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Description

[0001] The present invention relates to a process for synthesizing a product gas, comprising the steps:

- 5 (a) provision of a first feed gas;
- (b) provision of a second feed gas;
- (c) combining of the first feed gas and the second feed gas to give a gas mixture A comprising first feed gas and second feed gas;
- (d) combining of the gas mixture A with a circulated gas mixture to give a
- 10 gas mixture B comprising first feed gas, second feed gas and product gas;
- (e) reaction of the first feed gas and second feed gas present in the gas mixture B over a catalyst to give a product gas with production of a gas mixture C enriched in product gas;
- (f) removal of product gas from the gas mixture C enriched in product gas
- 15 with production of a gas mixture D depleted in product gas; and
- (g) recirculation of at least part of the gas mixture D depleted in product gas as circulated gas mixture to step (d);

- where the volume flow of the first feed gas provided in step (a) and/or the volume flow of the second feed gas provided in step (b) and/or the volume flow of the
- 20 product gas removed in step (f) is varied during the process, further comprising
- the temporary reduction of the amount of product gas formed per unit time during a first time interval by reduction of the first feed gas and/or the volume of flow of the second feed gas provided in step (b) and/or the volume of flow of the product gas removed in step (f); and
 - 25 - increasing of the amount of product gas formed per unit time during a second time interval by increasing the volume flow of the first feed gas provided in step (a) and/or the volume flow of the second feed gas provided in step (b) and/or the volume flow of the product gas removed in step (f).

- 30 **[0002]** The present invention further provides a plant for producing a product gas from a first feed gas and a second feed gas, comprising the following components which are operatively connected to one another:

- a first inlet device configured for providing the first feed gas;

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- a second inlet device configured for providing the second feed gas;
- a reactor configured for synthesizing a product gas from the first feed gas and the second feed gas to produce a gas mixture C enriched in product gas under heterogeneous gas catalysis;
- 5 - a removal device configured for removing product gas from the gas mixture C enriched in product gas with production of a gas mixture D depleted in product gas; and
- a control device which is configured for bringing the apparatus into a first operating state or into a second operating state, where the first operating
- 10 state differs from the second operating state
 - (i) in terms of a smaller volume flow of the first feed gas which is provided by the first inlet device and/or a small volume flow of the second feed gas which is provided by the second inlet device and/or a smaller volume flow of the product gas which is removed by the removal device,
 - 15 and/or
 - (ii) in terms of a smaller amount of product gas formed per unit time.

[0003] The process according to the present invention provides for product gas to be removed during the synthesis of a product gas from a first feed gas and a second

20 feed gas and unreacted feed gas to be circulated, with the volume flow of the first feed gas, of the second feed gas and/or of the removed product gas being varied during the process. In this way, the amount of product gas formed per unit time can be temporarily throttled back (part load operation) or the synthesis thereof be temporarily stopped entirely (standby operation) without the plant in which the

25 process is carried out having to be shut down completely. In this way, temporary bottlenecks in the availability of energy or of feed gas can be bridged. The process can then briefly be run up to optimized synthesis conditions (full load operation) again when the availability of energy or of feed gas has been established again. The two feed gases can, for example, be hydrogen and nitrogen and the product gas can

30 be ammonia.

[0004] The proportion of renewable energies in the supply of electric power is continually increasing. In Germany, the proportion at the end of 2012 was already

about 23% and is intended to increase further up to 35% by the year 2020. In addition, it is expected worldwide that the proportion of renewable energies in total electric power generation will increase significantly. Here, a particularly high growth is expected particularly for electric power generation from wind and solar power.

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[0005] However, these forms of energy present the challenge of being subject to great fluctuations over time (both hourly and seasonally) and their introduction prognosis being subject to errors. The demand for electricity, on the other hand, is subject to significantly different fluctuations and likewise prognosis errors. Times
10 having the highest consumption frequently do not coincide with times of greatest electric power generation from renewable energies.

[0006] Apart from this highly temporary generation, electric power is also generated from wind and solar power to a very different extent regionally since these energy
15 sources depend on regional circumstances such as wind conditions and solar radiation. Highly intermittent electric power generation leads firstly to great price fluctuations on the power market and secondly the requirements which conventional power stations have to meet in order to equalize the temporary fluctuations and by power grids to equalize the regional differences are becoming ever greater. Large
20 regional differences in electric power generation lead to a greater stress and capacity requirement imposed on the power grids in the long term, so that these have to be configured accordingly.

[0007] In order to counter these challenges, power stores and flexibilizing measures
25 on the supply and demand side will be required to an increased extent in the future in order to equalize fluctuations both in electric power generation and in the power price and take stress off the grids.

[0008] The production of ammonia from nitrogen and hydrogen is a large-scale,
30 catalytic process which has been established for many decades. Since it is an equilibrium reaction, only part of the nitrogen and hydrogen can be reacted in one pass through the reactor. After partial removal of the ammonia by condensation, the remaining, unreacted nitrogen and hydrogen are recirculated to the reactor, so that

the process is a circulation process. The ammonia is condensed and removed. Since the reaction is exothermic, the heat of reaction has to be removed.

[0009] In conventional processes for synthesizing ammonia, the reactors are usually
5 configured as adiabatic multibed reactors, i.e. a plurality of catalyst beds are
connected in series. A distinction is made here between reactors with direct cooling
(quench reactors) and reactors having indirect cooling in order to decrease the raised
gas temperature resulting from the exothermic reaction again. In quench reactors,
temperature regulation is achieved by cooling part of the gas mixture and feeding it
10 in again between downstream catalyst beds, by passing at least the first catalyst bed.
Compared to the preheated part of the gas mixture, which flows to the first catalyst
bed, the quench streams are significantly colder. In reactors with indirect cooling,
temperature regulation is achieved by a heat exchanger, by means of which the heat
of reaction can be removed via a cooling medium, being located between the catalyst
15 beds.

[0010] The conventional processes for synthesizing ammonia are designed for
continuous and steady-state operation. Part load operation of such plants is possible
to only a limited extent and always leads to significantly increasing specific energy
20 consumptions. Complete shutdown of the plants is always very complicated and
time-consuming. Furthermore, the extreme temperature and pressure fluctuations
would lead to a significant reduction in the durability of the apparatuses and
components used.

25 [0011] In order to produce NH_3 intermittently via the intermediate step of water
electrolysis, new technical concepts which allow large fluctuations in the load of the
plant are necessary. While the water electrolysis used for producing the hydrogen can
react highly flexibly to the intermittent availability of electricity from wind power
and solar energy, the synthesis of NH_3 from nitrogen and hydrogen is significantly
30 slower.

[0012] WO 2012/037571 A2 discloses a process in which a hydrogen store/buffer
between the electrolysis and the ammonia synthesis is utilized to equalize short-term

fluctuations in the production of hydrogen. Such a buffer can also be utilized to bridge short time periods in which no hydrogen can be produced.

5 [0013] In order not to have to design a hydrogen store of any size, which would lead to very high capital costs, it is necessary to have technical concepts which allow the ammonia synthesis to be carried out in part load operation without the plant being excessively stressed and without the specific consumptions being greatly increased.

10 [0014] In the production of hydrogen by electrolysis of water, there can every now and again be times at which it is not purposeful to produce hydrogen for several days or weeks. In order to equalize this, a very large buffer would be necessary but this will be fully utilized only rarely, so that the required capital investment would be disproportionate. In order to circumvent this, it is necessary to have technical concepts in order to run down the ammonia synthesis to a (material-saving) standby
15 mode and run it up again quickly.

[0015] These considerations apply in similar form to further products which are produced from synthesis gas, e.g. for the production of methanol.

20 [0016] EP 2 589 574 B1 discloses a process for regulating the load of an ammonia plant, in which the ammonia plant comprises a high-pressure ammonia synthesis circuit which comprises at least one ammonia reactor in which a fresh gas comprising a hydrogen feed and a nitrogen feed is converted at a synthesis pressure into ammonia with the circuit also comprising a purge conduit which is arranged so as to
25 draw off a purge gas containing inert gases from the circuit and the process also providing for part load operation of the plant. During this part load operation, the ammonia synthesis circuit is operated at the synthesis pressure and the purge gas stream is decreased, with the ratio of the molar flow of the purge gas to the molar flow of the fresh gas being reduced to a value which is smaller than a nominal value
30 of the ratio which is used when the ammonia plant is operated at nominal load. However, this concept has the disadvantage that in the case of small proportions of inert gas in the fresh gas there is only a very slow increase in the proportion of inert

gas in the synthesis circuit. For this reason, such a concept allows only a very slow lowering and increasing of the part load operation.

5 [0017] It is an object of the invention to provide an alternative process and an alternative plant for producing products by heterogeneous gas catalysis, which allow, in particular, a rapid change in load, i.e. a rapid change from full load operation to part load operation and vice versa.

10 [0018] This object is achieved by a process having the features of claim 1 and by a plant having the features of claim 13.

[0019] The variation of the volume flows as described preferably brings the process of the invention into various states in which different amounts of product gas are formed per unit time, and these are preferably referred to for the purposes of the invention as “*standby operation*”, “*part load operation*” and “*full load operation*”. As regards the amount of product gas formed per unit time, the states have the following ranking order: standby operation < part load operation < full load operation. In the process of the invention, preference is given to at least two of these states being gone through in succession. A person skilled in the art will know that, proceeding from full load operation, part load operation has to be conducted at least for a short time interval in order to finally bring the process into standby operation, and vice versa. The different amount of product gas formed per unit time can, according to the invention, be achieved by different measures and the combination thereof.

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[0020] The process of the invention is suitable for temporarily storing energy which is available in different amounts, e.g. electric energy from regenerative sources, in the form of chemical energy. For example, hydrogen can be dissociated by electrolysis into hydrogen and oxygen and the hydrogen obtained in this way can be converted back into electric power when required later. The temporary storage of hydrogen (first feed gas) can, according to the invention, be effected by reaction with nitrogen (second feed gas) to form ammonia (product gas). The ammonia obtained in this way can when required be redissociated into hydrogen and nitrogen or electric

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power can also be obtained back directly by combustion of ammonia. As an alternative, ammonia can also be sold as regeneratively produced product on the chemicals or fertilizer market.

5 **[0021]** The process of the invention makes it possible to transport varying loads in the production of feed gas, e.g. hydrogen, due to part load operation to the synthesis of product gas, e.g. ammonia. As a result of simultaneous combination with a buffer, e.g. a hydrogen buffer, such a system can react very quickly to changes in the availability of feed gas (e.g. H_2 and N_2) and bridge a certain time in which no feed
10 gas is produced via the buffer. In this way, the overall flexibility of the process increases with a simultaneously low buffer requirement.

[0022] A preferred further development of the process provides for the removal of the product gas in step (f) to be effected by condensing out by means of a condenser
15 and the outlet temperature of the condenser being increased in the first time interval. Higher product contents in the recycle stream are achieved by increasing the condensation temperature at part load. This leads to a shift in the reaction equilibrium and to a reduction in the partial pressures of the feed gases in the gas mixture.

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[0023] In the process of the invention, the abovementioned measure of increasing the outlet temperature of the condenser can in principle also be implemented as sole measure in a convertor having only one catalyst bed. However, this measure is preferably undertaken in combination with a change in the quench flows in part load
25 operation of the plant. According to simulation calculations, a decrease in the load down to 20% of the nominal load is possible by means of these measures (60% of the nominal load is usual).

[0024] At least partial bypassing of the catalyst makes a rapid change to standby
30 operation and back into normal operation possible without subjecting the convertor to great temperature change stresses.

[0025] The measures proposed in the present invention serve to set a suitable temperature profile in the reaction zone during operation of the plant in part load. This is advantageous in a plant for producing ammonia by reaction of nitrogen and hydrogen, but the present invention is not restricted to this reaction.

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[0026] According to the invention, a change in the volume flows of the feed gases and/or the discharged volume flow is carried out in part load operation of the plant, with it being particularly advantageous for the desired concentration change in the recycle stream to be achieved by a nonproportional change in the discharged volume flow relative to the feed streams.

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Figure 1 schematically illustrates a preferred variant of the apparatus of the invention for carrying out the process of the invention (circulation process), where in the interest of simplicity not all heat exchangers, in particular no heat integration between the process streams, are depicted.

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Figure 2 schematically illustrates a preferred variant of the apparatus of the invention for carrying out the process of the invention for standby operation for keeping the synthesis circuit warm.

Figure 3 schematically illustrates a variant of the embodiment of Figure 2, in which the separate circuit for standby operation is, however, smaller.

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Figure 4 shows the relative proportion of ammonia (product gas) in thermodynamic equilibrium as a function of the temperature at various pressures.

Figure 5 depicts a possible course of the amount of ammonia produced in the respective catalyst bed in the load range from 30% to 100% based on the total conversion in full load operation.

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Figure 6 shows the ratio of the relative proportion of ammonia (product gas) to temperature for a three-bed quench reactor (a) under full load and (b) under 30% part load. The solid-line graphs in each case relate to one of the three catalyst beds.

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Figure 7 illustrates a quench reactor in which the gas mixture can be conveyed between the outer shell of the reactor and the catalyst beds in standby operation.

[0027] The process of the invention is preferably characterized by at least two states which are passed through during at least two successive time intervals, preferably time intervals which follow one another in direct succession. For the purposes of the description, these two time intervals will hereinafter be referred to as “*first time interval*” and as “*second time interval*”. The first time interval can occur at a time before or after the second time interval. The state of the process during the first time interval is preferably characterized by a lower yield of product gas compared to the state of the process during the second time interval. The state of the process during the first time interval particularly preferably corresponds to part load operation while the state of the process during the second time interval corresponds to full load operation.

[0028] In a preferred further development of the invention, there is at least one third time interval in which the volume flow of the first feed gas provided in step (a) and/or the volume flow of the second feed gas provided in step (b) and/or the volume flow of the product gas removed in step (f) are completely stopped. This third time interval corresponds to standby operation of the plant.

[0029] The beginning and end of the first time interval are preferably determined by a reduced availability of resources, in particular electric power and/or first feed gas and/or second feed gas, compared to the second time interval. The first time interval preferably commences at the point in time at which the lower availability of electric power and/or first feed gas and/or second feed gas compared to the second time interval commences and preferably ends at the point in time at which the availability of electric power and/or first feed gas and/or second feed gas is completely re-established. The second time interval preferably commences at the point in time at which the first time interval ends. The first time interval is thus preferably characterized over its entire duration by a reduced availability of electric power and/or first feed gas and/or second feed gas compared to the second time interval.

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[0030] If the beginning and end of the first and second time intervals are determined by the different availability of electric power, this electric power is preferably

produced by means of renewable energies, in particular by photovoltaics and/or wind power.

5 [0031] In a particularly preferred embodiment, the availability of the first feed gas, preferably hydrogen, is causally coupled to the availability of electric power. In this preferred embodiment, the first feed gas, preferably hydrogen, is preferably obtained by electrolysis, with the electric power used in the electrolysis preferably being obtained from renewable energies, in particular by means of photovoltaics and/or wind power. If the availability of electric power is temporarily restricted, there is
10 also less electric power available for the electrolysis of water and consequently the availability of hydrogen (first feed gas) is temporarily restricted.

[0032] The second time interval is, according to the invention, preferably characterized, in contrast to the first time interval, by a sufficiently high availability
15 of electric power and/or by a sufficiently high availability of first feed gas, preferably hydrogen, and/or second feed gas in order to be able to carry out the process of the invention with maximum amount formed per unit time (i.e. at maximum conversion).

20 [0033] In all preferred embodiments according to the invention which are defined via a first time interval and a second time interval, the respective duration of the time interval is preferably independently in each case at least 30 minutes, preferably at least 1 hour, more preferably at least 2 hours, most preferably at least 6 hours and in particular at least 12 hours.

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[0034] If the process of the invention serves, for example, to synthesize ammonia from hydrogen and nitrogen and the hydrogen required for this is provided by electrolysis of water for which the required electricity is generated by photovoltaics, first and second time intervals can be predetermined by the day-night cycle. The first
30 time interval then preferably comprises the night phase and the second time interval the day phase. If, on the other hand, the electricity required for the electrolysis of water is generated by wind power, first and second time intervals can be prescribed

by the weather. The first time interval then preferably comprises the lull phase and the second time interval comprises the wind phase.

- [0035]** In a preferred embodiment of the process of the invention, the volume flow of the first feed gas provided in step (a) and/or the volume flow of the second feed gas provided in step (b) and/or the volume flow of the product gas removed in step (f) are varied during the process in such a way that
- (i) a first volume flow of the first feed gas is provided during the first time interval and a second volume flow of the first feed gas which differs from the first volume flow is provided during the second time interval; and/or
 - (ii) a first volume flow of the second feed gas is provided during the first time interval and a second volume flow of the second feed gas which differs from the first volume flow is provided during the second time interval; and/or
 - (iii) a first volume flow of the product gas is removed during the first time interval and a second volume flow of the product gas which differs from the first volume flow is removed during the second time interval.

- [0036]** Preference is given to both the volume flow of the first feed gas provided in step (a) and also the volume flow of the second feed gas provided in step (b) being varied simultaneously and preferably also equally.

- [0037]** In a preferred embodiment of the process of the invention, a smaller volume flow of the first feed gas is provided during the first time interval in step (a) and/or a smaller volume flow of the second feed gas is provided in step (b) and/or a smaller volume flow of the product gas is removed in step (f) than during the second time interval which follows the first time interval or which precedes the first time interval.

- [0038]** Preference is given to both a lower volume flow of the first feed gas being provided during the first time interval in step (a) and a lower volume flow of the second feed gas being provided in step (b) than during the second time interval which follows the first time interval or which precedes the first time interval.

- [0039]** In a preferred embodiment, the process of the invention comprises
- the temporary reduction of the amount of product gas formed per unit time during the first time interval by reduction (part load operation) or complete stoppage (standby operation) of the volume flow of the first feed gas provided in step (a) and/or the volume flow of the second feed gas provided in step (b) and/or the volume flow of product gas removed in step (f); and subsequently, preferably immediately subsequently,
 - increasing the amount of the product gas formed per unit time during the second time interval by increasing the volume flow of the first feed gas provided in step (a) and/or the volume flow of the second feed gas provided in step (b) and/or the volume flow of the product gas removed in step (f). The amount of product gas formed per unit time is preferably optimized here (full load operation).
- [0040]** The temporary reduction in the amount formed per unit time in part load operation or in standby operation is preferably determined by a reduced availability of electric power which is preferably produced by means of renewable energies, in particular photovoltaics and/or wind power, more preferably by a reduced availability of first and/or second feed gas, even more preferably by a reduced availability of hydrogen which is preferably obtained by electrolysis.
- [0041]** A preferred embodiment of the process of the invention comprises operation of the process in part load operation and subsequently in full load operation, and/or vice versa.
- [0042]** Another preferred embodiment of the process of the invention comprises operation of the process in standby operation, subsequently in part load operation and then in full load operation, and/or vice versa.
- [0043]** An alternating mode of operation is also possible according to the invention, for example
- full load operation -> part load operation -> full load operation -> part load operation, etc.; or

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- full load operation -> part load operation -> standby operation -> part load operation -> full load operation -> part load operation -> standby operation -> part load operation -> full load operation, etc.; or
- full load operation -> part load operation -> standby operation -> part load operation -> full load operation -> part load operation -> full load operation, etc.

[0044] The full load operation of the process of the invention preferably comprises operation at an optimized amount of product gas formed per unit time, preferably during the second time interval. The optimization is preferably carried out so that the availability of the volume flow of the first feed gas provided in step (a) and/or the volume flow of the second feed gas provided in step (b) and/or the volume flow of the product gas removed in step (f) does not have any decreasing influence on the amount of product gas formed per unit time, but instead the latter is limited by other factors, in particular the maximum capacity of the plant in which the process of the invention is carried out, under optimum conditions. Full load operation thus in principle comprises unlimited availability of the volume flow of the first feed gas provided in step (a) and/or the volume flow of the second feed gas provided in step (b) and/or an unlimited, i.e. virtually complete, removal of the volume flow of the product gas in step (f).

[0045] The part load operation of the process of the invention preferably comprises, compared to full load operation, the temporary reduction of the amount of product gas formed per unit time by reduction of the volume flow of the first feed gas provided in step (a) and/or the volume flow of the second feed gas provided in step (b) and/or the volume flow of the product gas removed in step (f), preferably in each case during the first time interval.

[0046] However, in various cases the part load operation cannot be sufficient in the event of a temporarily reduced availability of resources (e.g. electric power, first feed gas, second feed gas, etc.) to put the process of the invention into a state which does not mean complete shutdown and nevertheless makes it possible to overcome the reduced availability, i.e. the return of the resources makes the optionally rapid

running-up to full load operation possible. In these cases, the synthesis of product gas is, according to the invention, preferably temporarily stopped completely (standby operation).

5 **[0047]** The standby operation of the process of the invention preferably comprises, compared to part load operation, the temporary further reduction in the amount of product gas formed per unit time by stopping, i.e. completely interrupting, the volume flow of the first feed gas provided in step (a) and/or the volume flow of the second feed gas provided in step (b) and/or the volume flow of the product gas
10 removed in step (f), preferably in each case during the first time interval.

[0048] By means of standby operation, it is possible according to the invention firstly, in particular, to protect the apparatuses from excessive temperature and pressure fluctuations and secondly ensure rapid start-up and shutdown of the plant.
15 Here, the synthesis circuit or the reactor can be kept essentially at constant reaction temperature and constant reaction pressure in order to ensure rapid start-up and not to stress the components unnecessarily.

[0049] In a particularly preferred embodiment (cf. Figure 2), the provision of the first and second feed gas is switched off (steps (a) and (b)) in standby operation and product gas is also no longer removed (step (f)). Likewise, no purge gas is discharged. The operation of the circulation compressor of the synthesis circuit, on the other hand, is continued. To make up for the heat losses, an additional heating device is preferably integrated into the synthesis circuit. A pressure decrease caused
20 by progression of the reaction can be compensated for by brief introduction of first feed gas, e.g. hydrogen, and/or of second feed gas, e.g. nitrogen. Should cooling be necessary because of excessive evolution of heat as a result of a reaction of the remaining feed gases, this can likewise be realized in the circuit. The heating device can be electric or operated by means of a heating medium, preferably steam.
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[0050] In another, particularly preferred embodiment (cf. Figure 3), the complete synthesis circuit, circulation compressor and also an additional heating device are not utilized to make up for the heat losses, but instead a separate smaller circuit around

the reactor is utilized. Due to the smaller circuit size, the heat losses are lower in this mode of operation, so that a smaller supplementary heating device is sufficient to make up for any heat losses which occur. In addition, the heat losses are lower so that a blower for circulating the gas stream may be sufficient. A decrease in pressure due to progression of the reaction can be compensated for by brief introduction of a first feed gas, e.g. hydrogen, and/or second feed gas, e.g. nitrogen. Should cooling be necessary due to excessive evolution of heat due to reaction of the remaining feed gas, this can likewise be realized in the circuit.

- 10 **[0051]** In a preferred embodiment (cf. Figures 2 and 3), the process of the invention in standby operation comprises complete stoppage of the volume flow of the first feed gas provided in step (a) and/or the volume flow of the second feed gas provided in step (b) and/or the volume flow of the product gas removed in step (f) during the first time interval (standby operation), where the gas mixture A and/or the gas mixture B and/or the gas mixture C enriched in product gas and/or the gas mixture D depleted in product gas
- are compressed, preferably to make up decreases in pressure; and/or
 - are heated, preferably to make up for heat losses; and/or
 - are at least partly conveyed so as to bypass the catalyst, i.e. past the catalyst
- 20 (cf. Figure 7).

[0052] A compressor and/or a fan are preferably used for compression. Compressors which are suitable for compressing a gas are known to a person skilled in the art. Depending on the nature of the apparatus, a fan may be sufficient as compressor in order to make up for the comparatively low pressure decreases.

[0053] The gas mixture A and/or the gas mixture B and/or the gas mixture C enriched in product gas are preferably compressed during the first time interval in order to compensate for the pressure decreases occurring during the first time interval.

[0054] In a further preferred embodiment, the gas mixture A and/or the gas mixture B and/or the gas mixture C enriched in product gas are heated during the first time

interval, preferably to make up for heat losses. Heating of the gas mixtures is preferably carried out with the aid of a heating device.

[0055] In a preferred embodiment, the circulated gas mixture is thus

- 5 - heated by means of a heating device in order to make up for heat losses; and/or
- compressed by means of a compressor in order to compensate for pressure decreases

during the first time interval (standby operation).

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[0056] In the two above-described particularly preferred embodiments of standby operation, a circulation stream through the reactor is preferably maintained over the entire first time interval. In the simplest case, the flow conditions remain unchanged compared to normal operation, i.e. continuous flow through the catalyst beds occurs

15 in standby operation.

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[0057] In a further, particularly preferred embodiment, the flow conditions within the reactor are varied in standby operation, independently of the reactor type (quench reactor or reactor with intermediate cooling). For this purpose, “direct” flow through

20 the reactor can be realized according to the invention by means of appropriate construction of the reactor. In this case, the gas mixture flows mainly through the outer shell of the reactor without coming into contact with the catalyst beds, which leads to a lower pressure drop (cf. Figure 7). The flowing gas in the outer shell of the reactor, by passing the catalyst beds, also leads to thermal insulation of the catalyst

25 beds, so that these do not cool excessively.

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[0058] In preferred embodiments, the circulated gas mixture is therefore

- recirculated in a separate circuit to the reactor, where the circuit is equipped with means for heating and/or cooling the circulated gas mixture; and/or
- 30 - recirculated in a separate circuit to the reactor, where the circuit is equipped with means for compressing the circulated gas mixture; and/or
- conveyed within the reactor between the outer shell thereof and the interior catalyst bed or interior catalyst beds, so that the major part of the gas

mixture (gas mixture B), preferably the entire gas mixture, does not come into contact with the catalyst bed or the catalyst beds (cf. Figure 7) during the first time interval (standby operation).

5 **[0059]** In a preferred embodiment, the process of the invention is carried out in standby operation during the first time interval and a change from full load operation during the second time interval occurs with passage through part load operation during a time interval located between the first time interval and the second time interval by gradually increasing provision of first feed gas in step (a) and/or second
10 feed gas in step (b) or gas mixture A (fresh gas) in step (c).

[0060] In another preferred embodiment, the process of the invention is carried out in part load operation during the first time interval and a change from full load operation during the second time interval occurs by gradually increasing provision of
15 first feed gas in step (a) and/or second feed gas in step (b) or gas mixture A (fresh gas) in step (c).

[0061] In the two above-described embodiments, the second time interval follows the first time interval.

20

[0062] In a preferred embodiment, the process of the invention is carried out in full load operation during the second time interval and a change from standby operation during the first time interval occurs with passage through part load operation during a time interval located between the second time interval and the first time interval by
25 gradually decreasing provision of first feed gas in step (a) and/or second feed gas in step (b) or gas mixture A (fresh gas) in step (c).

[0063] In another preferred embodiment, the process of the invention is carried out in full load operation during the second time interval and a change to part load operation during the first time interval occurs by gradually decreasing provision of
30 first feed gas in step (a) and/or second feed gas in step (b) or gas mixture A (fresh gas) in step (c).

[0064] In the two above-described embodiments, the first time interval follows the second time interval.

5 [0065] In the process of the invention, the first feed gas and the second feed gas are converted into product gas.

[0066] In a preferred embodiment, the first feed gas is hydrogen. The hydrogen can originate from various sources. The hydrogen is preferably obtained by electrolysis of water. The electric power used for the electrolysis is particularly preferably
10 generated from renewable energies, in particular by photovoltaics, so that the availability of the power and thus also the availability of the electrolytically produced hydrogen can depend on solar radiation.

[0067] In a preferred embodiment, the second feed gas is selected from the group
15 consisting of nitrogen, carbon monoxide and carbon dioxide. Thus, preference is given according to the invention to the following combinations of first feed gas and second feed gas:

- (i) hydrogen and nitrogen,
- (ii) hydrogen and carbon monoxide, and
- 20 (iii) hydrogen and carbon dioxide.

[0068] In a preferred embodiment, the product gas is selected from the group consisting of ammonia, methanol, ethanol, higher alcohols, aldehydes, ketones, carboxylic acids and hydrocarbons (paraffins and olefins), with these also optionally
25 being able to be present in admixture. A person skilled in the art will know from which feed gases the product gases may be able to be produced. Particular preference is given to the product gas being ammonia, the first feed gas being hydrogen and the second feed gas being nitrogen.

30 [0069] In step (a) of the process of the invention, the first feed gas is provided.

[0070] If the first feed gas is hydrogen which is obtained by electrolysis of water, the provision of the first feed gas in step (a) is preferably effected by taking the hydrogen from the electrolysis cell.

5 [0071] As an alternative, however, the hydrogen can also originate from other sources, for example from synthesis gas.

[0072] In a preferred embodiment, the volume flow of the first feed gas provided in step (a) is varied during the course of the process so that a first volume flow of the
10 first feed gas is provided in a first time interval and a second volume flow of the first feed gas which differs from the first volume flow is provided in a second time interval.

[0073] In a preferred embodiment, step (a) of the process of the invention comprises
15 the substeps

(a₁) generation of electric power, preferably by means of regenerative energies, in particular by photovoltaics and/or by wind power; and
(a₂) electrolysis of water by means of the electric power generated in substep (a₁) to produce oxygen and hydrogen, with the hydrogen being
20 provided as first feed gas.

[0074] Preference is given in substep (a₁) to less electric power being generated during the first time interval and thus less hydrogen being provided as first feed gas in substep (a₂) than during the second time interval. In a preferred embodiment, no
25 electric power is generated during the first time interval and thus no hydrogen is provided as first feed gas in substep (a₂) (standby operation), but instead during the second time interval.

[0075] In step (b) of the process of the invention, the second feed gas is provided.
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[0076] If the second feed gas is nitrogen, the provision of the second feed gas in step (b) is preferably effected by introduction of virtually pure nitrogen, for example from an air fractionation plant.

[0077] If the second feed gas is carbon monoxide, this can, for example, originate from synthesis gas, so that in this case the provision of hydrogen as first feed gas in step (a) and the provision of carbon monoxide as second feed gas in step (b) can be effected simultaneously by the provision of synthesis gas.

[0078] In a preferred embodiment, the volume flow of the second feed gas provided in step (b) is varied during the course of the process so that a first volume flow of the second feed gas is provided in a first time interval and a second volume flow of the second feed gas which differs from the first volume flow is provided in a second time interval.

[0079] In step (c) of the process of the invention, the first feed gas and the second feed gas are combined to give a gas mixture A, so that the gas mixture A comprises the first feed gas and the second feed gas.

[0080] In addition, the gas mixture A can optionally comprise further gases, for example inert components.

[0081] Inert components are, for the purposes of the invention, inert in respect of the synthesis of the product gas, i.e. they are not reacted in the reaction of the first feed gas with the second feed gas to form product gas.

[0082] The relative molar ratio of first feed gas and second feed gas depends on the stoichiometry of the reaction by means of which the product gas is obtained.

[0083] If the first feed gas is hydrogen, the second feed gas is nitrogen and the product gas is ammonia, then the relative molar ratio of hydrogen to nitrogen is preferably about 3:1.

[0084] In a further preferred embodiment, the relative molar ratio of first feed gas to second feed gas in the gas mixture A is in the range from 10:1 to 1:10, more preferably from 8:1 to 1:8, even more preferably from 6:1 to 1:6, most preferably from 4:1 to 1:4 and in particular from 2:1 to 1:2. In another preferred embodiment,

the relative molar ratio of first feed gas to second feed gas in the gas mixture A is in the range from 10:1 to 1:1, more preferably from 8:1 to 1:1, even more preferably from 6:1 to 1:1, most preferably from 4:1 to 1:1 and in particular from 2:1 to 1:1. In a further preferred embodiment, the relative molar ratio of first feed gas to second feed gas in the gas mixture A is in the range from 1:1 to 1:10, more preferably from 1:1 to 1:8, even more preferably from 1:1 to 1:6, most preferably from 1:1 to 1:4 and in particular from 1:1 to 1:2.

[0085] The gas mixture A preferably comprises not more than 3% by volume of product gas, more preferably not more than 20% by volume, not more than 10, not more than 5.0% by volume, not more than 4.0% by volume, not more than 3.0% by volume, not more than 2.0% by volume, not more than 1.0% by volume or not more than 0.1% by volume of product gas. In a further preferred embodiment, the gas mixture A does not comprise any product gas.

[0086] The total content of first feed gas and second feed gas in the gas mixture A is preferably at least 90% by volume, more preferably at least 91% by volume, at least 92% by volume, at least 93% by volume, at least 94% by volume, at least 95% by volume, at least 96% by volume, at least 97% by volume, at least 98% by volume or at least 99% by volume.

[0087] In contrast to the process according to CA 2,790,545 A1, the process of the invention can be changed quickly and flexibly from full load operation to part load operation or standby operation even at an only small content of inert components in the gas mixture A.

[0088] The gas mixture A preferably comprises not more than 30% by volume of inert components, more preferably not more than 20% by volume, not more than 10, not more than 5.0% by volume, not more than 4.0% by volume, not more than 3.0% by volume, not more than 2.0% by volume, not more than 1.0% by volume or not more than 0.1% by volume of inert components. In a further preferred embodiment, the gas mixture A does not comprise any inert components.

[0089] The order of the steps (a) and (b) of the process of the invention is not fixed. Step (a) can be carried out before or after or at least partly simultaneously with step (b). Step (c) of the process of the invention can be carried out after steps (a) and (b) or at least partly simultaneously with step (a) and/or (b). In a particularly preferred embodiment, steps (a) and (b) are carried out simultaneously and step (c) is carried out subsequently thereto.

[0090] For example, hydrogen can be provided as first feed gas in step (a) and carbon monoxide or carbon dioxide can be provided as second feed gas in step (b). Both the hydrogen and the carbon monoxide or carbon dioxide can originate from synthesis gas, so that in this case the first and second feed gases can be provided simultaneously in steps (a) and (b) and can be combined to give the gas mixture A in step (c). In this case, steps (a) to (c) of the process of the invention are not realized by means of separate measures but are carried out together by the provision of synthesis gas.

[0091] The volume flow of the first feed gas provided in step (a) and/or the volume flow of the second feed gas provided in step (b) are preferably varied during the process in such a way that

- (i) a first volume flow of the first feed gas is provided during the first time interval and a second volume flow of the first feed gas is provided during the second time interval, with the second volume flow differing from the first volume flow; and/or
- (ii) a first volume flow of the second feed gas is provided during the first time interval and a second volume flow of the second feed gas is provided during the second time interval, with the second volume flow differing from the first volume flow.

[0092] The volume flow of the first feed gas provided during the first time interval in step (a) is preferably smaller than that during the second time interval. The volume flow of the first feed gas provided during the first time interval in step (a) is preferably not more than 99% of the volume flow of the first feed gas provided during the second time interval in step (a), more preferably not more than 90%, not

more than 80%, not more than 70%, not more than 60%, not more than 50%, not more than 40%, not more than 30%, not more than 20%, not more than 10%, not more than 5% or not more than 1%.

5 **[0093]** In a preferred embodiment, the relative ratio of the volume flow of the first feed gas provided during the first time interval in step (a) to the volume flow of the first feed gas provided during the second time interval is in the range from 1:1 to 1:100, more preferably from 1:2 to 1:90, even more preferably from 1:3 to 1:80, from 1:4 to 1:70, from 1:5 to 1:60, from 1:6 to 1:50, from 1:7 to 1:40, from 1:8 to 1:30 or
10 from 1:9 to 1:20. In a preferred embodiment, no first feed gas is provided during the first time interval (standby operation), but instead during the second time interval.

[0094] The volume flow of the first feed gas provided during the first time interval in step (a) is reduced by at least 5%, preferably by at least 10%, at least 20%, at least
15 30%, at least 40%, at least 50%, at least 60%, at least 70%, at least 80%, at least 90% or at least 99%, relative to the volume flow provided during the second time interval.

[0095] The volume flow of the second feed gas provided during the first time interval in step (b) is preferably smaller than during the second time interval. The
20 volume flow of the second feed gas provided during the first time interval in step (b) is preferably not more than 99% of the volume flow of the second feed gas provided during the second time interval in step (b), more preferably not more than 90%, not more than 80%, not more than 70%, not more than 60%, not more than 50%, not more than 40%, not more than 30%, not more than 20%, not more than 10%, not more
25 than 5% or not more than 1%.

[0096] In a preferred embodiment, the relative ratio of the volume flow of the second feed gas provided during the first time interval in step (b) to the volume flow of the second feed gas provided during the second time interval is in the range from
30 1:1 to 1:100, preferably from 1:2 to 1:90, more preferably from 1:3 to 1:80, from 1:4 to 1:70, from 1:5 to 1:60, from 1:6 to 1:50, from 1:7 to 1:40, from 1:8 to 1:30 or from 1:9 to 1:20. In a preferred embodiment, no second feed gas is provided during

the first time interval (standby operation), but instead during the second time interval.

5 [0097] The volume flow of the second feed gas provided during the first time interval in step (b) is preferably reduced by at least 5%, preferably by at least 10%, at least 20%, at least 30%, at least 40%, at least 50%, at least 60%, at least 70%, at least 80%, at least 90% or at least 99%, relative to the volume flow provided during the second time interval.

10 [0098] As a consequence of the variation according to the invention of the volume flow of the first feed gas provided in step (a) and/or the volume flow of the second feed gas provided in step (b) during the first time interval compared to the second time interval, the volume flow of the gas mixture A provided in each case can also change.

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[0099] The volume flow of the gas mixture A during the first time interval is preferably smaller than during the second time interval. The volume flow of the gas mixture A during the first time interval is preferably not more than 99% of the volume flow of gas mixture A during the second time interval, more preferably not more than 90%, not more than 80%, not more than 70%, not more than 60%, not more than 50%, not more than 40%, not more than 30%, not more than 20%, not more than 10%, not more than 5% or not more than 1%.

25 [0100] In step (d) of the process of the invention, the gas mixture A is combined with a circulated gas mixture to give a gas mixture B, with the gas mixture B comprising first feed gas, second feed gas, product gas and optionally further gases, e.g. inert components.

30 [0101] The circulated gas mixture typically comprises unreacted first feed gas and/or unreacted second feed gas and/or product gas and optionally further constituents, in particular inert components.

[0102] In a preferred embodiment, the content of gas mixture A in the gas mixture B is not more than 90% by volume, more preferably not more than 80% by volume, not more than 70% by volume, not more than 60% by volume, not more than 50% by volume, not more than 40% by volume, not more than 30% by volume, not more than 20% by volume, not more than 10% by volume, not more than 5% by volume or not more than 1% by volume.

[0103] In another preferred embodiment, the content of gas mixture A in the gas mixture B is at least 1% by volume, more preferably at least 5% by volume, at least 10% by volume, at least 20% by volume, at least 30% by volume, at least 40% by volume, at least 50% by volume, at least 60% by volume, at least 70% by volume, at least 80% by volume or at least 90% by volume.

[0104] In a preferred embodiment, the content of circulated gas mixture in the gas mixture B is not more than 90% by volume, more preferably not more than 80% by volume, not more than 70% by volume, not more than 60% by volume, not more than 50% by volume, not more than 40% by volume, not more than 30% by volume, not more than 20% by volume, not more than 10% by volume, not more than 5% by volume or not more than 1% by volume.

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[0105] In another preferred embodiment, the content of circulated gas mixture in the gas mixture B is at least 1% by volume, more preferably at least 5% by volume, at least 10% by volume, at least 20% by volume, at least 30% by volume, at least 40% by volume, at least 50% by volume, at least 60% by volume, at least 70% by volume, at least 80% by volume or at least 90% by volume.

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[0106] The relative ratio of the volume flow of the circulated gas mixture to the volume flow of the gas mixture A, which are combined in step (d) to give the gas mixture B, is preferably in the range from 10:1 to 1:10, more preferably from 8:1 to 1:8, even more preferably from 6:1 to 1:6, most preferably from 4:1 to 1:4 and in particular from 2:1 to 1:2. In another preferred embodiment, the relative ratio of the volume flow of the circulated gas mixture to the volume flow of the gas mixture A, which are combined in step (d) to give the gas mixture B, is in the range from 10:1 to

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1:1, more preferably from 8:1 to 1:1, even more preferably from 6:1 to 1:1, most preferably from 4:1 to 1:1 and in particular from 2:1 to 1:1. In a further preferred embodiment, the relative ratio of the volume flow of the circulated gas mixture to the volume flow of the gas mixture A, which are combined in step (d) to give the gas mixture B, is in the range from 1:1 to 1:10, more preferably from 1:1 to 1:8, even more preferably from 1:1 to 1:6, most preferably from 1:1 to 1:4 and in particular from 1:1 to 1:2.

[0107] The content of gas mixture A in the gas mixture B is preferably

10 - not more than 90% by volume, preferably not more than 80% by volume, not more than 70% by volume, not more than 60% by volume, not more than 50% by volume, not more than 40% by volume, not more than 30% by volume, not more than 20% by volume, not more than 10% by volume, not more than 5% by volume or not more than 1% by volume, during the first

15 time interval; and/or

- at least 1% by volume, more preferably at least 5% by volume, at least 10% by volume, at least 20% by volume, at least 30% by volume, at least 40% by volume, at least 50% by volume, at least 60% by volume, at least 70% by volume, at least 80% by volume or at least 90% by volume, during the

20 second time interval.

[0108] The content of circulated gas mixture in the gas mixture B is preferably

- at least 1% by volume, more preferably at least 5% by volume, at least 10% by volume, at least 20% by volume, at least 30% by volume, at least 40% by volume, at least 50% by volume, at least 60% by volume, at least 70% by volume, at least 80% by volume or at least 90% by volume, during the first

25 time interval; and/or

- not more than 90% by volume, more preferably not more than 80% by volume, not more than 70% by volume, not more than 60% by volume, not more than 50% by volume, not more than 40% by volume, not more than 30% by volume, not more than 20% by volume, not more than 10% by volume, not more than 5% by volume or not more than 1% by volume, during the second time interval.

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[0109] The relative ratio of the volume flows of the circulated gas mixture and of the gas mixture A, which are combined in step (d) to give the gas mixture B, is preferably

- at least 2:1, more preferably at least 3:1, at least 4:1, at least 5:1, at least 6:1, at least 7:1, at least 8:1, at least 9:1 or at least 10:1, during the first time interval; and/or
- not more than 10:1, preferably not more than 9:1, preferably not more than 8:1, not more than 7:1, not more than 6:1, not more than 5:1, not more than 4:1, not more than 3:1 or not more than 2:1, during the second time interval.

[0110] Steps (c) and (d) can, according to the invention, be carried out alphabetically one after the other or simultaneously.

[0111] In step (e) of the process of the invention, the first and second feed gases present in the gas mixture B are reacted over a catalyst to give product gas with production of a gas mixture C enriched in product gas.

[0112] Catalysts for gas-phase reactions are known to a person skilled in the art. With regard to the synthesis of ammonia as product gas, which is preferred according to the invention, reference may be made, for example, to A. Nielsen, I. Dybkjaer, Ammonia – Catalysis and Manufacture, Springer Berlin, Chapter 6, pages 202-326.

[0113] In the process of the invention, the reaction in step (e) preferably does not proceed to completion, so that the gas mixture C enriched in product gas comprises unreacted first feed gas and unreacted second feed gas.

[0114] The content of product gas in the gas mixture C enriched in product gas is preferably at least 5% by volume, more preferably at least 10% by volume, at least 20% by volume, at least 30% by volume, at least 40% by volume, at least 50% by volume, at least 60% by volume, at least 70% by volume, at least 80% by volume, at least 90% by volume or at least 99% by volume.

[0115] In a further preferred embodiment, the content of product gas in the gas mixture C enriched in product gas is in the range from 1 to 90% by volume, more preferably in the range from 5 to 50% by volume, even more preferably in the range from 10 to 40% by volume, most preferably in the range from 15 to 35% by volume and in particular in the range from 20 to 30% by volume.

[0116] The content of product gas in the gas mixture C enriched in product gas depends first and foremost on the reaction equilibrium. The content of product gas in the gas mixture C enriched in product gas under the given conditions is preferably greater during the first time interval than during the second time interval, preferably at least twice as great, more preferably at least three times as great, even more preferably at least four times as great, most preferably at least five times as great and in particular at least six times as great.

[0117] The content of first feed gas in the gas mixture C enriched in product gas is preferably at least 1% by volume, more preferably at least 2% by volume, at least 4% by volume, at least 6% by volume, at least 8% by volume, at least 10% by volume, at least 12% by volume, at least 14% by volume, at least 16% by volume, at least 18% by volume or at least 20% by volume.

[0118] The content of first feed gas in the gas mixture C enriched in product gas is dependent first and foremost on the reaction equilibrium. The content of first feed gas in the gas mixture C enriched in product gas under the given conditions is preferably greater during the second time interval than during the first time interval, preferably at least twice as great, more preferably at least three times as great, even more preferably at least four times as great, most preferably at least five times as great and in particular at least six times as great.

[0119] The content of second feed gas in the gas mixture C enriched in product gas is preferably at least 1% by volume, more preferably at least 2% by volume, at least 4% by volume, at least 6% by volume, at least 8% by volume, at least 10% by volume, at least 12% by volume, at least 14% by volume, at least 16% by volume, at least 18% by volume or at least 20% by volume.

[0120] The content of second feed gas in the gas mixture C enriched in product gas depends first and foremost on the reaction equilibrium. The content of second feed gas in the gas mixture C enriched in product gas under the given conditions is preferably greater during the second time interval than during the first time interval,
5 preferably at least twice as great, more preferably at least three times as great, even more preferably at least four times as great, most preferably at least five times as great and in particular at least six times as great.

[0121] The synthesis of product gas, e.g. ammonia, in part load operation is possible
10 to only a limited extent by means of simple reduction of the amount of the gas mixture A (fresh gas) or of the first feed gas and the second feed gas (nitrogen and hydrogen in the example of the synthesis of ammonia), since this can lead to an increase in the residence time of the gas mixture in the catalyst bed. The greater residence time can in turn bring about higher conversions, particularly in the front-
15 end catalyst beds. This can result in the temperature within the reactor increasing as a result of the exothermic reaction to such an extent that damage to the catalyst or to the reactor can occur.

[0122] In order to prevent this, it is possible according to the invention for only the
20 volume flow of one of the two feed gases (e.g. hydrogen) to be reduced and the volume flow of the other feed gas (e.g. nitrogen) to be kept constant or be increased. As a result, the residence time of the gas mixture in the catalyst bed is not increased as greatly and accordingly no impermissible temperature increase occurs.

[0123] If, however, the second feed gas is nitrogen, a comparatively large quantity
25 of energy is required for the provision thereof. In addition, it can be necessary to increase the proportion of the purge gas stream which may be required for discharge of the inert components present in the nitrogen. This can lead to an undesirable loss of first feed gas, e.g. hydrogen, and also product gas, e.g. ammonia.

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[0124] To avoid overheating in part load operation, it is in principle also possible to lower the synthesis pressure in order to shift the equilibrium further to the side of the starting materials. However, this can lead to an undesirable dynamic mechanical

stressing of the reactor and the other plant components and also impair the efficiency of the process.

5 **[0125]** In a preferred embodiment, the variation according to the invention
- of the volume flow of the first feed gas provided in step (a) and/or
- of the volume flow of the second feed gas provided in step (b) and/or
- of the volume flow of the product gas removed in step (f) is therefore
combined with a variation of the division of the volume (sub)streams of the
gas mixture B over the individual catalyst beds of a reactor which comprises
10 a plurality of catalyst beds.

[0126] In a preferred embodiment of the process of the invention
- the reaction in step (e) is carried out in at least a first catalyst bed and a
second catalyst bed;
15 - where a first substream of the gas mixture B is fed to the first catalyst bed
and the gas mixture leaving the first catalyst bed is fed to the second catalyst
bed;
- where a second substream of the gas mixture B is fed to the second catalyst
bed with bypassing of the first catalyst bed;
20 - where the relative volume flow ratio of the first substream to the second
substream is increased during the first time interval; and
- where the relative volume flow ratio of the first substream to the second
substream is reduced again during the second time interval.

25 **[0127]** The reactor is preferably a quench reactor having at least two catalyst beds
connected in series, preferably at least three catalyst beds connected in series. Such a
reactor is schematically illustrated by way of example in Figure 7, in which the
second catalyst bed (7b) and the third catalyst bed (7c) are arranged downstream of
the first catalyst bed (7a).

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[0128] To operate a quench reactor in part load operation, i.e. during the first time
interval, preference is given according to the invention to not only the total volume
flow of the gas mixture B, i.e. the sum of all substreams over the individual catalyst

beds, being reduced but the relative ratio of the volume flows over the individual catalyst beds of the quench reactor preferably also being varied so that, based on the relative ratio, more of the gas mixture B flows into the first catalyst bed during the first time interval in part load operation than during the second time interval in full load operation. In this way, the volume flow in the first catalyst bed is preferably reduced significantly underproportionally despite a lower throughput through the reactor. The temperature thus increases to only a limited extent in this catalyst bed. In the further, downstream catalyst beds, the conversion is lower due to temperature and partial pressure, so that the temperature in these zones also does not increase excessively despite a lower quench volume flow.

[0129] Owing to the good regulability of both the quench streams (gas mixture B) and the condensation temperature (removal of product gas), both concepts allow a comparatively quick change in load.

[0130] The objective of such a strategy in part load operation is to keep the temperature in the catalyst beds within the required temperature range in each case, e.g. for an ammonia synthesis preferably in the range from 380°C to 510°C.

[0131] In a preferred embodiment, the substreams of the gas mixture B (quench streams) are fed to the catalyst beds in such a way that the temperature in each catalyst bed does not exceed 530°C.

[0132] For this purpose, the exothermic synthesis reaction is preferably controlled, typically reduced to a suitable extent. If the load is reduced, i.e. less gas mixture A (fresh gas) is introduced into the circuit and the circulation flow is thus also reduced, the residence time of the gas mixture increases in all three catalyst beds. Accordingly, the reaction would progress further in all three catalyst beds, as occurs in full load operation, and the maximum temperature of 510°C would be exceeded in all cases.

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[0133] As a countermeasure, the temperature for removal of the product gas in step (f) (condenser temperature) is preferably increased according to the invention, which leads to an increase in the molar proportion of product gas in the gas phase at the

outlet of the condenser and thus also at the inlet into the reactor. Consequently, the conversion is reduced by shifting of the reaction equilibrium and the reaction kinetics.

- 5 **[0134]** Despite this measure, a decreasing load may nevertheless lead to a high exit temperature from the first catalyst bed. For this reason, an increased proportion of gas mixture is preferably additionally introduced into the first catalyst bed with decreasing load and the residence time increase is thereby limited there.
- 10 **[0135]** In a preferred embodiment, the first substream of the gas mixture B makes up at least 10% by volume of the total volume flow of the gas mixture B, more preferably at least 20% by volume, at least 30% by volume, at least 40% by volume, at least 50% by volume, at least 60% by volume, at least 70% by volume, at least 80% by volume, at least 90% by volume or at least 99% by volume.
- 15 **[0136]** The first substream of the gas mixture B preferably makes up
- at least 10% by volume of the total volume flow of the gas mixture B during the first time interval, more preferably at least 20% by volume, at least 30% by volume, at least 40% by volume, at least 50% by volume, at least 60% by
 - 20 volume, at least 70% by volume, at least 80% by volume, at least 90% by volume or at least 99% by volume, and/or
 - at least 10% by volume of the total volume flow of the gas mixture B during the second time interval, more preferably at least 20% by volume, at least 30% by volume, at least 40% by volume, at least 50% by volume, at least
 - 25 60% by volume, at least 70% by volume, at least 80% by volume, at least 90% by volume or at least 99% by volume.
- [0137]** In part load operation during the first time interval, the first substream of the gas mixture B preferably makes up at least 40% by volume, more preferably at least
- 30 45% by volume or at least 50% by volume, of the total volume flow of the gas mixture B.

[0138] Preferably,

- 33 -

- during the first time interval (part load operation) the first substream of the gas mixture B makes up more than 50% by volume, preferably at least 55% by volume or at least 60% by volume, of the total volume flow of the gas mixture B and the second substream, or in the case of more than two catalyst beds the sum of all further substreams makes up less than 50% by volume, preferably not more than 45% by volume or not more than 40% by volume, of the total volume flow of the gas mixture B, and/or
- during the second time interval (full load operation) the first substream of the gas mixture B makes up less than 50% by volume, more preferably not more than 45% by volume or not more than 40% by volume, of the total volume flow of the gas mixture B and the second substream, or in the case of more than two catalyst beds, the sum of all further substreams makes up more than 50% by volume, more preferably at least 55% by volume or at least 60% by volume, of the total volume flow of the gas mixture B.

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[0139] The second substream of the gas mixture B, which is fed to the second catalyst bed with bypassing of the first catalyst bed, preferably makes up not more than 99% by volume of the total volume flow of the gas mixture B, more preferably not more than 90% by volume, not more than 80% by volume, not more than 70% by volume, not more than 60% by volume, not more than 50% by volume, not more than 40% by volume, not more than 30% by volume, not more than 20% by volume, not more than 10% by volume, not more than 5% by volume or not more than 1% by volume.

25 **[0140]** The second substream of the gas mixture B, which is fed to the second catalyst bed with bypassing of the first catalyst bed, preferably makes up

- not more than 99% by volume of the total volume flow of the gas mixture B, more preferably not more than 90% by volume, not more than 80% by volume, not more than 70% by volume, not more than 60% by volume, not more than 50% by volume, not more than 40% by volume, not more than 30% by volume, not more than 20% by volume, not more than 10% by volume, not more than 5% by volume or not more than 1% by volume, during the first time interval; and/or

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- not more than 99% by volume of the total volume flow of the gas mixture B, more preferably not more than 90% by volume, not more than 80% by volume, not more than 70% by volume, not more than 60% by volume, not more than 50% by volume, not more than 40% by volume, not more than 30% by volume, not more than 20% by volume, not more than 10% by volume, not more than 5% by volume or not more than 1% by volume, during the second time interval.

[0141] In a preferred embodiment, the relative ratio of the first substream of the gas mixture B during the first time interval to the first substream of the gas mixture B during the second time interval is in the range from 10:1 to 1:1, more preferably in the range from 8:1 to 1.1:1, even more preferably in the range from 6:1 to 1.2:1, even more preferably in the range from 5:1 to 1.3:1, most preferably in the range from 4:1 to 1.4:1 and in particular in the range from 3:1 to 1.5:1.

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[0142] The ratio of the first substream of the gas mixture B during the first time interval to the first substream of the gas mixture B during the second time interval is preferably at least 1.1:1, more preferably at least 1.2:1, even more preferably at least 1.3:1, at least 1.4:1, at least 1.5:1, at least 1.6:1, at least 1.7:1, at least 1.8:1, at least 1.9:1, at least 2:1, at least 3:1, at least 4:1, at least 5:1, at least 6:1, at least 7:1, at least 8:1, at least 9:1 or at least 10:1.

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[0143] In a preferred embodiment, the relative ratio of the second substream of the gas mixture B during the first time interval to the second substream of the gas mixture B during the second time interval is in the range from 1:90 to 1:1, more preferably in the range from 1:70 to 1:2, more preferably in the range from 1:60 to 1:3, even more preferably in the range from 1:50 to 1:4, most preferably in the range from 1:40 to 1:5 and in particular in the range from 1:30 to 1:6.

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[0144] The ratio of the second substream of the gas mixture B during the first time interval to the second substream of the gas mixture B during the second time interval is preferably not more than 1:90, more preferably not more than 1:80, even more preferably not more than 1:70, not more than 1:60, not more than 1:50, not more than

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1:40, not more than 1:30, not more than 1:20, not more than 1:10, not more than 1:5, not more than 1:4, not more than 1:3 or not more than 1:2.

[0145] The relative ratio of the first substream of the gas mixture B to the second substream of the gas mixture B is preferably

- in the range from 99:0.1 to 1:1, more preferably in the range from 95:1 to 2:1, more preferably in the range from 90:2 to 3:1, from 85:3 to 4:1, from 80:4 to 5:1, from 75:4.5 to 8:1 or from 70:5 to 10:1, during the first time interval; and/or
- 10 - in the range from 1.1:1 to 10:1, more preferably in the range from 1.2:1 to 8:1, even more preferably from 1.3:1 to 6:1, most preferably from 1.4:1 to 4:1 and in particular from 1.5:1 to 2:1, during the second time interval.

[0146] The relative ratio of the first substream of the gas mixture B to the second substream of the gas mixture B is preferably

- at least 10:1, more preferably at least 20:1, at least 30:1, at least 40:1, at least 50:1, at least 60:1, at least 70:1, at least 80:1, at least 90:1 or at least 99:1, during the first time interval; and/or
- at least 1.1:1, more preferably at least 1.2:1, at least 1.3:1, at least 1.4:1, at least 1.5:1, at least 1.6:1, at least 1.7:1, at least 1.8:1, at least 1.9:1, at least 2:1, at least 5:1 or at least 10:1, during the second time interval.

[0147] The process of the invention is preferably carried out with the aid of at least one catalyst bed, more preferably using at least two catalyst beds and even more preferably using at least three catalyst beds.

[0148] The temperature in the catalyst beds is preferably in the range from 100°C to 800°C, more preferably in the range from 200°C to 700°C, even more preferably in the range from 300°C to 600°C, most preferably in the range from 350°C to 550°C and in particular in the range from 380°C to 510°C.

[0149] In a preferred embodiment, the temperature in the catalyst beds is at least 100°C, preferably at least 150°C, at least 200°C, at least 300°C, at least 350°C or at least 380°C.

5 **[0150]** In another preferred embodiment, the temperature in the catalyst beds is not more than 800°C, more preferably not more than 750°C, not more than 700°C, not more than 650°C, not more than 600°C, not more than 550°C or not more than 510°C.

10 **[0151]** In step (f) of the process of the invention, product gas is removed from the gas mixture C enriched in product gas to produce a gas mixture D depleted in product gas.

[0152] The removal preferably comprises cooling and condensing the product gas
15 by means of a suitable apparatus, in particular by means of a heat exchanger using a cooling medium, and subsequently separating off the condensed product gas in a suitable separation apparatus.

[0153] As regards the removal of the ammonia synthesized according to the
20 invention as product gas, reference may likewise be made by way of example to the full text of A. Nielsen, I. Dybkjaer, Ammonia – Catalysis and Manufacture, Springer Berlin, 1995, Chapter 6, pages 202-326.

[0154] Step (f) of the process of the invention is preferably carried out by
25 influencing and varying the temperature for the condensation of the product gas.

[0155] Gas-phase reactions such as the synthesis of ammonia are equilibrium
reactions in which the position of the equilibrium depends on the partial pressures of the feed gases and product gases and also on the temperature. In order to allow the
30 two feed gases, e.g. nitrogen and hydrogen, to react as completely as possible, product gas, e.g. ammonia, is removed, preferably condensed out, preferably continuously, in the synthesis circuit and the remaining gas phase is returned in a circuit (cf. Figure 1). This means that product gas is continuously withdrawn from

the reaction, so that according to the law of mass action the feed gases can react further in order to approach the chemical equilibrium. The remaining residual amount of product gas in the gas mixture is dependent not only the pressure but also on the condenser exit temperature.

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[0156] For part load operation, the condenser exit temperature is preferably increased. This leads to a higher partial pressure of the product gas in the equilibrium with the condensed-out liquid and thus to a higher proportion of product gas in the gas mixture. As a result, the position of the equilibrium in the catalyst beds is shifted
10 to the side of the product gas, e.g. ammonia, and accordingly a smaller conversion is achieved and less heat of reaction is liberated, so that overheating of the reactor can be avoided at a suitably chosen operating temperature of the condenser (cf. Figure 4).

[0157] In a preferred embodiment, at least 1% by volume, more preferably at least
15 10% by volume, at least 20% by volume, at least 30% by volume, at least 40% by volume, at least 50% by volume, at least 60% by volume, at least 70% by volume, at least 80% by volume, at least 90% by volume or at least 99% by volume, of the product gas are removed from the gas mixture C enriched in product gas, based on the total amount of the product gas which is present in the gas mixture C enriched in
20 product gas before the removal.

[0158] In another preferred embodiment, not more than 90% by volume, more preferably not more than 80% by volume, not more than 70% by volume, not more than 60% by volume, not more than 50% by volume, not more than 40% by volume,
25 not more than 30% by volume, not more than 20% by volume, not more than 10% by volume or not more than 1% by volume of the product gas are removed from the gas mixture C enriched in product gas, based on the total amount of product gas which is present in the gas mixture C enriched in product gas before the removal.

[0159] In a preferred embodiment, the relative ratio of the content of product gas in the gas mixture C to the content of product gas in the gas mixture D is in the range from 10:1 to 1:1, more preferably from 8:1 to 1:1, even more preferably from 6:1 to 1:1, most preferably from 4:1 to 1:1 and in particular from 2:1 to 1:1.
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[0160] In a preferred embodiment, the volume flow of the product gas removed in step (f) is varied during the course of the process so that a first volume flow of the product gas is removed in a first time interval and a second volume flow of the product gas which differs from the first volume flow is removed in a second time interval.

[0161] The volume flow of the product gas removed during the second time interval in step (f) is preferably greater than during the first time interval, preferably at least twice as great, more preferably at least three times as great, even more preferably at least four times as great, most preferably at least five times as great and in particular at least six times as great. In a preferred embodiment (standby operation), virtually no or absolutely no product gas is removed during the first time interval, but instead during the second time interval.

[0162] In a preferred embodiment, the relative ratio of the volume flow of product gas removed during the first time interval in step (f) to the volume flow of the product gas removed during the second time interval is in the range from 1:1 to 1:100, more preferably from 1:2 to 1:90, even more preferably from 1:3 to 1:80, from 1:4 to 1:70, from 1:5 to 1:60, from 1:6 to 1:50, from 1:7 to 1:40, from 1:8 to 1:30 or from 1:9 to 1:20.

[0163] The volume flow of the product gas removed during the first time interval in step (f) is preferably reduced by at least 5%, more preferably by at least 10%, at least 20%, at least 30%, at least 40%, at least 50%, at least 60%, at least 70%, at least 80%, at least 90% or at least 99%, relative to the volume flow removed during the second time interval.

[0164] In a preferred embodiment, no product gas is removed from the gas mixture C during the first time interval, so that the gas mixture D, which is recirculated, contains the entire amount of the product gas (standby operation).

[0165] In a preferred embodiment, the content of product gas in the gas mixture D depleted in product gas is at least 0.5% by volume, more preferably at least 5% by

volume, at least 10% by volume, at least 20% by volume, at least 30% by volume, at least 40% by volume, at least 50% by volume, at least 60% by volume, at least 70% by volume, at least 80% by volume, at least 90% by volume or at least 99% by volume.

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[0166] In another preferred embodiment, the content of product gas in the gas mixture D depleted in product gas is not more than 90% by volume, more preferably not more than 80% by volume, not more than 70% by volume, not more than 60% by volume, not more than 50% by volume, not more than 40% by volume, not more than 30% by volume, not more than 20% by volume, not more than 10% by volume or not more than 1% by volume.

[0167] The content of product gas in the gas mixture D depleted in product gas depends on the phase equilibrium/partial pressure. The content of product gas in the gas mixture D depleted in product gas under the given conditions is greater during the second time interval than during the first time interval, preferably at least twice as great, more preferably at least three times as great, even more preferably at least four times as great, most preferably at least five times as great and in particular at least six times as great. In a preferred embodiment (standby operation), virtually no or absolutely no product gas is removed during the first time interval, but instead during the second time interval.

[0168] The content of product gas in the gas mixture D depleted in product gas is preferably

- 25 - at least 1% by volume, more preferably at least 5% by volume, at least 10% by volume, at least 20% by volume, at least 30% by volume, at least 40% by volume, at least 50% by volume, at least 60% by volume, at least 70% by volume, at least 80% by volume or at least 90% by volume, during the first time interval; and/or
- 30 - not more than 90% by volume, more preferably not more than 80% by volume, not more than 70% by volume, not more than 60% by volume, not more than 50% by volume, not more than 40% by volume, not more than 30% by volume, not more than 20% by volume, not more than 10% by

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volume, not more than 5% by volume or not more than 1% by volume, during the second time interval.

5 [0169] In step (g) of the process of the invention, at least part of the gas mixture D depleted in product gas is returned as circulated gas mixture to step (d).

[0170] The relative proportion by volume of the gas mixture D which is recirculated to step (d) is preferably at least 1% by volume, more preferably at least 10% by volume, at least 20% by volume, at least 30% by volume, at least 40% by volume, at least 50% by volume, at least 60% by volume, at least 70% by volume, at least 80%
10 by volume, at least 90% by volume, at least 95% by volume or at least 99% by volume, in each case based on the total volume of the gas mixture D depleted in product gas.

[0171] The relative proportion by volume of the gas mixture D which is recirculated to step (d) is preferably not more than 99%, more preferably not more than 90%, not
15 more than 80%, not more than 70%, not more than 60%, not more than 50%, not more than 40%, not more than 30%, not more than 20%, not more than 10%, or not more than 1%, in each case based on the total volume of the gas mixture D depleted in product gas.

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[0172] In a preferred embodiment, the volume flow of the gas mixture D depleted in product gas which in step (g) is returned as circulated gas mixture to step (d) is varied during the course of the process so that a first volume flow of the gas mixture D depleted in product gas is recirculated to step (d) in a first time interval and a
25 second volume flow of the gas mixture D depleted in product gas which differs from the first volume flow is recirculated to step (d) in a second time interval.

[0173] The ratio of the volume flow of the gas mixture D depleted in product gas which is recirculated during the first time interval to step (d) to the volume flow of the gas mixture D depleted in product gas which is recirculated during the second
30 time interval to step (d) is preferably in the range from 10:1 to 1:10, more preferably from 8:1 to 1:8, even more preferably from 6:1 to 1:6, most preferably from 4:1 to 1:4, and in particular from 2:1 to 1:2. In another preferred embodiment, the ratio of

the volume flow of the gas mixture D depleted in product gas which is recirculated during the first time interval to step (d) to the volume flow of the gas mixture D depleted in product gas which is recirculated during the second time interval to step (d) is in the range from 10:1 to 1:1, more preferably from 8:1 to 1:1, even more preferably from 6:1 to 1:1, most preferably from 4:1 to 1:1, and in particular from 2:1 to 1:1. In a further preferred embodiment, the ratio of the volume flow of the gas mixture D depleted in product gas which is recirculated during the first time interval to step (d) to the volume flow of the gas mixture D depleted in product gas which is recirculated during the second time interval to step (d) is in the range from 1:1 to 1:10, more preferably from 1:1 to 1:8, even more preferably from 1:1 to 1:6, most preferably from 1:1 to 1:4 and in particular from 1:1 to 1:2.

[0174] The steps (a) to (g) of the process of the invention are preferably carried out in alphabetic order, but can also be carried out in an order different therefrom. Here, individual steps of the process of the invention can be carried out partly or completely simultaneously with other steps of the process of the invention.

[0175] The process of the invention preferably comprises influencing of the amount of product gas formed per unit time by varying the volume flow of the first feed gas provided in step (a) and/or varying the volume flow of the second feed gas provided in step (b) and/or varying the volume flow of product gas removed in step (f).

[0176] The variation of the volume flows of first feed gas and/or second feed gas provided and/or the variation of the volume flow of product gas removed are preferably carried out as a function of the availability of electric power, even more preferably as a function of the availability of electric power which is generated by means of renewable energies, in particular photovoltaics and/or wind power. The variation is even more preferably carried out as a function of the availability of first feed gas and/or second feed gas, preferably hydrogen which is preferably obtained from the electrolysis of water.

[0177] The “amount formed per unit time” is, for the purposes of the invention, preferably the conversion of the hydrogen used in the process into product gas.

[0178] In terms of the amount of product gas formed per unit time, the states of the process of the invention are in the following ranking order: standby operation < part load operation < full load operation.

5 **[0179]** The amount of product gas formed per unit time during part load operation is preferably relatively not more than 90% of the amount of product gas formed per unit time during full load operation, more preferably not more than 80%, even more preferably not more than 70%, or not more than 60% or not more than 50%, most preferably not more than 40% or not more than 30% and in particular not more than
10 20% or not more than 10% of the amount of product gas formed per unit time during full load operation.

[0180] The amount of product gas formed per unit time during standby operation is preferably virtually 0%.

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[0181] A further aspect of the invention provides an apparatus for producing a product gas from a first feed gas and a second feed gas, in particular by the process according to any of claims 1 to 12, comprising the following components which are operatively connected to one another:

- 20 - a first inlet device configured for providing the first feed gas;
- a second inlet device configured for providing the second feed gas;
- a reactor configured for synthesizing a product gas from the first feed gas and the second feed gas to produce a gas mixture C enriched in product gas under heterogeneous gas catalysis;
25 - a removal device configured for removing product gas from the gas mixture C enriched in product gas with production of a gas mixture D depleted in product gas; and
- a control device which is configured for bringing the apparatus into a first operating state or into a second operating state, where the first operating
30 state differs from the second operating state
- (i) in terms of a smaller volume flow of the first feed gas which is provided by the first inlet device and/or a smaller volume flow of the second feed gas which is provided by the second inlet device and/or a

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smaller volume flow of the product gas which is removed by the removal device, and/or

(ii) in terms of a smaller amount of product gas formed per unit time,

where the reactor (7) comprises at least one catalyst bed which has a radial distance
5 from an outer shell of the reactor, which results in a free space able to be flowed through between the catalyst bed and the outer shell of the reactor, in such a way that a gas mixture entering the reactor (7) in the first operating state can flow essentially axially through the reactor without flowing through the catalyst bed, where
the reactor (7) comprises at least two catalyst beds (7a, 7b, 7c) arranged in series in
10 the axial direction, where means by means of which at least one substream of the inflowing gas mixture can flow directly, with bypassing of a first catalyst bed (7a), into a second catalyst bed (7b) located downstream of this first catalyst bed (7a).

[0182] All preferred embodiments which have been described above in connection
15 with the process of the invention for synthesizing a product gas or for storing energy also apply analogously to the apparatus of the invention.

[0183] The apparatus of the invention preferably comprises a store for first feed gas, preferably hydrogen, the capacity of which is preferably sufficient for equalizing
20 relatively small temporary fluctuations in the availability of the first feed gas.

[0184] The apparatus of the invention preferably comprises a conduit which is configured for circulating a gas mixture from an outlet of the reactor to an inlet of the reactor, where the circuit comprises a heating device and the control device is
25 configured so that in the first operating state at least part of the gas mixture C leaving the reactor is conveyed from the outlet of the reactor through the conduit to the inlet of the reactor and during this passage is heated in the heating device.

[0185] The apparatus of the invention is particularly suitable for carrying out the
30 process of the invention.

[0186] A further aspect of the invention therefore provides for the use of the apparatus of the invention for synthesizing product gas by the process of the invention for synthesizing product gas.

5 [0187] The invention will be illustrated below with the aid of Figures 1 to 7. However, the figures serve merely for explanation of preferred embodiments of the invention and are not to be interpreted as constituting a restriction.

[0188] Figure 1 shows a circulation process which is particularly suitable for an ammonia and methanol synthesis, but also for the production of ethanol and of higher alcohols, aldehydes, ketones, carboxylic acids and also paraffins and olefins. The circuit comprises at least one reactor (7) in which a gas mixture B (16) is converted into product gas (17) with production of a gas mixture C enriched in product gas. The product gas (17) flows through a heat exchanger (18), e.g. a condenser (18). Here, product gas (19) is condensed out as a function of the temperature of the heat exchanger and the pressure. The uncondensed gas mixture D (16) depleted in product gas and the condensed product gas (19) can be conveyed further as circulating gas to a precipitator (15), with it being advantageous to introduce fresh gas mixture A (fresh gas) (14) together with the circulated gas mixture (19) into the precipitator (15) in order to be able to precipitate the condensed product gas (20) in liquid form better and discharge it from the process. The gas mixture D (16) depleted in product gas which has not been precipitated by the precipitator (15) is recirculated to the reactor (7).

25 [0189] The circuit depicted in Figure 2 is a possible variant for keeping the circulation process hot in standby operation. The reactor (7) is kept hot by means of the gas mixture which has been heated by means of a heat exchanger (18) and a heating device (13). Here, the circuit comprises a reactor (7), a heat exchanger (18), a circulation compressor (6) and a heating device (13).

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[0190] Figure 3 shows a small, separate circuit for standby operation, in which a heat exchanger or a heating device (13) and a blower (21) are provided in the circuit around a reactor (7) to compensate for the heat losses and the decreases in pressure.

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The gas mixture goes via an inlet (12) into the reactor (7) which the product gas mixture leaves via the outlet (11), with a proportion of the product gas being able to be discharged from the circuit via the removal device (8), for example a valve. Otherwise,

Patentkrav

1. Fremgangsmåde til syntese af en produktgas, omfattende følgende trin:

(a) tilvejebringelse af en første reaktantgas;

(b) tilvejebringelse af en anden reaktantgas;

5 (c) kombination af den første reaktantgas og den anden reaktantgas, idet der opnås en gasblanding A omfattende første og anden reaktantgas;

(d) kombination af gasblandingen A med en gasblanding cirkuleret i et kredsløb, idet der opnås en gasblanding B omfattende første reaktantgas, anden reaktantgas og produktgas;

10 (e) reaktion af den i gasblandingen B indeholdte første reaktantgas og anden reaktantgas i en katalysator til produktgas, idet der frembringes en gasblanding C, som er beriget med produktgas;

15 (f) udledning af produktgas fra gasblandingen C, som er beriget med produktgas, idet der frembringes en gasblanding D, som er fattig på produktgas; og

(g) recirkulering af i det mindste en del af gasblandingen D, fattig på produktgas, som den i kredsløbet cirkulerede gasblanding til trin (d);

20 hvor den i trin (a) tilvejebragte volumenstrøm af den første reaktantgas og/eller den i trin (b) tilvejebragte volumenstrøm af den anden reaktantgas og/eller den i trin (f) udledte volumenstrøm af produktgassen varieres under fremgangsmåden, som desuden omfatter

25 - midlertidig reduktion af den mængde produktgas, som frembringes pr. tidsenhed i et første tidsinterval ved at reducere den i trin (a) tilvejebragte volumenstrøm af den første reaktantgas og/eller den i trin (b) tilvejebragte volumenstrøm af den anden reaktantgas og/eller den i trin (f) udledte volumenstrøm af produktgassen; og

- 5 - øgning af den mængde produktgas, som frembringes pr. tidsenhed i et andet tidsinterval ved at øge den i trin (a) tilvejebragte volumenstrøm af den første reaktantgas og/eller den i trin (b) tilvejebragte volumenstrøm af den anden reaktantgas og/eller den i trin (f) udledte volumenstrøm af produktgassen,

kendetegnet ved, at

- 10 - reaktionen i trin (e) sker i mindst en første katalysatormasse og en anden katalysatormasse;
- en første delstrøm af gasblandingen B tilføres den første katalysatormasse, og gasblandingen, som forlader den første katalysatormasse, tilføres den anden katalysatormasse;
- en anden delstrøm af gasblandingen B tilføres den anden katalysatormasse ved at bypasse den første katalysatormasse;
- 15 - det relative volumenstrømforhold mellem den første delstrøm og den anden delstrøm øges i det første tidsinterval; og
- det relative volumenstrømforhold mellem den første delstrøm og den anden delstrøm reduceres igen i det andet tidsinterval.

- 20 **2.** Fremgangsmåde ifølge krav 1, hvor den første delstrøm af gasblandingen B udgør mindst 40 vol.-% af den samlede mængde af gasblandingen B.

- 25 **3.** Fremgangsmåde ifølge et af kravene 1 eller 2 omfattende fuldstændig afbrydelse af den i trin (a) tilvejebragte volumenstrøm af den første reaktantgas og/eller den i trin (b) tilvejebragte volumenstrøm af den anden reaktantgas og/eller den i trin (f) udledte volumenstrøm af produktgassen i mindst et tredje tidsinterval.

- 4.** Fremgangsmåde ifølge krav 3, hvor gasblandingen A og/eller gasblandingen B og/eller gasblanding C, som er beriget med produktgas, og/eller gasblanding D, som er fattig på produktgas,

- komprimeres; og/eller

- opvarmes; og/eller

- hvor gasblanding A og/eller gasblanding D, som er fattig på produktgas, i det mindste delvist cirkuleres ved at bypasse katalysatoren.

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5. Fremgangsmåde ifølge et af de foregående krav, hvor den første reaktantgas er hydrogen.

10 **6.** Fremgangsmåde ifølge krav 5, hvor hydrogenen udvindes gennem elektrolyse af vand.

7. Fremgangsmåde ifølge et af de foregående krav, hvor reaktionen i trin (e) forløber ufuldstændigt, sådan at gasblanding C, som er beriget med produktgas, omfatter ureageret første reaktantgas og ureageret anden reaktantgas.

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8. Fremgangsmåde ifølge et af de foregående krav, hvor indholdet af produktgas i gasblanding D, som er fattig på produktgas, udgør mindst 0,5 vol.-%.

20 **9.** Fremgangsmåde ifølge et af de foregående krav, hvor den anden reaktantgas er nitrogen, og produktgassen er ammoniak.

10. Fremgangsmåde ifølge et af kravene 1 til 8, hvor den anden reaktantgas er kulstofmonoxid eller kulstofdioxid.

25 **11.** Fremgangsmåde ifølge et af de foregående krav, hvor gasblanding A ikke omfatter produktgas.

12. Fremgangsmåde ifølge et af kravene 1 til 11, hvor udledningen af produktgassen i trin (f) sker ved udkondensering ved hjælp af en kondensator, og
30 at kondensatorens udgangstemperatur øges i første tidsinterval.

13. Anlæg til fremstilling af en produktgas af en første reaktantgas og en anden reaktantgas, særligt efter fremgangsmåden ifølge et af de foregående krav 1 til 12, omfattende følgende komponenter, som er funktionelt forbundet med hinanden:

- 5 - en første indløbsanordning konfigureret til tilvejebringelse af den første reaktantgas;
 - en anden indløbsanordning konfigureret til tilvejebringelse af den anden reaktantgas;
 - 10 - en reaktor (7) konfigureret til syntese af en produktgas af den første reaktantgas og den anden reaktantgas, til frembringelse af en gasblanding C, som er beriget med produktgas, under heterogen gaskatalyse;
 - en udledningsanordning (8) konfigureret til udledning af produktgas fra gasblandingen C, som er beriget med produktgas, idet der frembringes en gasblanding D, som er fattig på produktgas; og
 - 15 - en styringsanordning, som er konfigureret til at sætte anordningen i en første driftstilstand eller i en anden driftstilstand, hvor den første driftstilstand adskiller sig fra den anden driftstilstand
 - 20 (i) ved en mindre volumenstrøm af den første reaktantgas, som tilvejebringes via den første indløbsanordning, og/eller ved en mindre volumenstrøm af den anden reaktantgas, som tilvejebringes via den anden indløbsanordning, og/eller ved en mindre volumenstrøm af produktgassen, som udledes via udledningsanordningen (8), og/eller
 - (ii) ved en mindre mængde af produktgas genereret pr. tidsenhed,
- hvor reaktoren (7) mindst omfatter en katalysatormasse, som har en radial afstand til en udvendig muffe på reaktoren, hvorved der skabes et rum, som kan
- 25 gennemstrømmes, mellem katalysatormassen og reaktorens udvendige muffe, på en sådan måde, at i første driftstilstand en gasblanding, som strømmer ind i reaktoren (7), kan gennemstrømme reaktoren i det væsentlige aksialt uden at gennemstrømme katalysatormassen, hvor
- reaktoren (7) omfatter mindst to katalysatormasser (7a, 7b, 7c) placeret efter

hinanden i aksial retning, hvor der er tilvejebragt midler, ved hjælp af hvilke mindst en delstrøm af den indstrømmende gasblanding ved bypassing af en første katalysatormasse (7a) kan strømme direkte ind i en anden katalysatormasse (7b), som er efterkoblet denne første katalysatormasse (7a).

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14. Anlæg ifølge det foregående krav, som omfatter en rørledning (10), der er konfigureret til at cirkulere en gasblanding fra et udløb (11) på reaktoren (7) til et indløb (12) på reaktoren (7) i et kredsløb, hvor kredsløbet omfatter en varmeanordning (13), og hvor styringsanordningen er konfigureret sådan, at i den første driftstilstand i det mindste en del af gasblandingen C, som forlader reaktoren (7), cirkuleres fra reaktorens (7) udløb (11) via rørledningen (10) til reaktorens (7) indløb (12) og herved opvarmes i varmeanordningen (13).

15. Anvendelse af et anlæg ifølge et af kravene 13 eller 14 i en fremgangsmåde ifølge et af de foregående proceskrav.

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Figure 1

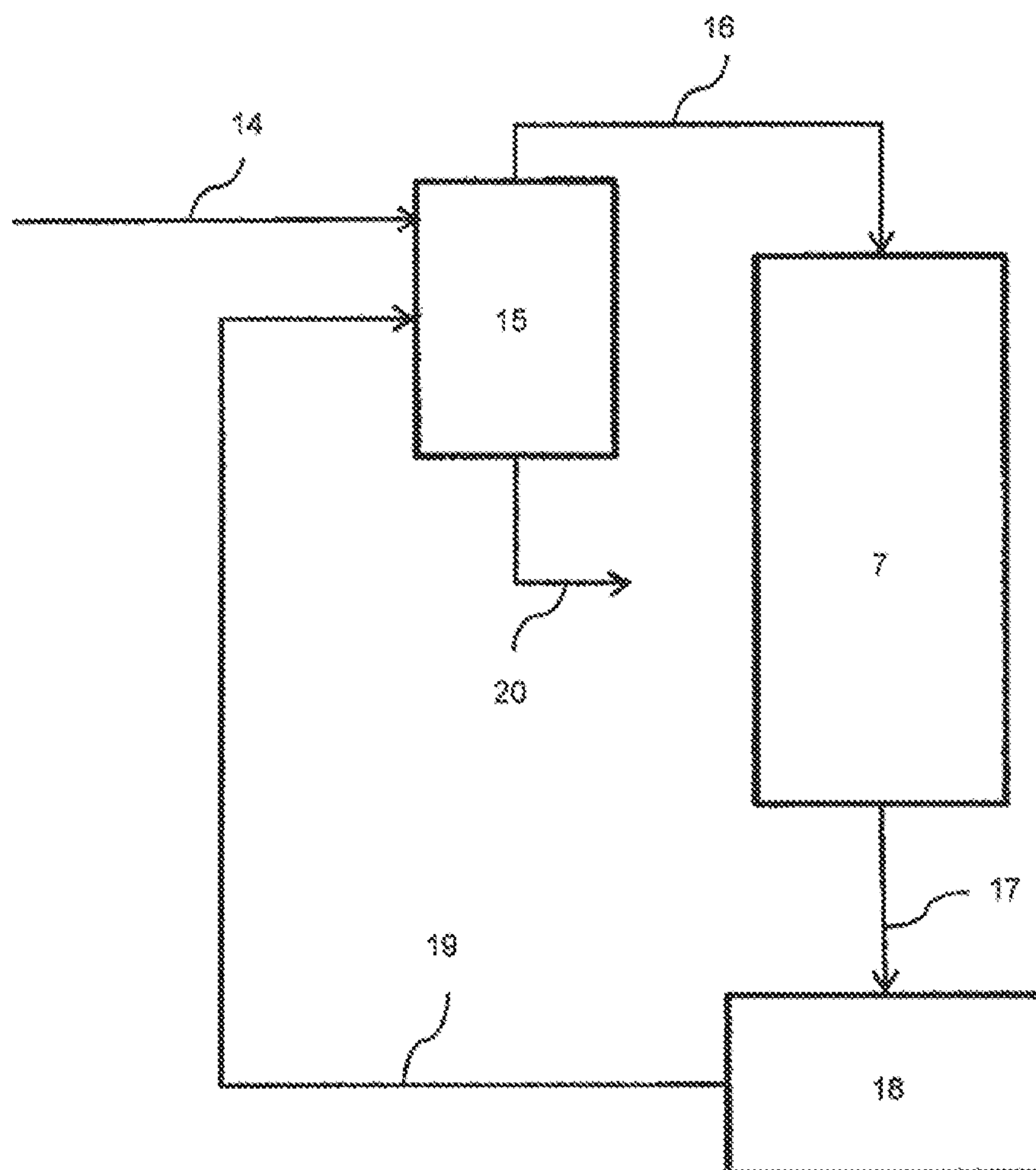


Figure 2

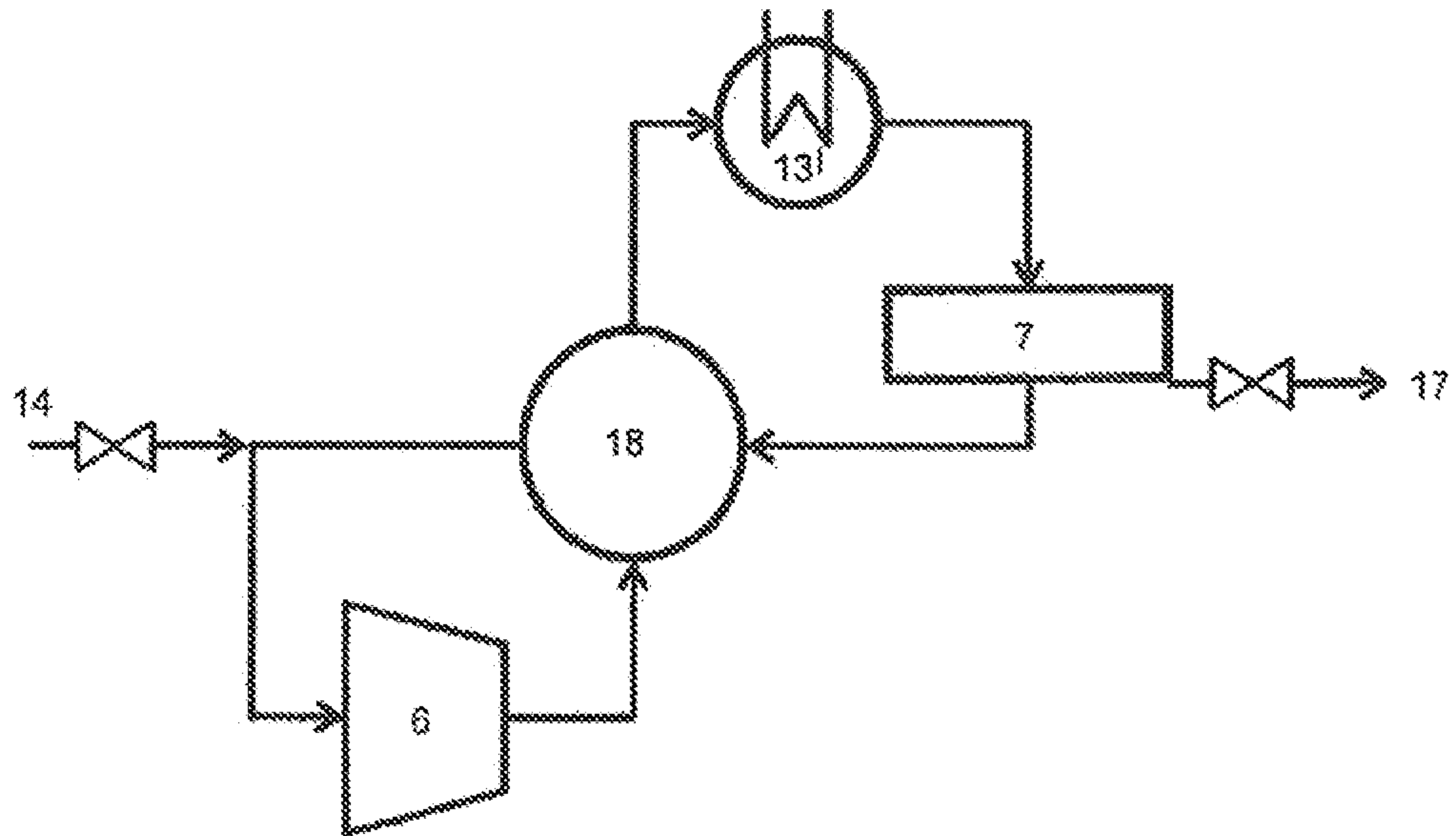


Figure 3

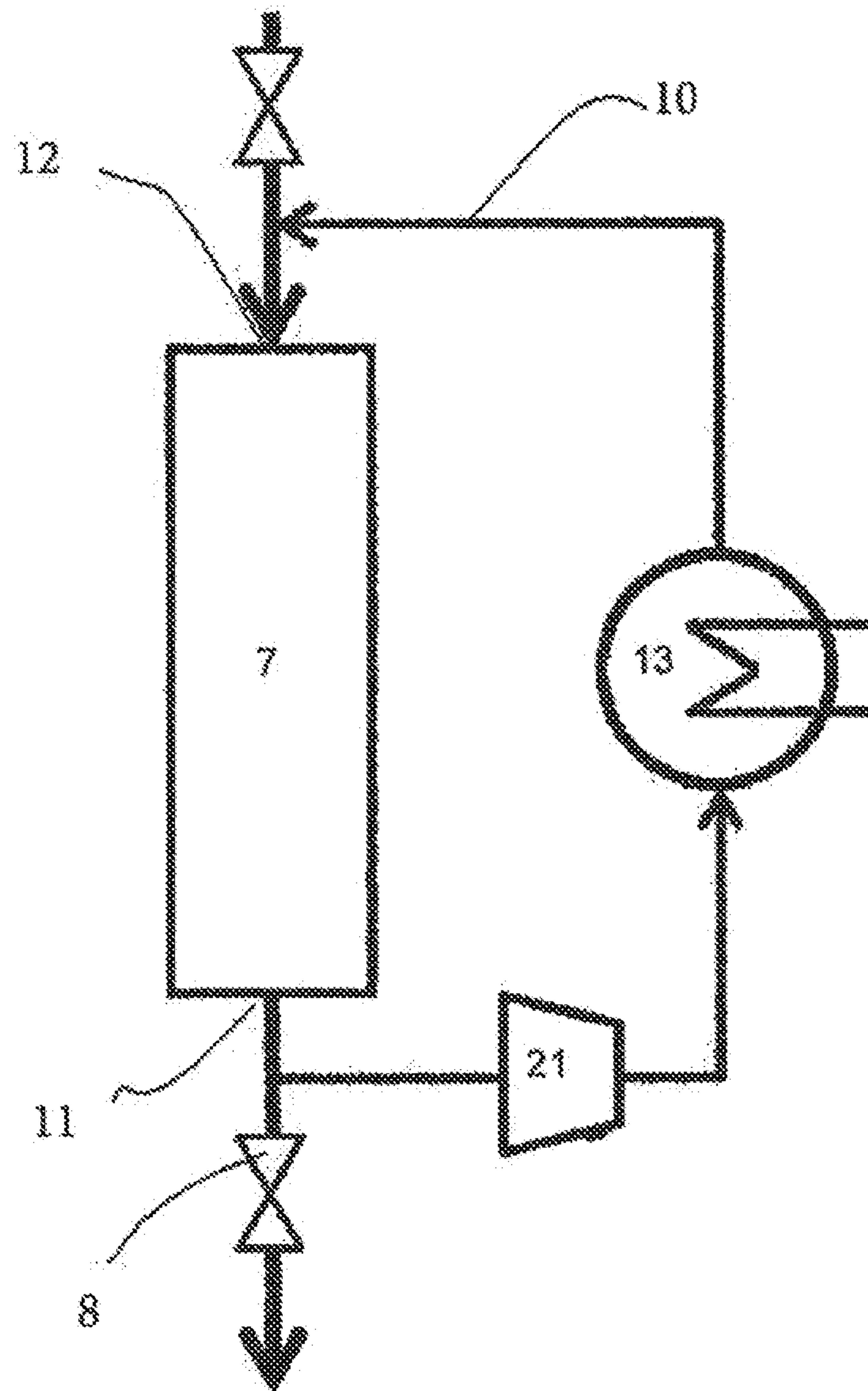


Figure 4

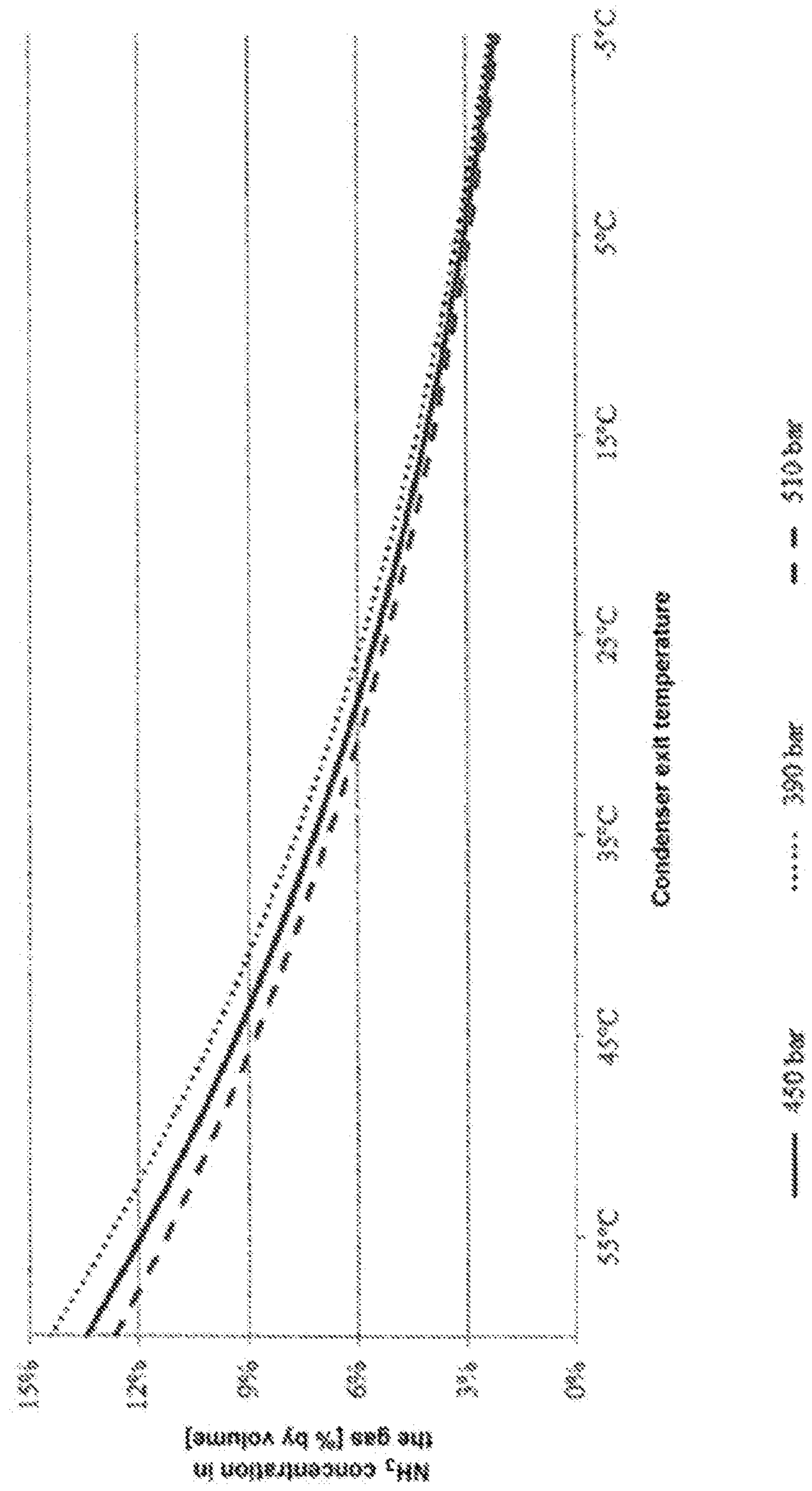


Figure 5

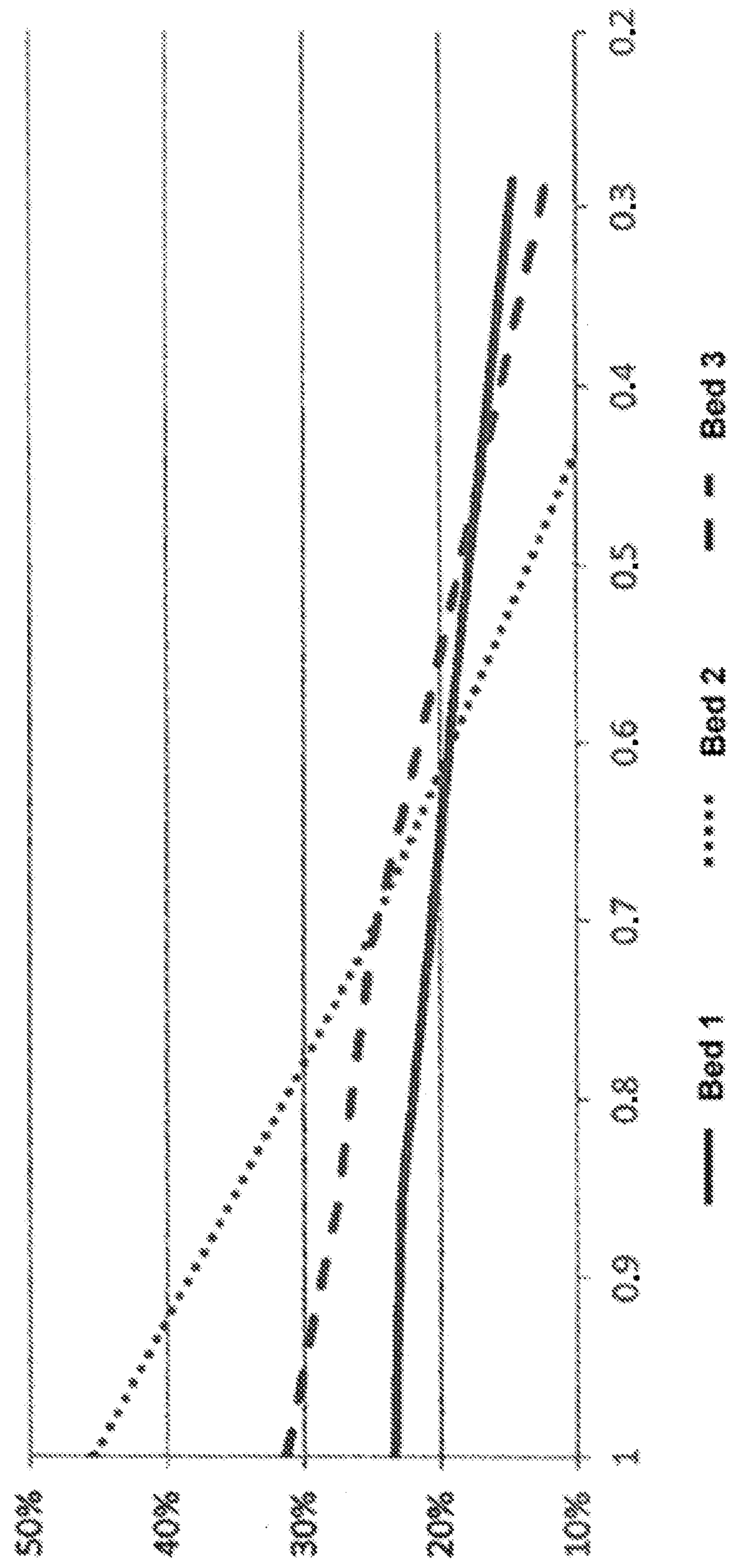
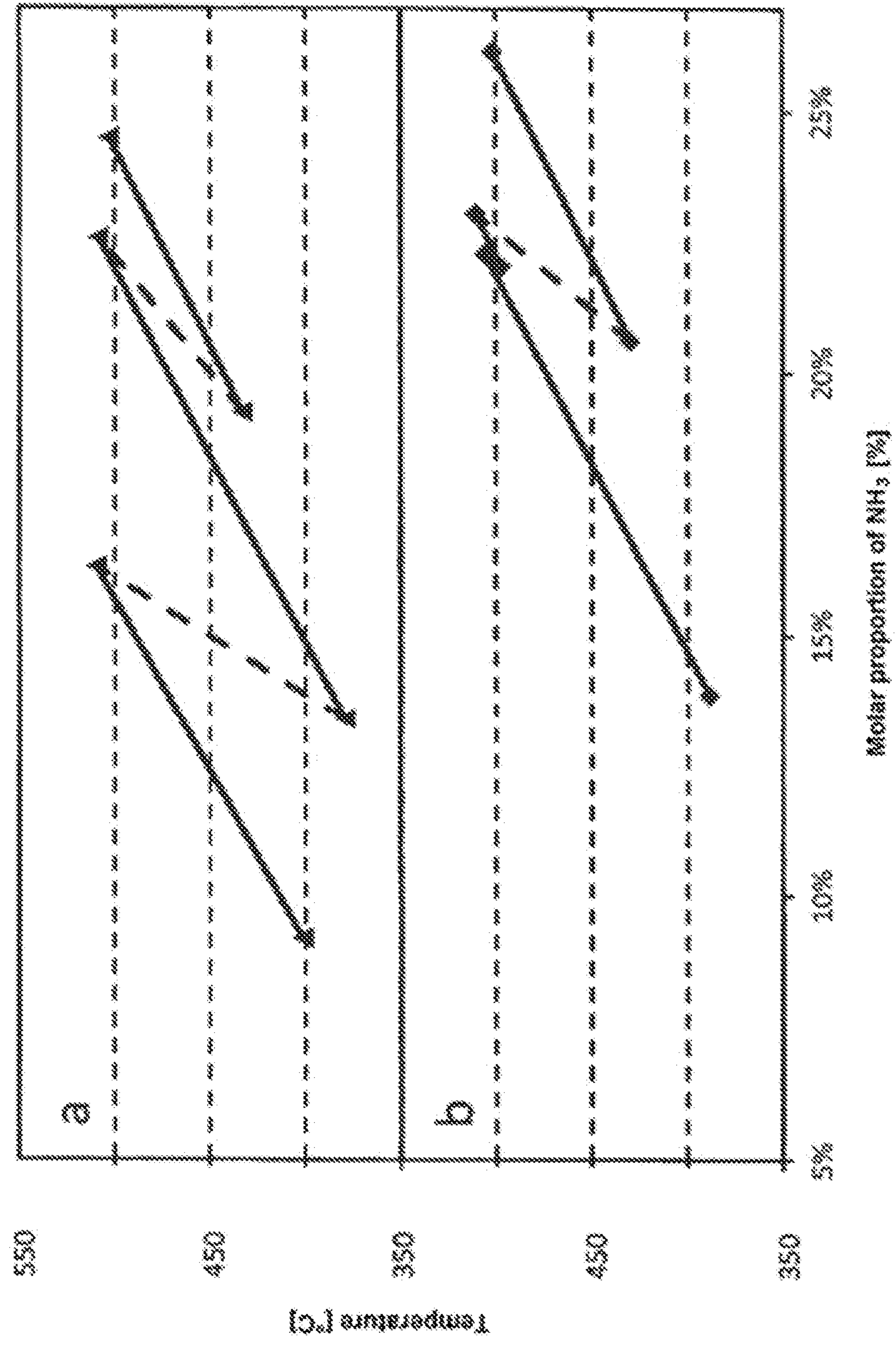


Figure 6



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Figure 7

