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Process and Apparatus for the Gasification of Solids

5 This invention relates to a process and an apparatus for the gasification of carbonaceous solids.

Gasification is referred to as a chemico-physical process, in which at least part of a solid matter is transferred into a gaseous end product. The gaseous end
10 product is a mixture which chiefly consists of carbon monoxide (CO) and hydrogen (H₂). Many reactions, which so far have only been known incompletely, proceed at the same time. The actual gasification is effected by exothermal combustion of the solids. The products of this reaction can react further with the solids and additionally introduced steam or among each other. Except for the
15 combustion reaction, all essential reactions are equilibrium reactions, so that the conversion also can proceed in the reverse direction. At the comparatively high temperatures in the coal gasification (600 to 1600 °C), the composition of the product gases generally is very close to the equilibrium.

20 In principle, three different process types for the gasification of solids are known: the gasification in fluidized beds, the gasification in a fixed bed formed of the solids, and finally the gasification in an entrained-bed reactor.

Independent of how the gasification reaction is effected, the synthesis gas CO
25 and H₂ obtained therein must be purified subsequently. Since during the reaction steam is required as educt and water is one of the possible reaction products, the water amongst others must be removed from the gas stream.

In the stream of water separated, further impurities are contained, such as solids, ammonia, Phenols etc., so that the stream of water must be purified.
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Such process is known for example from DE 41 07 109 C1. Solid fuels are gasified at a pressure in the range from 10 to 100 bar with gasification media containing oxygen and steam for generating a raw gas. The raw gas coming from the gasification is cooled to temperatures of 20 to 200 °C, whereby a condensate rich in water is obtained. The condensate is separated and at least partly evaporated, wherein condensate vapor and a salt-containing brine are withdrawn separately. The salt-containing brine is burnt, the condensate vapor is partly added to the purified raw gas.

DE 35 15 484 describes that the condensate obtained in the stepwise cooling of the product gas is cooled in a washer-cooler, which is charged with circulating water, whereby halogens are largely washed out. The used washing water, which has temperatures of 120 to 220 °C, is expanded to a lower pressure, whereby flash steam and a liquid phase are obtained. The flash steam virtually free of halogens is discharged, the major part of the liquid phase is again passed into the washer-cooler, and the residual liquid phase is supplied to a disposal.

DE 32 07 779 A1 describes that the condensate obtained from the synthesis gas is expanded and supplied to a separating means, from which a condensate phase largely consisting of water is withdrawn. The condensate phase is cooled in direct contact with colder gas, before it is used for cooling the raw gas stream. The heated, steam-containing cooling gas is supplied to a combustion and utilized e.g. for heating the reactor.

DD 147679 describes the recirculation of a stream substantially consisting of water, which originates from a hydroclone in which a stream rich in solids is separated from a stream poor in solids. The overflow stream poor in solids is admixed to the educt stream of the gasifier and serves as source for the re-

quired steam in the reaction. The underflow stream rich in solids is distilled at atmospheric pressure for further purification.

5 GB 2 198 744 A finally describes a coal gasification in the fixed bed, in which after separation of the gases the waste water stream is guided into an evaporation means. From there, the gaseous constituent is recirculated into the reactor as gasification medium.

10 Recirculations of streams of water, which partly contain major amounts of solids, otherwise are known from completely different processes. For example, US 5,586,510 describes the production of cement in a rotary kiln, wherein sludge which is obtained in the cement production is recirculated into the rotary kiln, atomized there and burnt.

15 All processes have in common that large amounts of waste water cannot be used further in the process. Therefore, an expensive and costly waste water aftertreatment is necessary, in order to process the stream or streams such that environmental standards are observed during the disposal.

20 It can be desirable for an embodiment of the invention to reduce the production of waste water in the gasification of solids.

25 An aspect provides a process for the gasification of carbonaceous solids, wherein in a first process step solids are at least partly converted to CO and H₂ in a gasification step to produce a gas stream, wherein in a second process step a stream containing water is separated, and wherein in a third process step the water-containing stream obtained in the second process step is subjected to a water purification, wherein in the water purification the stream containing water is separated into three streams with different degrees of purity, wherein a first
30 stream of water, which has the highest degree of purity, is supplied to a steam

generation, wherein a second stream of water, which has the medium degree of purity, is supplied to a solids slurring, preferably to a further ash treatment or to a slurry to be introduced into the reactor, wherein a third stream of water, which has the lowest degree of purity, is recirculated into the solids gasification, wherein the solids slurring comprises flushing out the ash from the solids gasification and wherein the gasification of the solids is carried out in a fixed bed.

A carbonaceous solid matter may be gasified and at least partly converted to carbon monoxide and hydrogen in the presence of oxygen and steam. The gas mixture produced then may then be supplied to a separating device, in which liquid fractions may be separated from gaseous fractions, whereby a so-called raw gas stream and a stream containing water can be obtained. The liquid stream containing water finally may be subjected to water purification.

According to the process, the water purification is effected such that three streams with different degrees of purity are obtained. The first stream has the highest degree of purity, almost exclusively consists of water, and may have the following composition:

Table 1: Substances contained in the first stream

pH		7 - 8
Total amount of iron (Fe)	µg/l	< 200, preferred < 20
Total amount of copper (Cu)	µg/l	< 30 preferred < 3
Total amount of silica (SiO ₂)	µg/l	< 200 preferred < 20
Total amount of sodium (Na)	µg/l	< 100

		preferred < 10
Organic components (chem. oxygen demand, short COD)	µg/l	< 2000 preferred < 200
Conductivity at 25 °C	µS/cm	< 0.2-2 preferred < 2
Oxygen (O ₂)	µg/l	50-1000 preferred 50-250

Such stream may thus be suitable to be utilized for steam generation. When the stream does not reach these limit values, the first stream can be used as cooling water inside the plant.

5

The second stream may have a medium degree of purity:

Table 2: Substances contained in the second stream

Organic components (chem. oxygen demand, short COD)	mg/l	100 - 10,000
Organic components (biochem. oxygen demand, short BOD)	mg/l	10 - 1000
Ammonium nitrogen (short NH ₄ -N)	mg/l	5 - 500
Nitrate nitrogen (short NO ₃ -N)	mg/l	5 - 1000
Phosphate phosphorus (short PO ₄ -P)	mg/l	2 - 100

Total content of suspended solids (short TSS)	mg/l	5 - 1000
Total content of dissolved solids (short TDS)	mg/l	100 - 25,000

The third stream may have the lowest degree of purity and carries high solids content:

5 Table 3: Substances contained in the third stream

Organic compounds	wt-%	5-80
Nitrogen- and/or phosphorus-containing compounds	wt-%	1-5
Metal hydroxides	wt-%	5-15

10 The procedure according to an embodiment of the invention can provide for the recirculation of each of these three streams. That stream of water which has the highest degree of purity may be supplied to a water inlet of a steam generation; that stream of water which has the medium degree of purity may be supplied to a further treatment of ash obtained in the gasification reactor; and/or that stream of water which has the lowest degree of purity and is rich in solids may be guided back into the gasification reactor.

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It may be particularly advantageous when all three streams are recirculated inside the gasification unit itself. The amount of waste water obtained in the

process thereby may only be reduced significantly, but may actually be lowered completely to zero.

At the same time, the streams with the medium and the lowest degree of purity also can jointly be recirculated into the gasification zone, whereby a particularly high utilization of the contained organic components can be effected.

In part, the degree of purity of the first stream may not reach the purity necessary for steam generation. With the same pH value it may then have a composition at which the individual components are present in three times, in part even six times the concentration as compared to the concentration indicated in Table 1. A stream with this composition can be utilized as cooling water stream at any point of the process, without the cooling water being evaporated or can be added to cooling tower of the gasification plant.

In an advantageous embodiment of the invention, the stream with the lowest degree of purity may be separated by decanting in a first step. It may be possible to further lower the water content for example by evaporation. In a second step, the remaining stream subsequently may be supplied to a reverse osmosis. The reverse osmosis is a physical process for concentrating substances dissolved in liquids, in which the natural process of osmosis is reversed with pressure. The medium in which the concentration of a certain substance should be reduced can be separated from a medium in which the concentration should be increased by a semipermeable membrane. In the present case, the concentration of solids in the entering stream of water can be lowered and can be increased in the exiting stream with the lowest degree of purity. That medium in which the concentration can be increased may be exposed to a pressure which must be higher than the pressure which is obtained by the osmotic demand for concentration equalization. This may result in a migration of particles against the direction of propagation. In the present process, the resulting purified first

stream may preferably be once again subjected to a second reverse osmosis, in order to obtain the stream with the highest and the stream with the medium degree of purity.

5 Parts of the stream with the medium degree of purity also can be obtained by an ion exchanger upstream of the reverse osmosis. The two partial streams with the medium degree of purity from ion exchanger and reverse osmosis subsequently may be mixed.

10 Before separating the three streams and/or between the separation of the stream with the lowest degree of purity, whose water content may lay below 89 wt-%, preferably below 50 wt-% and particularly preferably below 30 wt-%, and the reverse osmosis further purification steps, such as a denitrification, a nitrification and/or a removal of the organic compounds can be provided.

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Denitrification is understood to be the conversion of the nitrogen bound in the nitrate (NO_3^-) to molecular nitrogen (N_2) by certain heterotrophic and some autotrophic bacteria which are bound to a membrane. In this process, which serves the bacteria to generate energy, various oxidizable substances (electron
20 donors), such as organic substances, hydrogen sulfide (H_2S) and molecular hydrogen (H_2), can be oxidized with nitrate as oxidant (oxidizing agent) in the absence of molecular oxygen (O_2) (anoxic conditions).

Nitrification designates the bacterial oxidation of ammonia (NH_3) to nitrate
25 (NO_3). It consists of two coupled partial processes: In the first part ammonia is oxidized to nitrite, which in the second partial process is oxidized to nitrate.

A removal of the organic compounds preferably may be effected by the anaerobic treatment with bacteria in an oxygen-free environment.

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Furthermore, it was found to be advantageous in some embodiments to utilize the steam generated in the steam generation inside the steam supply system for the gasification process, e.g. for preheating educts, e.g. in distillation processes and/or to use the steam for generating electric energy, e.g. for operating a turbine. Thus, the water demand of the process can be reduced.

In particular when the gasification is effected in a fixed-bed reactor, the ash obtained there should be flushed out. For this purpose, the stream of water with the medium degree of purity may be used in accordance with a development of an embodiment of the invention.

In a fixed-bed gasification, this ash can be obtained by reaction of the carbonaceous solids such as coal or biomass and falls through a grate provided in the bottom region of the fixed bed. For the further transport of the ash, water can be introduced and the ash thus is flushed out.

In a gasification process which is carried out in a fluidized bed, it was found to be particularly favorable in some embodiments to also recirculate the stream with the medium degree of purity into the reactor.

In entrained-flow gasification it is recommendable in some embodiments to admix the stream with the medium degree of purity either to the educts introduced into the reactor as slurry and/or after quenching to the slag/waste water stream.

Furthermore, it may be necessary to subject that stream of water which has the highest degree of purity and/or that stream of water which has the medium degree of purity to a further purification before recirculation into the steam generation or the further ash treatment. Such further purification may provide for

largely recirculating the streams of waste water even when a high amount of soiling is introduced into the process by the solid starting material used.

Possible processes for the further purification may be chemical processes such as the Fenton reaction (an oxidation of organic substrates with hydrogen peroxide in an acidic medium catalyzed by iron salts), ozonization (sterilization by introducing ozone), the use of activated carbon (as adsorbent) and/or the addition of calcium hydroxide (for lowering the water hardness by ion exchange). The use of further precipitating or coagulating agents may also be conceivable. In addition, separators and/or a sewage treatment plant can also be used.

As set forth above, the solids are gasified in a fixed bed.

In a fixed-bed gasification it was found to be favorable in some embodiments to introduce that stream of water which has the lowest degree of purity above the fixed bed, wherein the stream is injected as finely distributed as possible.

When an entrained-bed reactor is used, it may be conceivable to feed the stream of water with the lowest degree of purity directly through the supply conduits into the burner flame. In a fluidized-bed reactor an atomization above the fluidized bed can also be considered. When the fuel is supplied as slurry in entrained-flow gasification, it is recommendable in some embodiments to admix the stream to this slurry before entry into the reactor region.

As solid matter, coal can be used on the one hand. Coal gasification processes have been practiced for decades. On the other hand, biomass also can serve as starting material, whereby renewable raw materials can be converted to synthesis gas. In particular in the gasification of biomass the above process may be of interest, since major amounts of non-burnt materials can be discharged by the stream of water.

Another aspect provides a plant for the gasification of a carbonaceous solids, such as for carrying out the process as set forth above, with a gasification fixed bed reactor configured so that the solids are at least partly converted to CO and H₂, with a separating device configured so that a liquid stream containing water is separated from the raw synthesis gas obtained by the gasification, and with a water purification device configured so that the stream containing water is purified, wherein the water purification device is designed such that the stream containing water is separated into three streams with different degrees of purity, and wherein the water purification device is connected via a conduit with a steam generator and/or via a conduit with a solids slurring, or a conduit for a slurry opening into the reactor and/or via a conduit with the gasification reactor, and wherein solids slurring is configured to flush out the ash from the solids gasification. An embodiment provides a plant that comprises a gasification reactor in which the solids are at least partly converted to carbon monoxide and hydrogen, a separating device in which the raw gas is separated from a liquid, aqueous stream, and a water purification device in which the aqueous liquid stream obtained in the separating device is purified. In the water purification the water is separated into three streams with different degrees of purity. Via a first conduit, the stream of water which has the highest degree of purity may be supplied to a water inlet of a steam generation; via a second conduit, that stream of water which has the medium degree of purity may be supplied to a means for the further treatment of ash from the gasification reactor; and/or via a third conduit that stream of water which has the lowest degree of purity may be guided back into the gasification reactor.

Advantageously, the device for separating the gaseous fraction from the liquid stream may be designed either as condenser or as droplet separator. The design as condenser may have the advantage that at the same time the gas stream is cooled further. When a droplet separator is used, the gas stream can

already be cooled previously and the thermal energy withdrawn in the cooling can be utilized at some other point.

5 The plant according to an embodiment of the invention may advantageously also include a gas cooling between the gasification reactor and the separating device, which can be recommendable in particular when the gas-liquid separator is formed as droplet separator and thus cooling can be effected at some other point in the process. Furthermore, the plant according to an embodiment of the invention may preferably include an ammonia recovery device between the
10 separating device and the further ash treatment.

In the gas-liquid separation, the contained liquid stream from the gas cooling may be separated further by decanting, wherein substantially tars, oils, phenols and ammonia (NH₃) are separated.

15 A further purification of the water can be effected by a downstream Phenosolvan[®] process. In the Phenosolvan[®] process the phenol-containing water may be intimately mixed with Phenosolvan[®] in a multistage extractor according to the mixer-separator principle. After subsequent phase separation, the largest part of
20 the phenols may be present in the solvent. This process may be repeated several times, wherein the phenol-containing water and the solvent can be guided in counterflow. The solvent may be separated from the phenols by distillation and flows back into the extractor for again washing out the phenols.

25 After passing the Phenosolvan[®] process, a CLL process (Chemie Linz-Lurgi[®]) may be conducted. In this process, acid gases and ammonia can be removed from the condensate of the Phenosolvan[®] process by selective stripping.

30 Due to this design of the plant, in particular with increasingly restrictive environmental protection requirements, the investment costs and the operating costs

can be lowered distinctly, since an expensive purification of the wastewater to be disposed of can wholly or partly be omitted. By introducing the stream carrying the main fraction of the solids into the gasification back, it can be omitted to separate the solids from the water with an energy-expensive drying operation.

5

When the stream of water with the medium degree of purity is used for flushing out the ash, additional fresh water cost and further treatment cost of water with medium degree of purity can be saved.

10

Further features, advantages and possible applications of embodiments of the invention can also be taken from the following description of an exemplary embodiment and the drawings. All features described or illustrated form the subject-matter of embodiments of the invention per se or in any combination, independent of their inclusion in the claims or their back-references.

15

In the drawings:

Fig. 1 shows a flow diagram of a conventional gasification process with waste water aftertreatment according to the prior art;

20

Fig. 2 schematically shows a flow diagram of a process according to an embodiment of the invention.

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In the conventional process as shown in Fig. 1, solids are introduced into a gasification reactor 10 via conduit 1 and oxygen is introduced via conduit 2. Via the same or a non-illustrated further conduit steam is fed into the reactor 10. Via conduit 14, the gas mixture formed by the reaction is supplied from the reactor 10 to a gas cooling 20. From this gas cooling, the raw synthesis gas obtained is withdrawn via conduit 21. Via conduit 22, the liquid stream obtained is supplied to a gas/liquid separation 23. From there, it is introduced into an

30

ammonia recovery 25 via conduit 24. Between the gas/liquid separation 23 and the ammonia recovery 25, a Phenosolvan[®] process can be provided (not shown in Fig. 1)

5 From the ammonia recovery 25, the liquid stream containing water is transferred into the water treatment 30 via conduit 26. From said water treatment, aqueous waste water on the one hand is discharged via conduit 31 and possibly treated such that the waste waters can be disposed of. Via conduit 32 a stream is withdrawn, which contains a large part of the solid particles and therefore is also referred to as slag stream. The stream containing solids is supplied to a drier 33 in which the contained water is evaporated by supplying energy and escapes into the atmosphere. The dried slag then for example can be brought to a disposal site.

15 From the gasification reactor 10, in particular when it is a fixed-bed gasification, the ash is discharged via conduit 11 and supplied to a further ash treatment 12. To transport the solid ash, the ash stream is made flowable by means of so-called make-up water. Via conduit 40, the make-up water is fed from a source into the further ash treatment 12. The flowable ash then is withdrawn via conduit 20 13.

In this process, the water obtained from the waste water aftertreatment is disposed of as waste water, but not recirculated into the process. Instead, fresh water is fed in at some other point of the process.

25 Fig. 2 shows the configuration of the process according to an embodiment of the invention in a flow diagram, wherein the solids to be gasified likewise are supplied to the gasification reactor 10 via conduit 1 and oxygen is supplied via conduit 2. From the gasification reactor 10 ash is withdrawn via conduit 11 and

supplied to a further ash treatment 12. From this further ash treatment, the ash then is withdrawn via conduit 13.

5 In the illustrated embodiment of the invention, the gasifier 10 is designed as fixed-bed reactor and includes a substantially cylindrical vertical reactor with external water jacket. The coal or biomass is introduced from above through a sluice into the solids distributor present in the interior of the reactor, whereby a fixed bed is formed, which rests on a rotary grate arranged in the lower region of the reactor 10. From this lower region oxygen and steam are also injected. Due to the ascending hot gases drying of the employed coal or biomass as well as desorption of the physisorbed gases takes place in the upper part of the gasifier 10. Below the drying zone the reaction zone is located, in whose upper part degassing of the coal or biomass takes place. Inside the reaction zone, degassing is followed by the actual gasification of the coal or biomass according to the Boudouard reaction. In the succeeding bottommost zone, the combustion of the coal or biomass as well as the water gas reaction and the water gas shift reaction take place. The resulting ash falls through the grate and is further discharged from there. The hot gases, which are guided in counterflow to the coal or biomass falling in from above, are withdrawn via a gas vent provided above the fixed bed.

20 Via conduit 14, the gas mixture obtained by the gasification reaction is withdrawn from the reactor 10 and supplied to a cooler 20. Due to cooling a raw synthesis gas is obtained, which is withdrawn via conduit 21. Via conduit 22, a liquid stream containing water flows into the gas-liquid separator (separating device) 23. It is of course also conceivable that the separation of gas and liquid exclusively is effected by the condensation in the gas cooler 20.

30 From the gas-liquid separator 23, which may be formed as condenser or as droplet separator, the liquid stream obtained is recirculated to the ammonia

recovery 25 via conduit 24. Between the gas/liquid separation 23 and the ammonia recovery 25, a Phenosolvan[®] process can be provided (not shown in Fig. 2). From the ammonia recovery 25, conduit 26 then leads into the water treatment 30. The aqueous stream obtained there is divided into three streams.

5 That stream which has the highest degree of purity is supplied via conduit 31 to a non-illustrated steam generation. The steam generated there can then either be used as heat carrier in the actual gasification process, e.g. for heating the educts, or be used for energy generation in a downstream turbine. In principle it is also conceivable to feed the stream of water as coolant into a cooling circuit.

10 Possibly, a further non-illustrated purification of this stream will be necessary.

Via conduit 37, the stream with the medium degree of purity is guided to the further ash treatment 12 and there serves as fluidizing agent. The introduction of make-up water thereby can be omitted completely. Possibly, a further water

15 purification 38 can also be provided for this stream in conduit 37.

Via conduit 36, that stream which carries a large part of the solids finally is transported back into the gasification. When the reactor 10 is designed as fixed-bed reactor, it is recommended to spray the stream carrying solids onto the fixed

20 bed from above. Thus, an energy-intensive and hence expensive drying of the solids can be omitted. In addition, valuable products still contained can thus be supplied to the gasification.

With this process it may be possible to provide gasification for solids, in which

25 no waste waters are obtained.

It is to be understood that, if any prior art publication is referred to herein, such reference does not constitute an admission that the publication forms a part of the common general knowledge in the art, in Australia or any other country.

5

In the claims which follow and in the preceding description, except where the context requires otherwise due to express language or necessary implication, the word “comprise” or variations such as “comprises” or “comprising” is used in an inclusive sense, i.e. to specify the presence of the stated features but not to preclude the presence or addition of further features in various embodiments of the process or plant.

List of Reference Numerals

	1, 2	conduit
	10	gasification reactor
	11	conduit
5	12	further ash treatment
	13, 14	conduit
	20	cooler
	21, 22	conduit
	23	separating device
10	24	conduit
	25	ammonia recovery
	26	conduit
	30	water aftertreatment
	31, 32	conduit
15	33	solids drying
	34-37	conduit
	38	water aftertreatment
	40	conduit

Claims:

1. A process for the gasification of carbonaceous solids, wherein in a first process step the solids are at least partly converted to CO and H₂ in a gasification step to produce a gas stream, wherein in a second process step a liquid stream containing water is separated from the gas stream, and wherein in a third process step the stream containing water is subjected to a water purification,

wherein in the water purification the stream containing water is separated into three streams with different degrees of purity,

wherein a first stream of water, which has the highest degree of purity, is supplied to a steam generation,

wherein a second stream of water, which has the medium degree of purity, is supplied to a solids slurring,

wherein a third stream of water, which has the lowest degree of purity, is recirculated into the solids gasification,

wherein the solids slurring comprises flushing out the ash from the solids gasification, and

wherein the gasification of the solids is carried out in a fixed bed.

2. The process according to claim 1, wherein the second stream of water is supplied to a further ash treatment or to a slurry to be introduced into the gasification reactor.

3. The process according to claim 1 or 2, wherein the second and the third stream of water are mixed and together recirculated into the solids gasification.

4. The process according to any one of the preceding claims, wherein the third stream of water is separated by decanting and subsequently the first and the second stream of water are obtained by a reverse osmosis process.

5. The process according to any one of the preceding claims, wherein the steam generated in the steam generation is utilized for supplying the gasification process with steam and/or for generating electric energy.

6. The process according to any one of the preceding claims, wherein the first stream of water, which has the highest degree of purity, and/or the second stream of water, which has the medium degree of purity, is/are subjected to a further purification before being recirculated.

7. The process according to any one of the preceding claims, wherein the third stream of water, which has the lowest degree of purity, is sprayed into the solids gasification above the fixed bed.

8. A plant for the gasification of carbonaceous solids with a gasification fixed bed reactor configured so that the solids are at least partly converted to CO and H₂, with a separating device configured so that a liquid stream containing water is separated from the raw synthesis gas obtained by the gasification, and with a water purification device configured so that the stream containing water is purified, wherein the water purification device is designed such that the stream containing water is separated into three streams with different degrees of purity, and wherein the water purification device is connected via a conduit (31) with a steam generator and/or via a conduit (37) with a solids slurring, or a conduit for a slurry opening into the reactor and/or via a conduit (36) with the gasification

reactor, and wherein solids slurring is configured to flush out the ash from the solids gasification.

9. The plan according to claim 8, wherein the water purification device is connected via the conduit (37) for further ash treatment.

10. The plant according to claim 8 or 9, wherein the separating device is a condenser or a droplet separator.

11. The plant according to any one of claims 8 to 10, wherein the plant is for carrying out a process according to any one of claims 1 to 7.

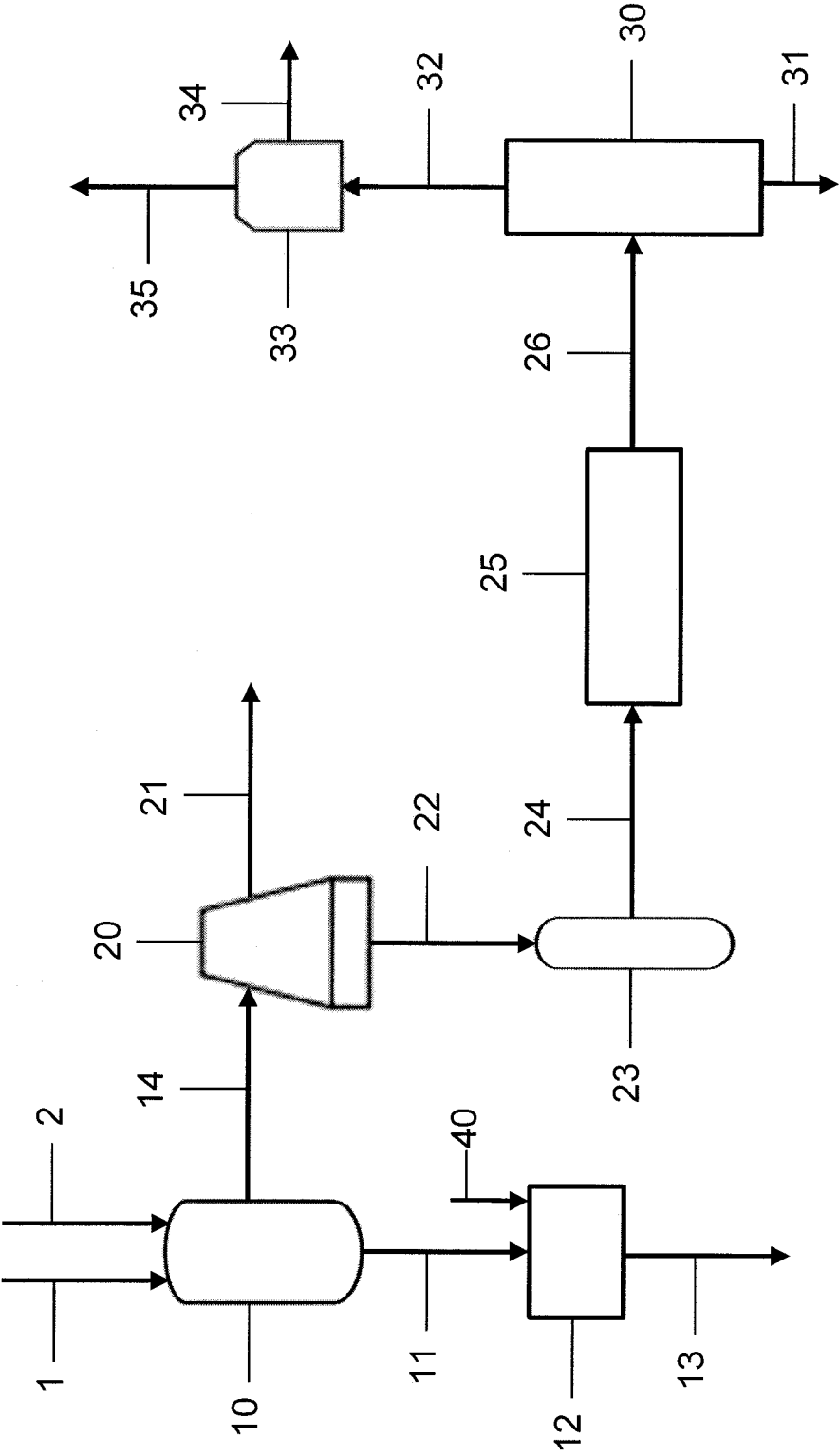


Fig. 1

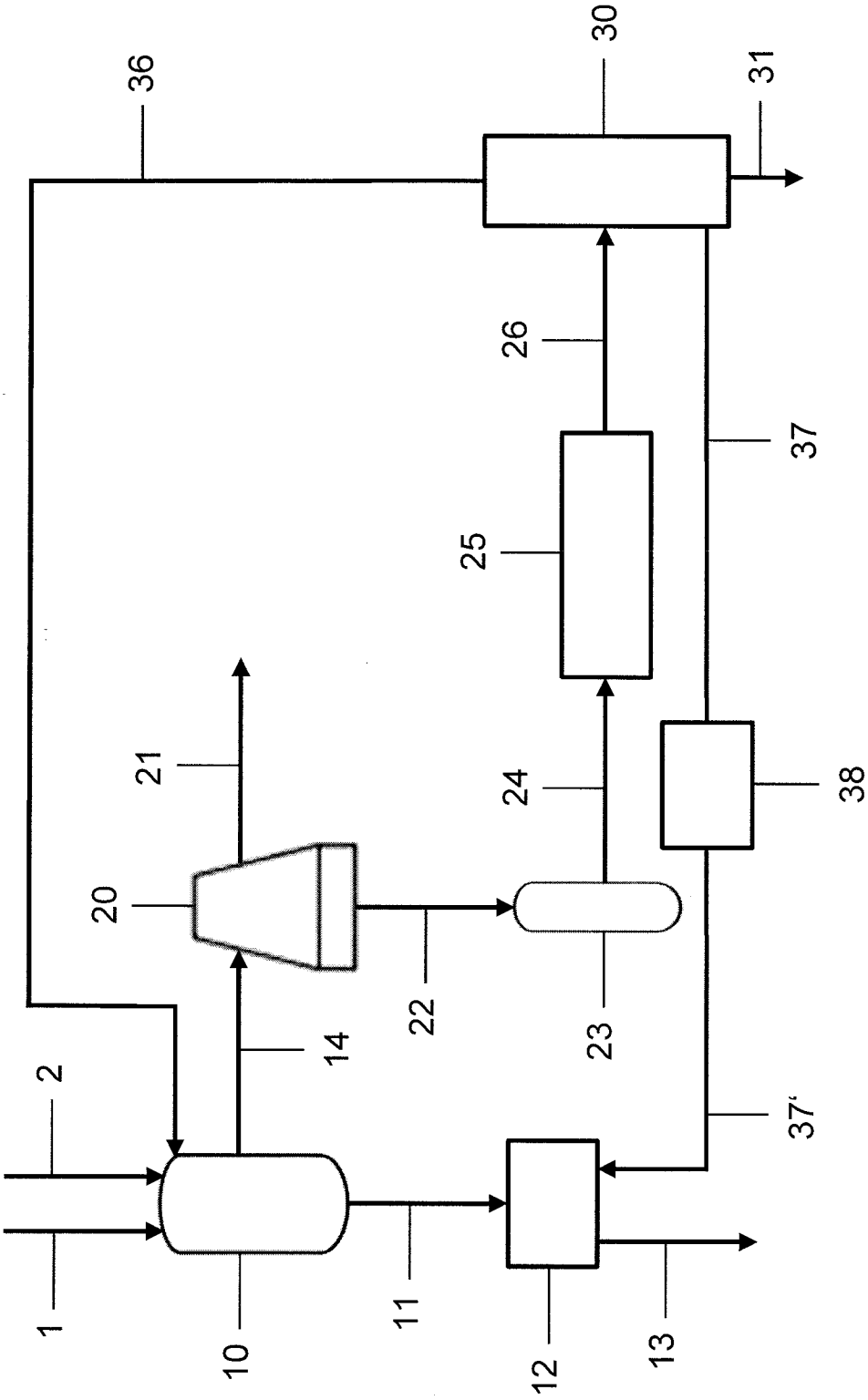


Fig. 2