insufficient quality of the overall permeate.

US5753011 discloses a process in which combination of membrane separation steps with staged arrangement for the permeate and pressure swing adsorption, PSA, achieves high purities for two product streams. A disadvantage of this process is the use of a costly and inconvenient PSA.

EP1634946 discloses a process for processing of biogas. Described therein is a thermal utilization of the methane from the methane-depleted stream. Disadvantages are the high costs and the loss of the gas.

EP0596268, finally, discloses various connection arrangements for production of three different gas compositions.

US 6,168,649 B1 discloses membrane separation processes with three membrane separation stages, wherein a retentate stream and a permeate stream from stages 2 and 3 respectively are recycled to the crude gas stream.

US 6 565 626 B1 discloses a process for separating gases in a crude gas stream, which is conducted in an apparatus comprising a feed stream separation stage, a retentate separation stage and a permeate separation stage, each in the form of membrane separation stages, and at least one compressor, wherein the feed stream separation stage separates a feed stream consisting of at least two components A and B, A being the more readily permeating component, into a first permeate stream and a first retentate stream, the retentate separation stage divides the first retentate stream into a second permeate stream which is sent to the crude gas stream, and a second permeate stream, product enriched in component B is withdrawn, the permeate separation stage divides the first permeate stream into a third retentate stream which is sent to the crude gas stream, and a third permeate stream which is withdrawn as a second product enriched in component A, the first permeate stream is not subjected to any recompression, the gas volume recycled in the second permeate stream and in the third retentate stream totals less than 60% of the volume of the crude gas stream, and wherein the concentration of the component

Example, in the processing and feeding of biogas into the natural gas grid as a result of the high demands on the methane purity and as a result of the limitation of methane slippage necessary is, there are no processes which do not need a recompression unit or a further purification of the permeate or retentate stream (for example thermal postcombustion of the permeate stream or pressure swing adsorption for the retentate stream).

Proceeding from this prior art, it was an object of the present invention to provide processes for separation and purification of gas mixtures, which have the disadvantages of the prior art processes only to a reduced degree, if at all. More particularly, processes and apparatuses which afford both permeate gas and retentate gas are to be provided in high purities. In a further specific object, this process and this apparatus are to be advantageous in terms of investment and operating costs and/or enable a simpler process regime.

In a next specific object, a process/apparatus usable with maximum universality is to be provided for any desired gas mixtures. More particularly, it should be possible to separate gas streams which are obtained in the production of biogas and natural gas, or which comprise carbon dioxide and methane in the presence of other gases such as water, hydrogen, hydrogen sulfide.

It was a further specific object of the present invention to provide a process which enables purification of methane-containing crude gas streams, with reduced methane emission compared to the prior art processes at the same throughput, and hence reduced environment pollution by this strong greenhouse gas.

Further objects which are not stated explicitly are evident from the overall context of the claims, description, examples and figures which follow.

It has now been found that, surprisingly, the process according to Claim 1 can afford pure streams of permeate and retentate without requiring more than one compressor or any need to purify the permeate or retentate stream further by other methods. The process according to the invention thus enables simultaneous achievement of permeate and retentate streams in high purity. The capital costs for the plant are low; it does not need any additional downstream purification processes. It was thus possible to achieve the stated objects with a pure membrane separation process.

The present invention is described in detail hereinafter. First, some important terms are defined.

The quotient of the permeances of the individual gases gives the **selectivity** of the membrane for separation with regard to the two gases, and thus states how well the membrane can separate a gas mixture with regard to the two components. **Permeate** refers to the overall stream obtained on the low-pressure side of the membrane, membrane module or membrane separation step.

Permeate gas refers in each case to the component(s) enriched in each case **in the permeate stream** relative to the respective input stream at the membrane, at the membrane module or in the membrane separation step.

Retentate refers to the entire stream which is obtained on the high-pressure side of the membrane, membrane modules or membrane separation step and does not pass through the membrane.

Retentate gas refers to the component(s) enriched in each case in the **retentate stream** relative to the respective input stream at the membrane, at the membrane module or in the membrane separation step.

Crude gas or crude gas mixture or crude gas stream (17) refers to a gas mixture of at least two gases or a stream of this gas mixture which is to be separated by means of the process according to the invention.

Feed stream (5) refers to a gas stream which is supplied to the feed stream separation stage (1). This stream, for the performance of the process, may correspond to the crude gas stream (17) or to the crude gas stream compressed by a compressor. After recycling of the second permeate stream (9) or of the third retentate stream (10), the feed stream (5) is composed of the gases of the crude gas stream (17), of the second permeate stream (9) and of the third retentate stream (10). The feed stream (5) can be produced either by mixing streams (9) and (10) both with the uncompressed crude gas stream (17), or mixing both with the compressed crude gas stream or mixing one with the uncompressed crude gas stream and one with the compressed crude gas stream, or by mixing streams (9) and/or (10) with the crude gas stream (17) in the compressor. Combinations of the variants described above are also encompassed by the present invention.

Feed stream separation stage (1) refers to a membrane separation stage for separation of the feed stream (5) into a first permeate stream (6) and a first retentate stream (7).

Retentate separation stage (2) refers to a membrane separation stage, which may be of identical or different construction to the feed stream separation stage (1), for separation of the first retentate stream (7) into a second permeate stream (9) and a second retentate stream (8).

Permeate separation stage (3) refers to a membrane separation stage, which may be of identical or different construction to the feed stream separation stage (1) and retentate separation stage (2), for separation of the first permeate stream (6) into a third permeate stream (11) and a third retentate stream (10).

The descriptions which follow of preferred and specific embodiments of the process according to the invention and the preferred and particularly suitable executions and the drawings and descriptions of the drawings illustrate the invention in detail merely by way of example, which means that it is not restricted to these execution and use examples or to the respective combinations of features within individual execution examples.

Individual features which are specified and/or described in connection with specific execution examples are not restricted to these execution examples or the combination with the other features of these execution examples; instead, they can be combined, within the range of technical possibilities, with any other variants, even if they are not discussed specifically in the present documents.

Identical reference symbols in the individual figures and diagrams of the drawings refer to identical or similar or identically or similarly acting components. The diagrams in the drawing also make clear those features which are not provided with reference numerals, irrespective of whether such features are described hereinafter or not. On the other hand, features which are included in the present description but are not visible or represented in the drawing are also immediately understandable to a person skilled in the art.

The process according to the invention comprises the use of a chain of three membrane separation stages. Each stage consists of one or more physical gas separation modules, which are connected in parallel and/or in series within one stage. The driving force produced for the gas separation in the modules is a partial pressure differential between the retentate side and the permeate side in the respective membrane separation stages. The partial pressure differential can be generated either by means of a compressor (4) arranged on the feed side of the feed stream separation stage (1) and/or by means of at least one, preferably one or two, vacuum pump(s) (not shown in figs. 11 to 13), preferably in the second permeate stream (9) on the permeate side of the retentate separation stage (2) and/or in the third permeate stream (11) on the permeate side permeate separation stage (3).

It may optionally be advantageous, in one or more of the membrane separation stages, to generate or to enhance the partial pressure difference by means of a permeate-side purge gas stream.

In a preferred configuration of the present invention, a compressor (4) brings the crude gas mixture or the gas mixture of the crude gas stream (17) and the second permeate stream (9) and/or the third retentate stream (10) to the desired pressure in the range from 5 to 100 bar, but preferably to a pressure of 9 to 75 bar. The resulting feed stream (5) is introduced into the feed stream separation stage (1). In the feed stream separation stage (1), a preseparation of the crude gas mixture into more easily permeating components (permeate gas), which pass mainly into the permeate of the first stage, and components which permeate less quickly (retentate gas), which are predominantly retained by the membrane and are enriched in the retentate, is obtained.

The process according to the invention is notable in that it/it is configured such that 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 by at least 2%, more preferably by at least 3% and most preferably by 3 to 40%, in each case compared to the concentration in the crude gas stream (17). The increase may depend on the composition of the crude gas stream (17) and is particularly marked in the case of low concentrations of a permeate gas (10 to 20%). In general, the increase in concentration of one of the permeate gases is between 2 and 15%, more preferably between 3 and 8%, when the content of the permeate gas in the crude gas stream (17) is between 30 and 70%. This is because the inventors have found that the yield of retentate gas in the overall process increases and hence the loss of retentate gas decreases when the concentration of the permeate gas in the feed stream separation stage (1) is increased. At the same cut in the separation stage (=ratio of permeate stream to feed stream in the stage in question), much less permeate gas passes into the permeate of the feed stream separation stage (1) when the concentration of at least one component A which permeates relatively easily in the feed stream separation stage (1) or of a permeate gas (A) is increased in feed stream (5). Analogously, a lowering was found when the concentration of component A or of a permeate gas A in the feed stream (5) to be purified is reduced. For instance, the cut in the separation stage for a concentration of 50% of a component A or of a permeate gas A in the feed stream (5) to be purified is between 10 and 60%, preferably between 15 and 55% and more preferably between 20 and 50%. In a particularly preferred embodiment of the present invention, the process according to the invention is therefore configured such that the content of permeate gas(es) of the feed stream separation stage (1) in feed stream (5) is greater than or equal to 40% by volume, preferably more than 50% by volume and very particularly more than 55% by volume, based on the volume of the feed stream (5), after recycling of the second permeate stream (9) and of the third retentate stream (10).

This increase in concentration of the permeate gases in the feed stream (5), as already explained, increases the efficiency of the feed stream separation stage (1), which in turn has the result that less retentate gas B passes into the first permeate stream (6). This in turn increases the efficiency of the permeate separation stage (3) and ensures that less unwanted retentate gas passes into the third permeate stream (10) here too. Especially in the case of separation of methane-containing crude gases, this leads to the advantage that the unwanted emissions of methane, which is harmful to the climate, were reduced significantly.

In general, it can be stated that, in the feed stream separation stage (1), preferably 20 to 100%, more preferably 40 to 70%, of component A or of a permeate gas A are transferred from the feed stream (5) to the permeate.

The retentate of the feed stream separation stage (1), optionally with pressure reduction by an optionally present pressure-reducing valve (12), or with a pressure increase, is supplied by means of the first retentate stream (7) to the retentate separation stage (2) in which the fine purification is effected. On the retentate side of the retentate separation stage (2), i.e. in the second retentate stream (8), there is preferably a pressure-reducing valve (13), by means of which the pressure in the system can be maintained and kept constant. The content of the less easily permeating components or of a retentate gas B is increased further in the retentate separation stage (2), such that the content of component B or of a retentate gas B in the second retentate stream (8) is more than 90%, preferably more than 95% and more preferably more than 97%. In a particularly preferred variant, the process according to the invention is thus notable 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).

The cut in the separation stage of retentate separation stage (2), at a concentration of component A or of a permeate gas A of 50% in the first retentate stream (7), is between 10 and 60%, preferably between 20 and 50%. The permeate of the retentate separation stage (2) is recycled by means of the second permeate stream (9), supplied to the feed stream (5) and reprocessed. This can, as already explained above in the definition of the term "feed stream", be effected in different ways according to whether a compressor (4) or even a multistage compressor (4) is used. In the case of a one-stage compressor (4), the second permeate stream (9) is preferably supplied to the suction side of the compressor (4) (see fig. 11). If a multistage compressor is used, it is preferable that the second permeate stream (9) is introduced into the compressor between two compression stages.

The permeate of the feed stream separation stage (1) which has been greatly enriched with component A or a permeate gas A is supplied by means of the first permeate stream (6) to the permeate separation stage (3). If necessary, it is possible by means of a pressure-reducing valve (14) in the retentate stream of the permeate separation stage (3), i.e. the third retentate stream (10), to prevent the pressure of the permeate of the feed stream separation stage (1) from declining to ambient pressure (see fig. 11). In this way, the driving force for the permeate separation stage (3) can be maintained. The permeate separation stage (3) produces a permeate with a content of component A or of a permeate gas A of greater than 95%, preferably greater than 97% and more preferably greater than 99%, which is discharged from the apparatus via the third permeate stream (11). In a particularly preferred embodiment, the apparatus is configured such 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).

The cut in the separation stage of the permeate separation stage (3) is between 50 and 95%, preferably between 70 and 93%.

The third retentate stream (10) is recycled, supplied to the feed stream (5) and reprocessed. This can, as already explained above, be effected in different ways and may depend, for example, on whether a compressor (4) or even a multistage compressor (4) is used. In the case of a one-stage compressor (4), the third retentate stream (10) is preferably supplied to the suction side of the compressor (4) (see fig. 11). If a multistage compressor is used, it is preferable that the third retentate stream (10) is introduced into the compressor between two compression stages.

In a preferred embodiment of the apparatus used in accordance with the invention, the retentate from the permeate separation stage (3) is recycled without total decompression into an elevated compression stage of the compressor (4).

In a further preferred embodiment of the apparatus used in accordance with the invention, the retentate from the permeate separation stage (3) without total decompression and the permeate from the second separation stage are recycled into an elevated compression stage of the compressor (4).

The process according to the invention is notable particularly in that it/it 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, preferably 10 to 50% by volume, most preferably 20 to 40% by volume, of the volume of the crude gas stream (17). The control of the amount of the retentate gas streams to be recycled can be controlled, for example, through selection of the respective membrane modules in the membrane separation stages (1) to (3) or by the pressures in the system or by the flows. The process according to the invention is thus notable in that, in spite of very low return streams, the increase in the concentration of the permeate component in the feed stream (5), which has been explained in detail above, is ensured. This distinctly increases the efficiency of the overall process.

The first permeate stream (6) is preferably conducted such that the feed pressure of the permeate separation stage (3), preferably by means of a pressure-reducing valve (14) on the retentate side of the permeate separation stage (3), is between 1 and 30 bar, preferably between 2 and 20 bar and more preferably 3 and 10 bar.

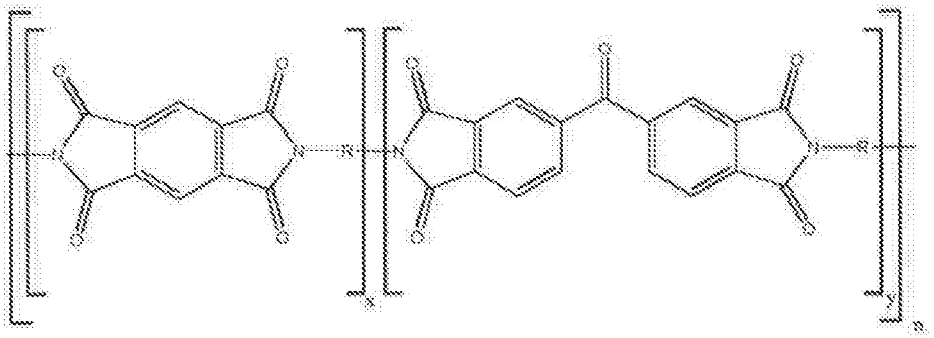
As already explained, it is particularly advantageous when a multistage compressor (4) is used. This is because it is possible in this case to dispense with a complete decompression of the retentate of the permeate separation stage (3), since the retentate of the permeate separation stage (3) can be fed in between two compressor stages of the compressor (4).

Since the retentate separation stage (2), in the case of decompression to feed pressure, would generally be operated in the selectivity-limited range, it may be advisable to decompress the second permeate stream (9) merely to a higher pressure level of a multistage pressure-increasing unit, i.e. of a multistage compressor (4), since this reduces the operating costs for the compression unit without distinctly worsening the separation outcome. In a particularly preferred embodiment of the present invention, therefore, a multistage compressor (4) is used and the gas streams (9) and (10) are supplied to this compressor in each case between two compression stages. Such a connection arrangement is shown in fig. 12.

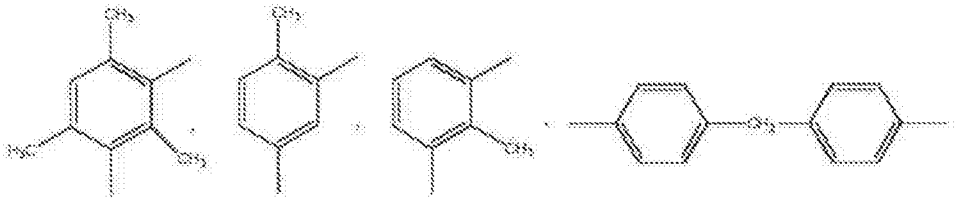
As already mentioned, the apparatus may comprise one or more pressure-reducing valves (12), (13) or (14). In a preferred embodiment, preferably by means of a pressure-reducing valve (14), it is ensured that the pressure drop over the feed stream separation stage (1) is restricted to 1 and 30 bar, preferably between 2 and 20 bar and more preferably between 3 and 10 bar. Simultaneously or alternatively, preferably by means of a pressure-reducing valve (13), it is ensured that the pressure drop over the feed stream separation stage (1) and the retentate separation stage (2), is restricted to 1 and 100 bar, preferably between 5 and 80 bar and more preferably between 10 and 70 bar.

The process according to the invention can in principle be implemented with all membranes which are capable of separating binary gas mixtures or multigas mixtures. The membrane materials used are preferably but not exclusively polymers. Useful polymers in the separation-active layer are more preferably polyimides, polyamides, polysulfones, cellulose acetates and derivatives, polyphenylene oxides, polysiloxanes, polymers with intrinsic microporosity, mixed matrix membranes, facilitated transport membranes, polyethylene oxides, polypropylene oxides, carbon membranes or zeolites, or mixtures thereof.

Particularly preferred membranes comprise, as materials for the separation-active layer or as a material for the complete membrane, a polyimide of the general formula



R is selected from the group consisting of



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

Such membranes are obtainable from Evonik Fibres GmbH under the Polyimid P84 and Polyimid P84 HT names. A process for producing these preferred membranes is disclosed in WO 2011/009919 A1. All membranes disclosed in this publication can be used with preference in the process according to the invention. To avoid straightforward repetition, reference is hereby made completely to the content of this patent application. It has been found that these membranes can achieve the best separation outcomes.

The membranes are preferably used in the form of hollow fiber membranes and/or flat membranes. The membranes are assembled into modules, which are then used in the separation task. The modules used may be all gas separation modules known in the prior art, for example but not exclusively hollow fiber gas separation modules, spiral-wound gas separation modules, cushion gas separation modules or tube bundle gas separation modules.

According to the invention, the gas separation membrane modules have a mixed gas selectivity of components A and B (= ratio of stream A to stream B through the membrane) of at least 30, preferably at least 35, more preferably at least 40, even more preferably of at least 45 and especially preferably of at least 45 to 80. Membranes of higher selectivity have the advantage that the separation becomes more effective and less permeate has to be recycled from retentate separation stage (2), or less retentate from permeate separation stage (3). Thus, especially in the case of use of a one-stage compressor (4), it is necessary to compress less gas twice, which entails economic advantages in the operation of the plant. In the case of very selective membrane modules having a selectivity of 45, only approx. 35% of the gas introduced into the feed stream separation stage (1) as crude gas need be compressed twice; with a membrane module having a selectivity of only 10, it may be the case that the double compression is up to 300%. The figures of 35% and 300% are based on experiments in which a gas mixture with equimolar amounts of components A and B (= feed) was applied, with 98.5% component B present in the retentate gas of stage (2) and 99% of component B in the permeate stream of stage (3).

It is evident that the inventive operation can be conducted in a much more economically viable manner with more selective membranes, and the necessary size of the compressor and the energy required can be reduced.

The process according to the invention has the particular advantages that it is a pure membrane process and does not need any additional purification of the permeate and/or retentate streams (11) or (8) for many applications. For example, in the case of purification of biogas or natural gas (= removal of carbon dioxide from methane), there is no longer any need for a pressure swing adsorption or amine scrubbing for fine purification of the retentate, and so it can be fed into the natural gas grid.

In addition, with the process according to the invention, it is possible at the same time to produce a pure retentate stream (8) and a pure permeate stream (11) in the biogas and natural gas purification. It can therefore be released into the atmosphere without any great losses of methane and without any great impairment of the environment, without any need for further treatment of the gas by a catalytic postcombustion or utilization in a combined heat and power plant. There is therefore no need for investment in further plant parts, which leads to a more economically viable purification process for biogas and natural gas.

A further advantage is considered to be that the process according to the invention needs a much lower level of apparatus complexity and energy expenditure than the known prior art processes.

Especially through the combination of the inventive features of mixed gas selectivity, control of the amount of the retentate streams recycled and increase in the permeate component in the feed stream (5), a process much superior to the prior art processes can be provided.

The process according to the invention can especially be used for separation of gas mixtures comprising at least two gases, the gas mixture separated most preferably being 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 crude biogas or crude natural gas.

The examples which follow are intended to illustrate and describe the present invention in detail, but do not restrict it in any way.

In the examples which follow, it was shown that, in the case of inventive use of the connection arrangement of the modules and application of particular pressures, it was possible to simultaneously produce methane with a purity of more than 96% and carbon dioxide with a purity of more than 97% from a mixture of carbon dioxide and methane in a ratio of 50 to 50.

Comparative example 1: Separation of a mixture of methane and carbon dioxide with a mixing ratio of 50 to 50 with a moderately selective polyimide membrane

The fig. 11 shown connection arrangement was used. Each stage consisted of a hollow fiber membrane module consisting of hollow polyimide fibers from UBE (NM B01 A type). 1.78 m^3/h of a crude gas mixture of 50% methane and 50% carbon dioxide, which corresponds approximately to a gas mixture of biogas, is introduced into a mixing chamber (not shown in fig. 11) and then compressed to 25 bar together with recycled gas composed of gas streams (9) and (10). The gas which has been compressed and cooled to 20°C is applied to the feed stream separation stage (1). The retentate of the feed stream separation stage (1) is subsequently passed via the first retentate stream (7) to the retentate separation stage (2). A reducing valve (13) on the retentate side of the retentate separation stage (2) is set to 18.2 bara and thus determines the driving force through the membrane of membrane separation stages (1) and (2). The retentate of the retentate separation stage (2) has a content of 98.5% methane and 1.5% carbon dioxide. 0.895 m^3/h of this mixture leaves retentate separation stage (2). The permeate of retentate separation stage (2) has a volume flow rate of 0.743 m^3/h with a methane content of 34.5% and a carbon dioxide content of 65.5%, and is recycled via the second permeate stream (9) into the mixing chamber and compressed again by the compressor (4).

The permeate of the feed stream separation stage (1) has a volume flow rate of 1.67 m^3/h with a carbon dioxide content of 91.0% and a methane content of 9.0%, and is passed via the first permeate stream (6) as feed into the permeate separation stage (3). The pressure drop over the membrane of stage (1) does not extend to ambient pressure, but is limited to 4.2 bara by a reducing valve (14) on the retentate side of the permeate separation stage (3). This gives a third permeate stream (11) from permeate separation stage (3) of 0.885 m^3/h with a composition of 99.0% carbon dioxide and only 1.0% methane. The third retentate stream (10) from the permeate separation stage (3) is 0.801 m^3/h with a composition of 17.9% methane and 82.1% carbon dioxide and is recycled into the mixing chamber and compressed again. The sum of recycled gas streams (9) and (10) is therefore 1.544 m^3/h or 86.7% based on the amount of gas to be separated supplied. Pure product streams are obtained with a relatively high double compression rate. The membranes used exhibit a moderate mixed gas selectivity for carbon dioxide over methane of 20.

Example 1: Separation of a mixture of methane and carbon dioxide with a mixing ratio of 50 to 50 with a highly selective polyimide membrane

The fig. 11 shown connection arrangement was used. Each stage consisted of a hollow fiber membrane module consisting of highly selective hollow polyimide fibers with a separation area of about 5 m^2 per module. These hollow polyimide fibers were produced according to example 19 of Austrian patent application A1164/2009, except working with a precipitation bath temperature of 40°C rather than 10°C. 1 m^3/h of a crude gas mixture of 50% methane and 50% carbon dioxide, which corresponds approximately to a gas mixture of biogas, is introduced into a mixing chamber and then compressed to 25 bar together with recycled gas composed of gas streams (9) and (10). The gas which has been compressed and cooled to 20°C is applied to the feed stream separation stage (1). The retentate of this stage is passed via the first retentate stream (7) to the retentate separation stage (2). A reducing valve (13) on the retentate side of the retentate separation stage (2) is set to 18.4 bara and thus determines the driving force through the membrane of membrane separation stages (1) and (2). The retentate of the retentate separation stage (2) has a content of 98.5% methane and 1.5% carbon dioxide. 0.503 m^3/h of this mixture leaves retentate separation stage (2). The permeate of retentate separation stage (2) has a volume flow rate of 0.262 m^3/h with a methane content of 24.6% and a carbon dioxide content of 75.4%, and is recycled via the second permeate stream (9) into the mixing chamber and compressed again by the compressor (4).

The permeate of the feed stream separation stage (1) has a volume flow rate of 0.547 m^3/h with a carbon dioxide content of 92.4% and a methane content of 7.6%, and is passed via the first permeate stream (6) as feed into the permeate separation stage (3). The pressure drop over the membrane of stage (1) does not extend to ambient pressure, but is limited to 5.0 bara by a reducing valve (14) on the retentate side of the permeate separation stage (3). This gives a third permeate stream (11) from permeate separation stage (3) of 0.497 m^3/h with a composition of 99.0% carbon dioxide and only 1.0% methane. The third retentate stream (10) from the permeate separation stage (3) is 0.050 m^3/h. The sum of recycled gas streams (9) and (10) is therefore 0.312 m^3/h or 31.2% based on the amount of gas to be separated supplied. Pure product streams are obtained with a moderate double compression rate. The membranes used exhibit a high mixed gas selectivity for carbon dioxide over methane of 45. The high selectivity of 45 compared to 20 in the comparative example therefore enables a more economically favorable process by a lower recompression of 31.2% rather than 86.7%.

Example 2: Separation of a mixture of methane and carbon dioxide with a mixing ratio of 50 to 50 with a highly selective polyimide membrane with a methane content in the permeate stream of less than 0.5%

In order to satisfy various legislators, it is necessary to minimize the methane loss into the atmosphere. The fig. 11 shown connection arrangement was used to lower the concentration of methane in the third permeate stream (11) to below 0.5% by volume. Each stage consisted of a hollow fiber membrane module consisting of highly selective polyimide with a separation area of about 5 m^2 per module. These hollow polyimide fibers were produced according to example 19 of Austrian patent application A1164/2009, except working with a precipitation bath temperature of 40°C rather than 10°C. 1 m^3/h of a crude gas mixture of 50% methane and 50% carbon dioxide, which corresponds approximately to a gas mixture of biogas, is introduced into a mixing chamber and then compressed to 25 bar together with recycled gas composed of gas streams (9) and (10). The gas which has been compressed and cooled to 20°C is applied to the feed stream separation stage (1). The retentate of this stage is passed via the first retentate stream (7) to the retentate separation stage (2). A reducing valve (13) on the retentate side of the retentate separation stage (2) is set to 18.1 bara and thus determines the driving force through the membrane of membrane separation stages (1) and (2). The retentate of the retentate separation stage (2) has a content of 98.5% methane and 1.5% carbon dioxide. 0.505 m^3/h of this mixtures leaves retentate separation stage (2). The permeate of retentate separation stage (2) has a volume flow rate of 0.244 m^3/h with a methane content of 26.1% and a carbon dioxide content of 73.9%, and is recycled via the second permeate stream (9) into the mixing chamber and compressed again by the compressor (4).

The permeate of the feed stream separation stage (1) has a volume flow rate of 0.607 m^3/h with a carbon dioxide content of 93.1% and a methane content of 6.9%, and is passed via the first permeate stream (6) as feed into the permeate separation stage (3). The pressure drop over the membrane of stage (1) does not extend to ambient pressure, but is limited to 4.4 bara by a reducing valve (14) on the retentate side of the permeate separation stage (3). This gives a third permeate stream (11) from permeate separation stage (3) of 0.495 m^3/h with a composition of 99.5% carbon dioxide and only 0.5% methane. The third retentate stream (10) from the permeate separation stage (3) is 0.112 m^3/h and has a composition of 35% methane and 65% carbon dioxide and is recycled into the mixing chamber and compressed again. The sum of recycled streams (9) and (10) is therefore 0.356 m^3/h or 35.6% based on the amount of gas to be separated supplied. Pure product streams are obtained with a moderate double compression rate. The membranes used exhibit a high mixed gas selectivity for carbon dioxide over methane of 45.

Example 3: Separation of a mixture of methane and carbon dioxide with a mixing ratio of 50 to 50 with a highly selective polyimide membrane with a methane content in the permeate stream of less than 0.5% with the aid of a vacuum pump for the permeate of stage 2

In order to satisfy various legislators, it is necessary to minimize the methane loss into the atmosphere. The fig. 11 shown connection arrangement, supplemented with a vacuum pump not shown in fig. 11, was used to lower the concentration of methane in the third permeate stream (11) to below 0.5% by volume. Each stage consisted of a hollow fiber membrane module consisting of highly selective hollow polyimide fibers with a separation area of about 5 m^2 per module. These hollow polyimide fibers were produced according to example 19 of Austrian patent application A1164/2009, except working with a precipitation bath temperature of 40°C rather than 10°C. 1 m^3/h of a crude gas mixture of 50% methane and 50% carbon dioxide, which corresponds approximately to a gas mixture of biogas, is introduced into a mixing chamber and then compressed to 25 bar together with recycled gas composed of gas streams (9) and (10). The gas which has been compressed and cooled to 20°C is applied to the feed stream separation stage (1). The retentate of this stage is passed via the first retentate stream (7) to the retentate separation stage (2). A reducing valve (13) on the retentate side of the retentate separation stage (2) is set to 14.5 bara and thus determines the driving force through the membrane of membrane separation stages (1) and (2). The retentate of the retentate separation stage (2) has a content of 98.5% methane and 1.5% carbon dioxide. 0.505 m^3/h of this mixture leaves retentate separation stage (2). The permeate of retentate separation stage (2) has a pressure of 0.2 bara, established by a vacuum pump, and has a volume flow rate of 0.371 m^3/h with a methane content of 13.3% and a carbon dioxide content of 86.7%. The gas stream is recycled from the pressure side of the vacuum pump via the second permeate stream (9) into the mixing chamber and compressed again by the compressor (4).

The permeate of the feed stream separation stage (1) has a volume flow rate of 0.542 m^3/h with a carbon dioxide content of 94.8% and a methane content of 5.2%, and is passed via the first permeate stream (6) as feed into the permeate separation stage (3). The pressure drop over the membrane of stage (1) does not extend to ambient pressure, but is limited to 4.4 bara by a reducing valve (14) on the retentate side of the permeate separation stage (3). This gives a third permeate stream (11) from permeate separation stage (3) of 0.495 m^3/h with a composition of 99.5% carbon dioxide and only 0.5% methane. The third retentate stream (10) from the permeate separation stage (3) is 0.047 m^3/h and has a composition of 54.9% methane and 45.1% carbon dioxide and is recycled into the mixing chamber and compressed again. The sum of recycled streams (9) and (10) is therefore 0.417 m^3/h or 41.7% based on the amount of gas to be separated supplied. Pure product streams are obtained with a moderate double compression rate. The membranes used exhibit a high mixed gas selectivity for carbon dioxide over methane of 45. In contrast to the above example 2, less pressure is required (14.5 bar rather than 18.1 bar = 80.1% of the pressure), and only 6.1% more double compression, which leads to a saving in compression work.

Example 4: Separation of a mixture of methane and carbon dioxide with a mixing ratio of 50 to 50 with a highly selective polyimide membrane with a methane content in the permeate stream of less than 0.5% with the aid of a vacuum pump for the permeate of stage 2 and improved retentate purity

In order to satisfy various legislators, it is necessary to minimize the methane loss into the atmosphere.

The fig. 11 shown connection arrangement, supplemented with a vacuum pump not shown in fig. 11, was used to increase the concentration of methane in the second retentate stream (8). The vacuum improves the pressure ratio in the retentate separation stage (2) by reducing the pressure in the second permeate stream (9), such that the driving force is increased and higher purities can be achieved in the second retentate stream (8).

Each stage consisted of a with a separation area of about 5 m^2 per module. These hollow polyimide fibers were produced according to example 19 of Ausarian patent application A1164/2009, except working with a precipitation bath temperature of 40°C rather than 10°C. 1 m^3/h of a crude gas mixture of 50% methane and 50% carbon dioxide, which corresponds approximately to a gas mixture of biogas, is introduced into a mixing chamber and then compressed to 25 bar together with recycled gas composed of gas streams (9) and (10). The gas which has been compressed and cooled to 20°C is applied to the feed stream separation stage (1). The retentate of this stage is passed via the first retentate stream (7) to the retentate separation stage (2). A reducing valve (13) on the retentate side of the retentate separation stage (2) is set to 18.1 bara and thus determines the driving force through the membrane of membrane separation stages (1) and (2). The retentate of the retentate separation stage (2) has a content of 99.7% methane and 0.3% carbon dioxide. 0.499 m^3/h of this mixture leaves retentate separation stage (2). The permeate of retentate separation stage (2) has a pressure of 0.2 bara, established by a vacuum pump, and has a volume flow rate of 0.258 m^3/h with a methane content of 25.8% and a carbon dioxide content of 74.2%. The gas stream is recycled from the pressure side of the vacuum pump via the second permeate stream (9) into the mixing chamber and compressed again by the compressor (4).

The permeate of the feed stream separation stage (1) has a volume flow rate of 0.608 m^3/h with a carbon dioxide content of 93.2% and a methane content of 6.8%, and is passed via the first permeate stream (6) as feed into the permeate separation stage (3). The pressure drop over the membrane of stage (1) does not extend to ambient pressure, but is limited to 4.4 bara by a reducing valve (14) on the retentate side of the permeate separation stage (3). This gives a third permeate stream (11) from permeate separation stage (3) of 0.501 m^3/h with a composition of 99.5% carbon dioxide and only 0.5% methane. The third retentate stream (10) from the permeate separation stage (3) is 0.107 m^3/h and has a composition of 36.2% methane and 63.8% carbon dioxide and is recycled into the mixing chamber and compressed again. The sum of recycled streams (9) and (10) is therefore 0.366 m^3/h or 36.6% based on the amount of gas to be separated supplied. Pure product streams are obtained with a moderate double compression rate. The membranes used exhibit a high mixed gas selectivity for carbon dioxide over methane of 45. In contrast to the above example 3, use of a vacuum pump on the permeate side of the retentate separation stage (2) gives a much cleaner retentate with the same compressor output (99.7% methane rather than 98.5%).

List of figures:

Fig. 6:	Staged arrangement for retentate and permeate with recompression and recycling in the second retentate stage upstream of the second retentate stage
Fig. 7:	Staged arrangement for permeate with recompression
Fig. 8:	Staged arrangement for retentate with permeate recycling from the second stage and staged arrangement for permeate with recompression
Fig. 9:	Staged arrangement for retentate with permeate recycling from the second stage
Fig. 10:	Connection arrangement with internal staged arrangement for permeate
Fig. 11:	Illustrative connection arrangement of several membrane modules according to invention

List of reference numerals:

1: Feed stream separation stage
2: Retentate separation stage
3: Permeate separation stage 3
4: One-stage or multistage compressor
5: Feed stream
6: First permeate stream
7: First retentate stream
8: Second retentate stream
9: Second permeate stream
10: Third retentate stream
11: Third permeate stream
12: Optional pressure-reducing valve in the first retentate stream 7
13: Optional pressure-reducing valve in the second retentate stream 8
14: Optional pressure-reducing valve in the third retentate stream 10
17: Crude gas stream

Eljárás gázok szétválasztására

Szabadalmi igénypontok

1. Eljárás egy nyersgáz áram (17) szétválasztására, amely eljárást egy olyan berendezésben valósitjuk meg, amely tartalmaz egy betáplálásiáram-szétválasztó fokozatot (1), egy retentátum-szétválasztó fokozatot (2) és egy permeátum-szétválasztó fokozatot (3), minden esetben membrános szétválasztó fokozatokként, valamint egy kompresszort (4) és/vagy legalább egy, előnyösen egy vagy két vákuumszivattyút, ahol
a betáplálásiáram-szétválasztó fokozatban (1) egy betáplálási áramot (5), amely legalább két, A és B komponensből áll, ahol A a könnyebben permeálódó komponens, egy első permeátum áramra (6) és egy első retentátum áramra (7) osztunk fel,

a retentátum-szétválasztó fokozatban (2) az első retentátum áramot (7) egy a nyersgáz áramhoz (17) hozzávezetett második permeátum áramra (9) és egy második retentátum áramra (8) osztjuk fel, mely utóbbit mint első, B komponensben feldúsított terméket veszünk el,

a permeátum-szétválasztó fokozatban (3) az első permeátum áramot (6) egy a nyersgáz áramhoz (17) hozzávezetett harmadik retentátum áramra (10) és egy harmadik permeátum áramra (11) osztjuk fel, mely utóbbit mint második, A komponensben feldúsított terméket veszünk el,

az első permeátum áramot (6) nem vetjük alá ismételt sűrítésnek,

a második permeátum áramban (9) és a harmadik permeátum áramban (10) visszavezetett gáztérfogat összegében kevesebb, mint a nyersgáz áram (17) térfogatának 60 térfogatszázaléka,

az A komponens koncentrációját a második permeátum áram (9) és a harmadik retentátum áram (10) visszavezetése után megnöveljük a betáplálási áramban (5), előnyösen legalább 2 %-kal, különösen előnyösen legalább 3 %-kal és még előnyösebben 3-40 %-kal, minden esetben annak a nyersgáz áramban (17) fennálló koncentrációjához képest, és

legalább a betáplálásiáram-szétválasztó fokozatban (1), előnyösen azonban mindhárom membrános szétválasztó fokozatban (1-3) olyan gázszétválasztó membránmodulokat alkalmazunk, amelyek kevertgáz-szétválasztó képessége az A és B komponensek tekintetében legalább 30.

2. Az 1. igénypont szerinti eljárás, ***azzal jellemezve***, hogy a második permeátum áramot (9) és a harmadik retentátum áramot (10) újrafeldolgozáshoz a kompresszor (4) szívóoldalára vezetjük.

3. Az 1. vagy 2. igénypont szerinti eljárás, ***azzal jellemezve***, hogy egy többlépcsős kompresszort (4) alkalmazunk.

4. A 3. igénypont szerinti eljárás, ***azzal jellemezve***, hogy a második permeátum áramot (9) és/vagy a harmadik retentátum áramot (10) két sűrítési fokozat közé vezetjük be a kompresszorba (4).

5. Az 1-4. igénypontok bármelyike szerinti eljárás, ***azzal jellemezve***, hogy az első retentátum áramot (7) és/vagy a második retentátum áramot (8) és/vagy a harmadik retentátum áramot (10) egy nyomáscsökkentő szelepen keresztül vezetjük.

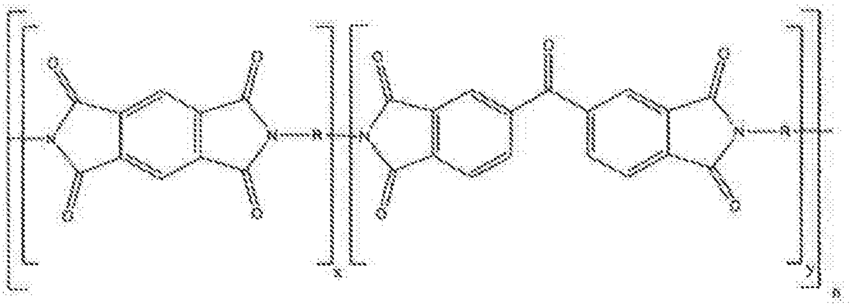
6. Az 1-5. igénypontok bármelyike szerinti eljárás, ***azzal jellemezve***, hogy a membrános szétválasztó fokozatok (1-3) legalább egyikében olyan gázszétválasztó membránmodult alkalmazunk, amelynek kevertgáz-szétválasztó képessége az A és B komponensek tekintetében legalább 35, előnyösen legalább 40, különösen előnyösen legalább 45.

7. Az 1-6. igénypontok bármelyike szerinti eljárás, ***azzal jellemezve***, hogy a membrános szétválasztó fokozatok (1-3) legalább egyike több mint egy gázszétválasztó membránmodult tartalmaz, amelyek párhuzamosan vagy sorosan vannak kapcsolva.

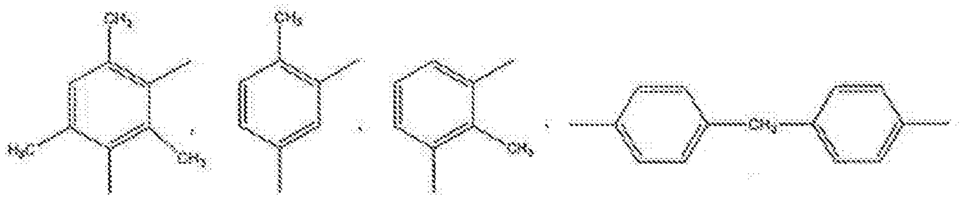
8. Az 1-7. igénypontok bármelyike szerinti eljárás, ***azzal jellemezve***, hogy az egy vagy több gázszétválasztó membránmodul üreges szálú membránokból és/vagy lapos membránokból áll.

9. Az 1-8. igénypontok bármelyike szerinti eljárás, ***azzal jellemezve***, hogy a membránok szétválasztást elősegítő rétegének anyagaként amorf vagy részben kristályos műanyagokat alkalmazunk, mint például, de nem kizárólagosan, poliimideket, poliamidokat, poliszulfonokat, cellulóz-acetátot és származékait, polifenilénoxidokat, polisziloxánokat, belső mikropórusos polimereket, kevert mátrixú membránokat (mixed matrix membranes), könnyített áteresztésű membránokat (facilitated transport membranes), polietilén-oxidokat, polipropilén-oxidokat vagy ezek keverékeit.

10. A 9. igénypont szerinti eljárás, ***azzal jellemezve***, hogy a membránok szétválasztási elősegítő rétegének anyagaként az alábbi általános képletű poliimidet alkalmazzuk:



ahol a képletben R az alábbiakból álló csoportból van megválasztva:



és x,y: móltört,

$0 < x < 0,5$ és $1 > y > 0,5$ értékekkel.

11. Az 1-10. igénypontok bármelyike szerinti eljárás, ***azzal jellemezve***, hogy az A és B komponensek szén-dioxid és metán, vagy hidrogén és metán, vagy szén-monoxid és hidrogén.

12. Az 1-11. igénypontok bármelyike szerinti eljárás, ***azzal jellemezve***, hogy gázkeverékként nyers biogázt vagy nyers földgázt vagy egy olyan keveréket használunk, amely főként, de nem kizárólagosan az A és B komponenseket tartalmazza.

13. Az 1-12. igénypontok bármelyike szerinti eljárás, ***azzal jellemezve***, hogy a betáplálásiáram-szétválasztó fokozat (1) permeátum oldalának (6) nyomását előnyösen egy a permeátum-szétválasztó fokozat (3) retentátum oldalán elhelyezett nyomáscsökkentő szelep (14) segítségével 1-30 bar-ra, előnyösen 2-20 bar-ra, és különösen előnyösen 3-10 bar-ra állítjuk be.

14. Az 1-13. igénypontok bármelyike szerinti eljárás, ***azzal jellemezve***, hogy az első és a második retentátum áram (7 és 8) nyomását előnyösen egy a második retentátum áramban (8) elhelyezett nyomáscsökkentő szelep (13) segítségével 1-100 bar-ra, előnyösen 5-80 bar-ra és különösen előnyösen 10-70 bar-ra állítjuk be.

15. Az 1-14. igénypontok bármelyike szerinti eljárás, ***azzal jellemezve***, hogy a szétválasztási feladathoz hajtóerőként a retentátum oldal és a permeátum oldal közötti parciálisnyomás-különbséget használjuk fel a mindenkori membrános szétválasztó fokozatokban, ahol a parciálisnyomás-különbséget egy a betáplálási áramban (5) elrendezett kompresszorral és/vagy a második és/vagy a harmadik permeátum áramban (9, 10) elrendezett vákuumszivattyúval (15) és/vagy egy permeátum oldali öblítőgáz árammal hozzuk létre.

16. Az 1-15. igénypontok bármelyike szerinti eljárás, ***azzal jellemezve***, hogy a betáplálásiáram-szétválasztó fokozat (1) permeátumának nyomása a környezeti nyomásával azonos vagy ahhoz képest megnövelt, így még egy parciálisnyomás-különbség áll fenn a permeátum-szétválasztó fokozat (3) retentátuma és permeátuma között, és ezzel adva van egy hajtóerő arra az esetre, ha a permeátum-szétválasztó fokozat (3) permeátuma környezeti nyomáson van, vagy vákuum hatásának van kitéve.

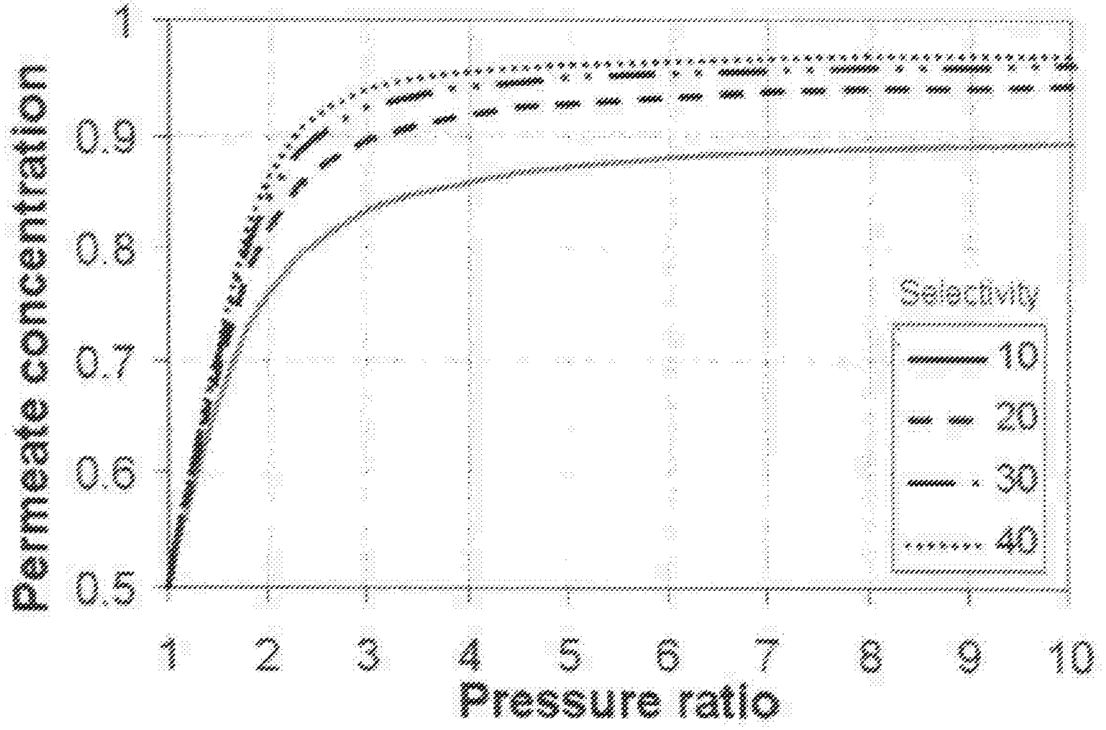


Fig. 1: Influence of the pressure ratio and of the selectivity on the separation outcome

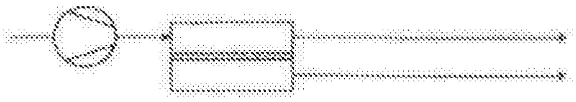


Fig. 2: One-stage membrane separation stage without recycling

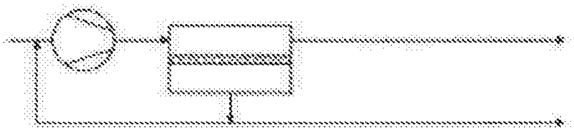


Fig. 3: One-stage membrane separation stage with recycling

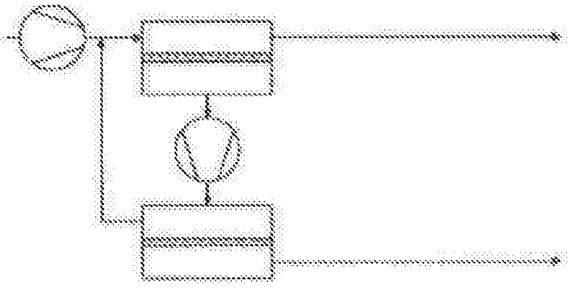


Fig. 4: Two-stage membrane separation stage with recompression and recycling

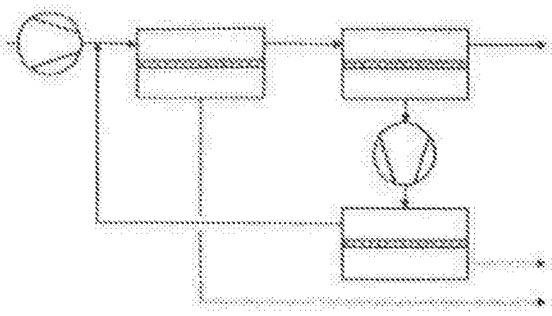


Fig. 5: Staged arrangement for retentate and permeate with recompression and recycling in the second retentate stage upstream of the first retentate stage

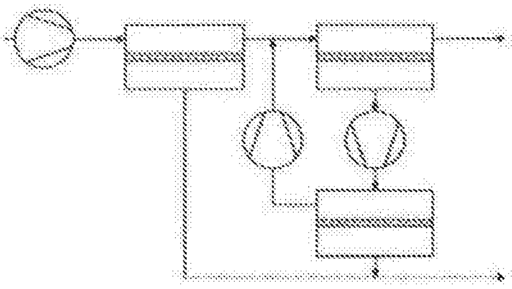


Fig. 6: Staged arrangement for retentate and permeate with recompression and recycling in the second retentate stage upstream of the second retentate stage

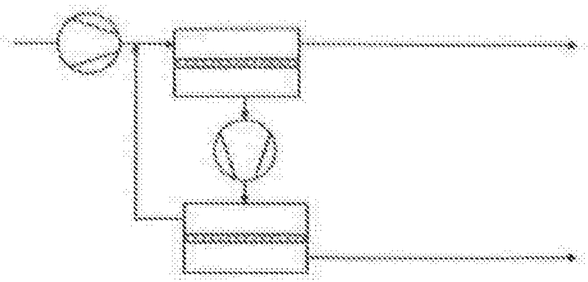


Fig. 7: Staged arrangement for permeate with recompression

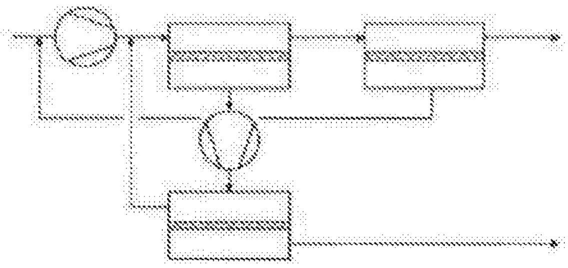


Fig. 8: Staged arrangement for retentate with permeate recycling from the second stage and staged arrangement for permeate with recompression

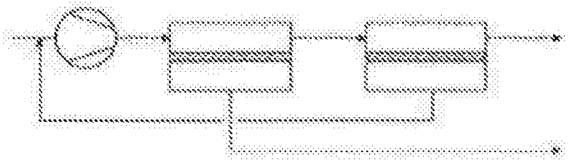


Fig. 9: Staged arrangement for retentate with permeate recycling from the second stage

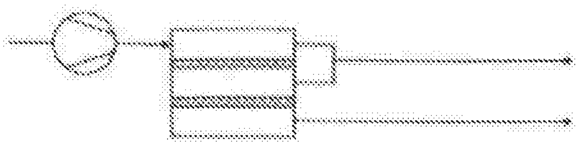


Fig. 10: Connection arrangement with internal staged arrangement for permeate

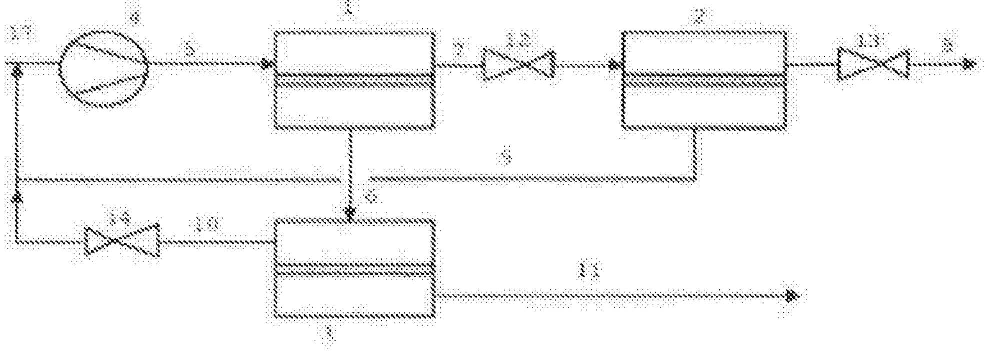


Fig. 11: Illustrative connection arrangement of several membrane modules according to invention