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(54) Titre : ELIMINATION DU DIOXYDE DE CARBONE A PARTIR D'UN GAZ BRULE AVEC UN MILIEU COMPRENANT DE L'AMMONIAC
(54) Title: REMOVAL OF CARBON DIOXIDE FROM FLUE GAS WITH AMMONIA COMPRISING MEDIUM

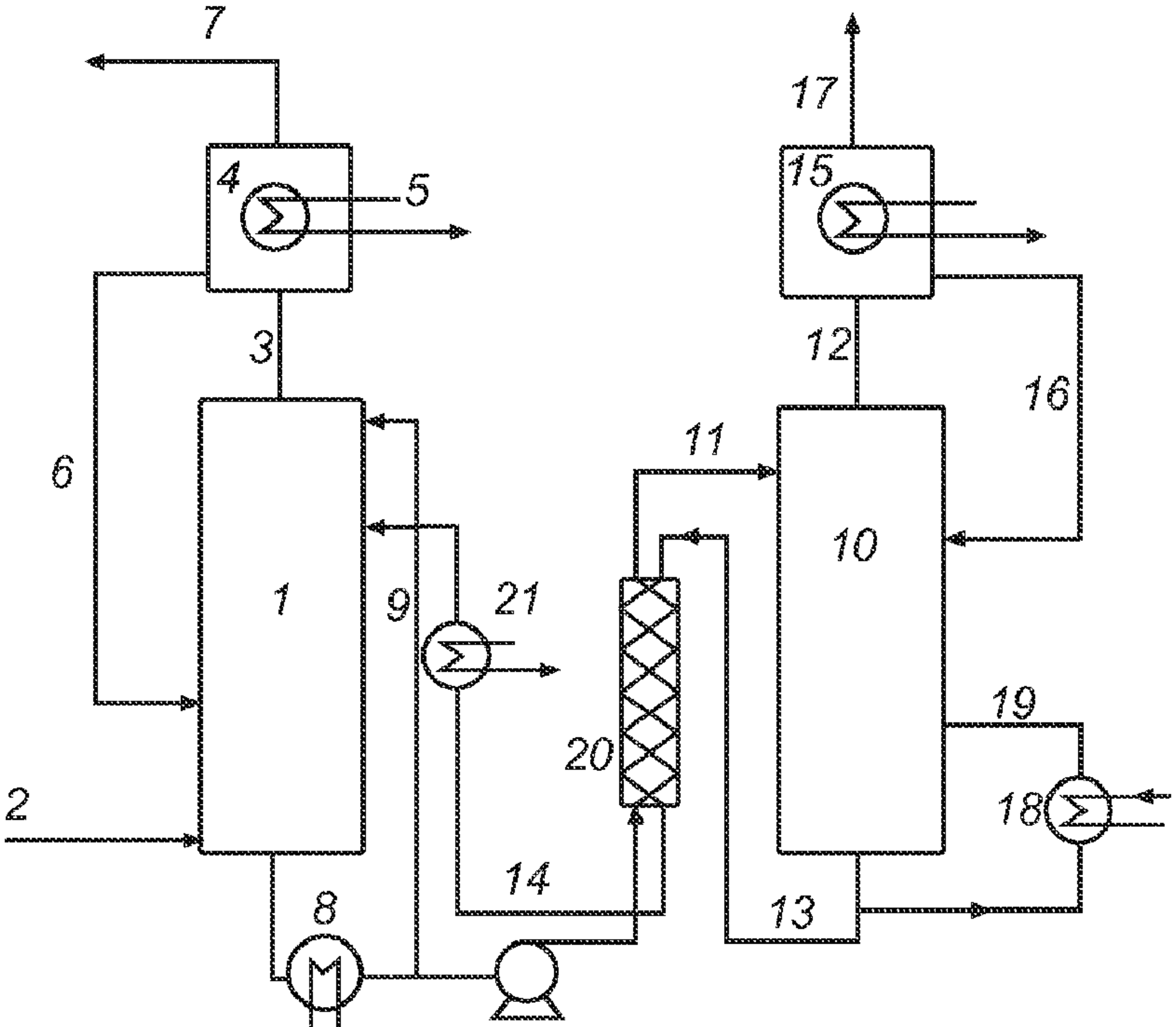

Fig. 2

(57) Abrégé/Abstract:
A process of removal of CO_2 from a flue gas (2). The process comprises the steps of: a) providing a flue gas comprising CO_2 (2), b) contacting the flue gas of step (a) with an ammonia-comprising medium (9), to absorb CO_2 from said flue gas; and c) condensing

(57) **Abrégé(suite)/Abstract(continued):**
ammonia (4) present in the flue gas leaving step (b), to remove ammonia from said flue gas. A system for removal of CO_2 from a flue gas. A system for removal of CO_2 from a flue gas. The system comprises a CO_2 absorber (1) receiving the flue gas (2) and comprising an ammonia-comprising medium (9). The system further comprises an ammonia condenser (4) receiving flue gas (3) leaving the CO_2 absorber.

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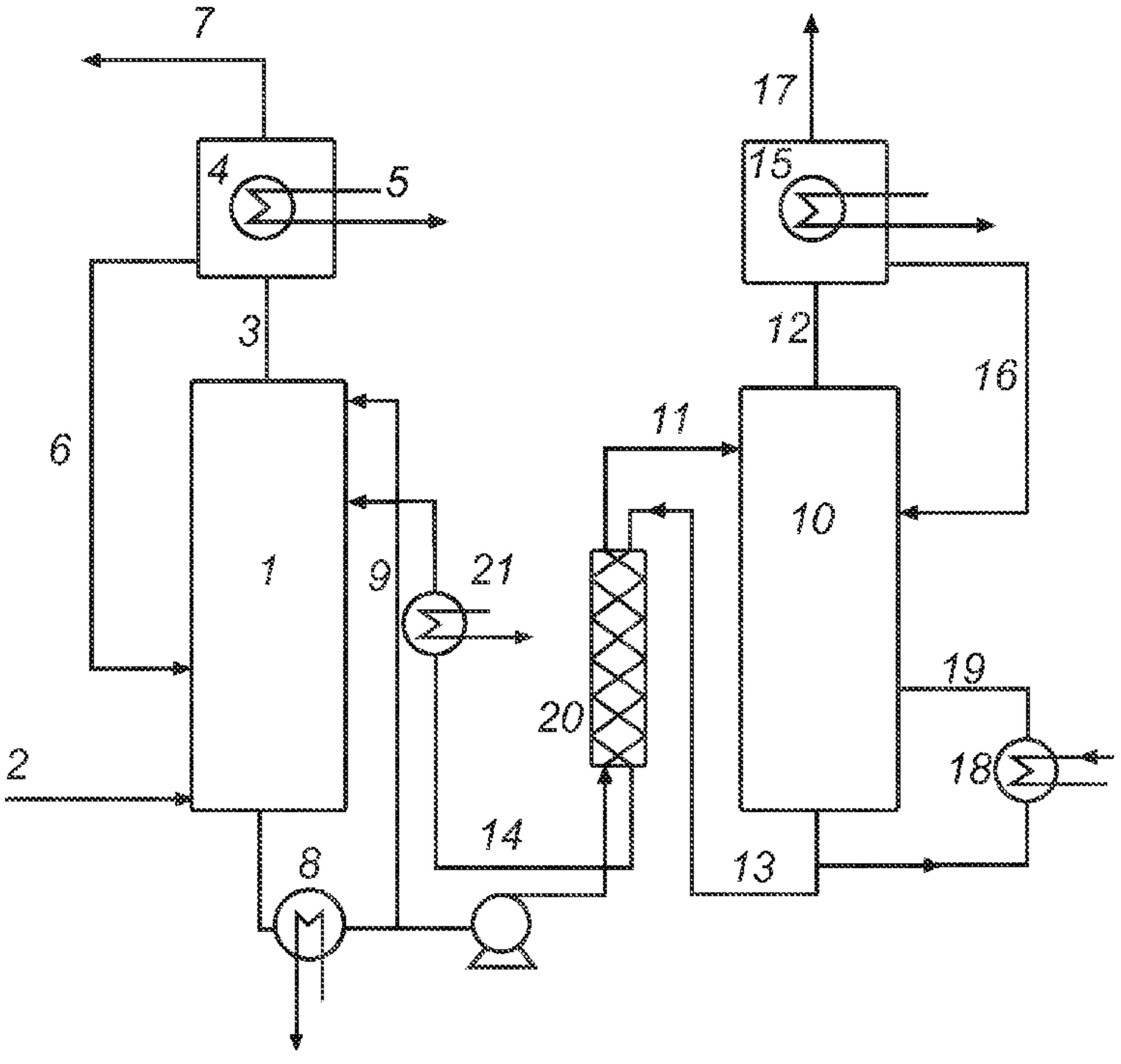

Fig. 2

(57) Abstract: A process of removal of CO_2 from a flue gas (2). The process comprises the steps of: a) providing a flue gas comprising CO_2 (2), b) contacting the flue gas of step (a) with an ammonia-comprising medium (9), to absorb CO_2 from said flue gas; and c) condensing ammonia (4) present in the flue gas leaving step (b), to remove ammonia from said flue gas. A system for removal of CO_2 from a flue gas. A system for removal of CO_2 from a flue gas. The system comprises a CO_2 absorber (1) receiving the flue gas (2) and comprising an ammonia-comprising medium (9). The system further comprises an ammonia condenser (4) receiving flue gas (3) leaving the CO_2 absorber.

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REMOVAL OF CARBON DIOXIDE FROM FLUE GAS WITH AMMONIA COMPRISING MEDIUM

Technical field

The present invention relates to a process of removal of CO_2 from a flue gas and to a system for removal of CO_2 from a flue gas. In said process and by said system, CO_2 is removed by absorption in an ammonia-comprising medium.

Background art

Environmental concern raises a demand for removal of carbon dioxide (CO_2) from, e.g., combustion gases, and subsequent processing or storage of the CO_2, to reduce emission to the atmosphere thereof. In known technologies for ammonia or ammonium based CO_2 capture, CO_2 is converted to ammonium carbonate or ammonium bicarbonate in dissolved or solid form. It is known to regenerate the ammonia or ammonium compounds used for CO_2 capture by release of CO_2 under controlled conditions.

Resnik, K.P. et al. (2004) Aqua ammonia process for simultaneous removal of CO_2, SO_2 and NO_x, *Int. J. Environmental Technology and Management,* Vol. 4, Nos. 1/2, pp. 89–104, discloses that the aqueous ammonia process can remove CO_2 and other contaminants that may exist in flue gas. Test results pertaining to the ammonia/carbon dioxide reaction in a semi-continuous reactor system are presented. Regeneration test results, including solution-cycling between the regeneration and absorption steps, are also presented.

WO 2006/022885 discloses cleaning of combustion gas to near zero concentration of residual contaminants followed by the capture of CO_2. The CO_2 is captured from cooled and clean flue gas in a CO_2 absorber utilizing an ammoniated solution or slurry in the NH_3–CO_2–H_2O system. Regeneration is accomplished by elevating the pressure and temperature of the CO_2-rich solution from the absorber.

It is, however, an ever existing desire to further improve CO_2 capture technologies in respect of, e.g., ammonia loss, power consumption, or chemical reaction rate.

Summary of the invention

An object of the present invention is to improve known ammonia or ammonium based technologies for CO_2 capture.

Accordingly, and depending on the operational and design parameters of a known technology for CO_2 capture, an object may reside in the reduction of energy and/or chemical consumption as well as in the reduction of investment and/or operation cost.

Additionally, an object may reside in the environmental, health and/or economical improvements of reduced emission of chemicals used in such a technology for CO_2 capture, e.g. in reduced ammonia slip.

In one aspect of the invention, the above-mentioned objects as well as further objects, which will become apparent to a skilled man after studying the description below, will be achieved by a process of removal of CO_2 from a flue gas, comprising the steps of: (a) providing a flue gas comprising CO_2; (b) contacting the flue gas of step (a) with an ammonia-comprising medium, to absorb CO_2 from said flue gas; and (c) condensing ammonia present in the flue gas leaving step (b), to remove ammonia from said flue gas.

In another aspect of the invention, said objects will be achieved by a system for removal of CO_2 from a flue gas, comprising a CO_2 absorber comprising an ammonia-comprising medium, to absorb CO_2 from said flue gas; and an ammonia condenser receiving the flue gas leaving the CO_2 absorber, to remove ammonia from said flue gas.

Thus, by condensation of ammonia present in the flue gas after absorption of CO_2 in an ammonia-comprising medium it is achieved an improved technology for CO_2 capture.

A flue gas may typically result from combustion of organic material such as renewable or non-renewable fuels. However, in the present context the term “flue gas “ may refer to any gas mixture comprising CO_2. Should a flue gas to be treated according to the present invention comprise chemical species or particles detrimental to the absorption of CO_2 in an ammonia-comprising medium, or to other features of the present invention, such species or particles may be initially removed by separation technologies known to a skilled man. Examples of such pre-treatments are given in, e.g., WO 2006/022885 referred to above.

As used herein, ammonia-comprising medium is any medium used to absorb CO_2, which includes ammonia, ammonium, or any compounds or mixtures comprising ammonia or ammonium. As an example, the CO_2

absorption may take place in an aqueous medium where the ammonia can be in the form of ammonium ion, NH_4^+, or in the form of dissolved molecular NH_3. Contacting of the flue gas comprising CO_2 with an ammonia-comprising medium results in formation of ammonium carbonate or ammonium bicarbonate in dissolved or solid form. In other words, as often used in the art, CO_2 is absorbed by the ammonia-comprising medium and thus removed from the flue gas. The ammonia-comprising medium of the present invention may be prepared by dissolution or mixing of ammonia or an ammonium compound such as ammonium carbonate in water. The term “medium” refers to a solution as well as to a suspension or slurry.

Ammonia present in the CO_2 depleted flue gas after CO_2 absorption, e.g. ammonia carried over from the ammonia-comprising medium, may be removed from the flue gas by condensation. Such condensation may take place in a condenser or scrubber, e.g. by acid or water wash, or by other direct contact or indirect contact heat exchange. Condensation can be performed at temperatures close to 0 °C, such as 0 to 5 °C, and at pressures up to 50 bar, such as 0 to 10 bar.

Step (b) of the inventive process or the CO_2 absorber of the inventive system operates within a wide temperature range. Practically, the lower temperature limit is set by the freezing point of the ammonia-comprising medium. On the other hand, the upper temperature limit is for practical reasons set by the boiling point of the ammonia-comprising medium. It is to be understood that the references to freezing point and boiling point are made at the operating pressure of step (b) or the CO_2 absorber. At pressures close to atmospheric pressure, the upper and lower temperature limits may be close to 0 and 100 °C, respectively, whereas temperatures above 100 °C may well be practical and preferred at higher pressures. A pressure range of 0 to 10 bar, preferably 5 to 10 bar, is contemplated. A higher pressure in the absorber increases both the solubility of the carbon dioxide in the medium and the residence time of the flue gas in the absorber, resulting in a smaller absorber size. A higher pressure also decreases the partial pressure of ammonia.

However, to achieve favourable mass transfer and chemical reaction rate, it may be desirable to operate at a temperature higher than the lower limit. Accordingly it is suggested to operate step (b) or the CO_2 absorber at a temperature higher than 20 °C, preferably higher than 38 °C, more preferably higher than 40 °C, most preferably higher than 50 °C.

To reduce loss of ammonia (due to evaporation) and/or energy (in the form of evaporating steam), it may be desirable to operate at a temperature lower than the upper limit. Accordingly it is suggested to operate step (b) or the CO_2 absorber at a temperature lower than 80 °C, preferably lower than 60 °C. In some applications, e.g. when the flue gas provided is of very low temperature, it is suggested to operate step (b) or the CO_2 absorber at a temperature lower than 16 °C, preferably lower than 15 °C, more preferably lower than 5 °C.

A temperature range of 20 to 35 °C is contemplated for operation of step (b) or the CO_2 absorber.

Step (c) of the inventive process or the ammonia condenser of the inventive system may operate by indirect cooling of the gas leaving step (b) or the gas leaving the CO_2 absorber, respectively. In indirect cooling, the cooling medium is physically separated from the gas to be cooled. Thus, condensed ammonia will not be diluted by cooling medium but only with any component, e.g. water, present in the flue gas and condensed together with said ammonia. A low temperature enhances the solubility of the ammonia in the condensed water. Such an arrangement favourably allows for recovery of condensed ammonia at a higher concentration than should it additionally have been diluted with cooling medium. Further utilisation of the recovered ammonia is thus facilitated.

Ammonia condensed in step (c) of the inventive process or the ammonia condenser of the inventive system may be returned to step (b) or the CO_2 absorber, respectively. Thus, the need for make-up ammonia or ammonium as well as the amount of ammonia emitted to the environment is reduced.

The inventive process may comprise the further step (d) of releasing CO_2 from the medium resulting from step (b), to regenerate the ammonia-comprising medium. Correspondingly, the inventive system may further comprise a medium regenerator, to release CO_2 from the medium resulting from the CO_2 absorber. Released CO_2 may optionally be further processed or stored as suitable in view of technical, economical or environmental concerns.

Step (d) of the inventive process or the medium regenerator of the inventive system operates within a wide temperature range. It is desirable to operate at a temperature in the range of 100 to 200 °C, preferably 110 to 160 °C. Thus, the release of CO_2 is performed at a temperature allowing efficient release of CO_2.

Step (d) of the inventive process or the medium regenerator of the inventive system operates within a wide pressure range. It is desirable to operate at a pressure higher than atmospheric pressure, preferably higher than 10 bar. Due to the high regeneration pressure, the ammonia formed during regeneration is captured in the medium from which CO_2 is released. Thus, release, or loss, of ammonia is avoided.

The inventive process may comprise the further step (e) of returning the ammonia-comprising medium regenerated in step (d) to step (b). Correspondingly, the inventive system may further comprise a passage from the medium regenerator to the CO_2 absorber, to return regenerated ammonia-comprising medium to the CO_2 absorber. Thus, it has been created an integrated process or system allowing for continuous or semi-continuous removal of CO_2 from a flue gas by absorption in an ammonia-comprising medium, recovery of CO_2, and regeneration and recycling of said medium. Ammonia emission from, and ammonia make-up to, the integrated process or system is reduced.

Brief description of the drawings

Figs. 1a and 1b are schematic representations of an ammonium based CO_2 capture system.

Fig. 2 is a schematic representation of an ammonium based CO_2 capture system.

Detailed description of embodiments

Figs. 1a and 1b are schematic representations of an ammonium based CO_2 capture system. The system comprises a CO_2 absorber 1. In all embodiments, CO_2 absorber 1 may be arranged as a plurality of vessels or operational steps in parallel or in series. Flue gas 2, from which CO_2 is to be removed, is fed to CO_2 absorber 1. In CO_2 absorber 1 the flue gas is contacted with ammonia-comprising medium, e.g. by bubbling the flue gas through said medium or by spraying the medium into the flue gas. It is within the knowledge of a skilled man to arrange for contacting of flue gas with ammonia-comprising medium. In CO_2 absorber 1, CO_2 from flue gas 2 is absorbed in the ammonia-comprising medium, e.g. by formation of carbonate or bicarbonate of ammonium either in dissolved or solid form. Flue gas depleted of CO_2 leaves CO_2 absorber 1 via line 3. As used herein, ammonia-comprising medium is any medium used to absorb CO_2, which includes ammonia, ammonium, or any compounds or mixtures comprising ammonia or ammonium. As an example, the CO_2 absorption may take place in an aqueous medium where the ammonia can be in the form of ammonium ion, NH_4^+, or in the form of dissolved molecular NH_3.

The system illustrated in Figs. 1a and 1b further comprises an ammonia condenser 4. In all embodiments, ammonia condenser 4 may be arranged as a plurality of vessels or operational steps in parallel or in series. Ammonia condenser 4 is an indirect cooler having a cooling medium circulation 5. Via line 3, flue gas from CO_2 absorber 1 enters ammonia condenser 4. In ammonia condenser 4, ammonia present in the flue gas is condensed, e.g. on the heat exchange surfaces separating cooling medium of cooling medium circulation 5 and flue gas to be treated. Condensed ammonia, typically dissolved in water which may also have condensed from water vapour present in flue gas, leaves ammonia condenser 4 via line 6 and is returned to CO_2 absorber 1. Flue gas depleted of ammonia leaves ammonia condenser 4 via line 7.

The operating temperature of CO_2 absorber 1 is controlled by passing ammonia-comprising medium through a heat exchanger 8 and returning the medium to CO_2 absorber 1 via line 9. Heat exchanger 8 may, as desirable, heat or cool said medium. Heat exchanger 8 may be arranged on line 9, as illustrated in Fig. 1a, or in a vessel comprising CO_2 absorber 1, as illustrated in Fig. 1b.

Fig. 2 is a schematic representation of an ammonium based CO_2 capture system. The system comprises a CO_2 absorber, an ammonia condenser, a heat exchanger and related piping as described and numbered above. In Fig. 2, heat exchanger 8 is shown as in Fig. 1a but the alternate arrangement of Fig. 1b is also feasible.

The system illustrated in Fig. 2 further comprises a regenerator 10. In all embodiments, regenerator 10 may be arranged as a plurality of vessels or operational steps in parallel or in series. Ammonia-comprising medium, including dissolved or solid carbonate or bicarbonate of ammonium as formed in CO_2 absorber 1, enters regenerator 10 via line 11. In regenerator 10 the medium is exposed to temperature and pressure conditions sufficient to release CO_2 from the medium and to regenerate ammonia-comprising medium. Basically, carbonate or bicarbonate of ammonium either in dissolved or solid form is decomposed to release CO_2 as a gas. It is within the knowledge of a skilled man to obtain such conditions, e.g. utilising heat exchangers and pumps. Released CO_2 leaves regenerator 10 via line 12. Regenerated ammonia-comprising medium is returned to CO_2 absorber 1 via lines 13 and 14.

The system illustrated in Fig. 2 further comprises an ammonia recovery condenser 15, the purpose of which is to recover ammonia leaving regenerator 10 with released CO_2. In all embodiments, ammonia recovery condenser 15 may be arranged as a plurality of vessels or operational steps in parallel or in series. Ammonia recovery condenser 15 may be designed as described above for ammonia condenser 4. Via line 12, gas comprising CO_2 from regenerator 10 enters ammonia recovery condenser 15. In ammonia recovery condenser 15, ammonia present in the gas is condensed. Condensed ammonia, typically dissolves in water which is condensed from water vapour present in gas leaving regenerator 10. Dissolved ammonia leaves ammonia recovery condenser 15 via line16 and is returned to regenerator 10. CO_2 comprising gas depleted of ammonia leaves ammonia recovery condenser 15 via line 17.

The operating temperature of regenerator 10 is controlled by passing ammonia-comprising medium through a heat exchanger 18 and returning the medium to regenerator 10 via line 19. Heat exchanger 18 may be arranged on line 19, as illustrated in Fig. 2, or in a vessel comprising regenerator 10, similarly with the configuration of heat exchanger 8 in CO_2 absorber 1 in Fig. 1b.

As described above, in the system illustrated in Fig. 2, ammonia-comprising medium, including dissolved or solid carbonate or bicarbonate of ammonium, is fed from CO_2 absorber 1 to regenerator 10, whereas regenerated ammonia-comprising medium is fed from regenerator 10 to CO_2 absorber 1. The absorption process being exothermic and the regeneration process being endothermic, and said processes typically being operated at substantially different temperatures, means for heat recovery may improve the performance of the system. Thus, ammonia-comprising medium, including dissolved or solid carbonate or bicarbonate of ammonium, from CO_2 absorber 1 in line 11 is heat exchanged in an heat exchanger 20 with regenerated ammonia-comprising medium from regenerator 10 in lines 13 and 14.

Further means for temperature control of the system illustrated in Fig. 2 is represented by heat exchanger 21. Through heat exchanger 21, the medium in line 14 may be heated or cooled as desirable.

Example

A flue gas comprising 13 % by volume CO_2 is contacted with an ammonia/ammonium-containing solution in an absorber at ambient temperature, such as at approximately 25 °C. The pressure of the absorber is kept between 0 and 10 bar. A high pressure increases both the solubility of the carbon dioxide in the solvent and the residence time of the flue gas in the absorber, resulting in a smaller absorber size. A high pressure in the absorber decreases the partial pressure of the ammonia also. At the mentioned temperature the content of the CO_2 in the outgoing gas from absorber is expected to be below 2 % by volume, meaning an overall CO_2 removal efficiency close to 85 %. The removal efficiency can be improved by addition of promoters. Flue gas leaving the absorber passes an indirect cooler, reducing the gas temperature to a value between 0 and 5 °C. Thereby, water present in the flue gas leaving the absorber is condensed. The ammonia present in the gas is dissolved in the condensed water. Condensed water comprising ammonia is returned from the indirect cooler to the absorber. The flue gas stream leaving the indirect cooler may contain some ammonia. Depending on the operational temperature this value can vary between tens and hundreds of ppm. A low pH wash system can be used to clean the flue gas from trace ammonia.

Absorption of CO_2 into ammonia/ammonium containing solution involves the following chemical reactions:

$$NH_3\,(a) + H_2O = NH_4^+ + OH^- \qquad 1$$
$$2NH_4^+ + CO_2 + H_2O = (NH_4)_2CO_3\,(a) \qquad 2$$
$$(NH_4)_2CO_3\,(a) = NH_2CO_2^- + NH_4^+ + H_2O \qquad 3$$
$$(NH_4)_2CO_3\,(a) + CO_2 + H_2O = 2NH_4HCO_3\,(a) \qquad 4$$
$$NH_4HCO_3\,(a) = NH_4HCO_3\,(s) \qquad 5$$

If CO_2 available is in excess, the solution in the absorber becomes saturated by ammonium bicarbonate. Further reaction of the solution with CO_2 results in precipitation of the ammonium bicarbonate. The longer the residence time of the solution in the absorber the higher becomes the fraction of the solid ammonium bicarbonate in the solution.

During regeneration CO_2 is recovered from the saturated ammonium bicarbonate solution. Regeneration is done at high pressure, preferably above 10 bar. Decomposition of solid ammonium bicarbonate starts at 30 °C at atmospheric pressure, resulting in formation of ammonia, CO_2 and water. Decomposition of the ammonium bicarbonate in slurry requires a higher temperature, due to the excess energy needed to increase the temperature of the solution. It is expected that the decomposition of the ammonium bicarbonate should result in an equi-molar formation of the ammonia, water and CO_2. Regeneration at high pressure causes that the water stays in liquid form. Due to its higher solubility the formed NH_3 dissolves in the water, whilst the CO_2 can leave the system in gas form. The temperature of the regeneration is dependant on the fraction of the solid ammonium bicarbonate in the solution. At solid contents above 50 % by weight, the regeneration temperature can be close to 100 °C, whereas a temperature of 130 °C could be required for solid contents close to 15 % by weight.

CLAIMS

1. A process of removal of CO_2 from a flue gas, comprising the steps of:

(a) providing a flue gas comprising CO_2;

(b) contacting at a temperature lower than 16 °C the flue gas of step (a) with an ammonia-comprising medium prepared by dissolution or mixing of ammonium carbonate in water, to absorb CO_2 from said flue gas; and

(c) condensing ammonia present in the flue gas leaving step (b), to remove ammonia from said flue gas.

2. A process according to claim 1, wherein step (b) operates at a temperature lower than 15 °C.

3. A process according to claim 2, wherein step (b) operates at a temperature lower than 5 °C.

4. A process according to any one of the preceding claims, wherein step (c) is perfomed by indirect cooling of the flue gas leaving step (b).

5. A process according to any one of the preceding claims, wherein ammonia condensed in step (c) is returned to step (b).

6. A process according to any one of the preceding claims, comprising the further step of:

(d) releasing CO_2 from the medium resulting from step (b), to regenerate the ammonia-comprising medium.

7. A process according to claim 6, wherein step (d) operates at a temperature in the range of 100 to 200 °C.

8. A process according to claim 7, wherein step (d) operates at a temperature in the range of 110 to 160 °C.

9. A process according to any one of claims 6 to 8, wherein step (d) operates at a pressure higher than atmospheric pressure.

10. A process according to claim 9, wherein step (d) operates at a pressure higher than 10 bar.

11. A process according to any one of claims 6 to 10, comprising the further step of:

(e) returning the ammonia-comprising medium regenerated in step (d) to step (b).

12. A system for removal of CO_2 from a flue gas, comprising a CO_2 absorber (1), operating at a temperature lower than 16 °C, receiving the flue gas (2) and comprising an ammonia-comprising medium prepared by dissolution or mixing of ammonium carbonate in water; and an ammonia condenser (4) receiving flue gas (3) leaving the CO_2 absorber.

13. A system according to claim 12, wherein the CO_2 absorber (1) operates at a temperature lower than 15 °C.

14. A system according to claim 13, wherein the CO_2 absorber (1) operates at a temperature lower than 5 °C.

15. A system according to any one of claims 12 to 14, wherein the ammonia condenser (4) is an indirect cooler for the flue gas (3) leaving the CO_2 absorber (1).

16. A system according to any one of claims 12 to 15, further comprising a passage (6) from the ammonia condenser (4) to the CO_2 absorber (1), for returning condensed ammonia to the CO_2 absorber.

17. A system according to any one of claims 12 to 16, further comprising a medium regenerator (10), for releasing CO_2 from the medium resulting from the CO_2 absorber (1).

18. A system according to claim 17, wherein the medium regenerator (10) operates at a temperature in the range of 100 to 200 °C.

19. A system according to claim 18, wherein the medium regenerator (10) operates at a temperature in the range of 110 to 160 °C.

20. A system according to any one of claims 17 to 19, wherein the medium regenerator (10) operates at a pressure higher than atmospheric pressure.

21. A system according to claim 20, wherein the medium regenerator (10) operates at a pressure higher than 10 bar.

22. A system according to any one of claims 17 to 21, further comprising a passage (13, 14) from the medium regenerator (10) to the CO_2 absorber (1), for returning regenerated ammonia-comprising medium to the CO_2 absorber.

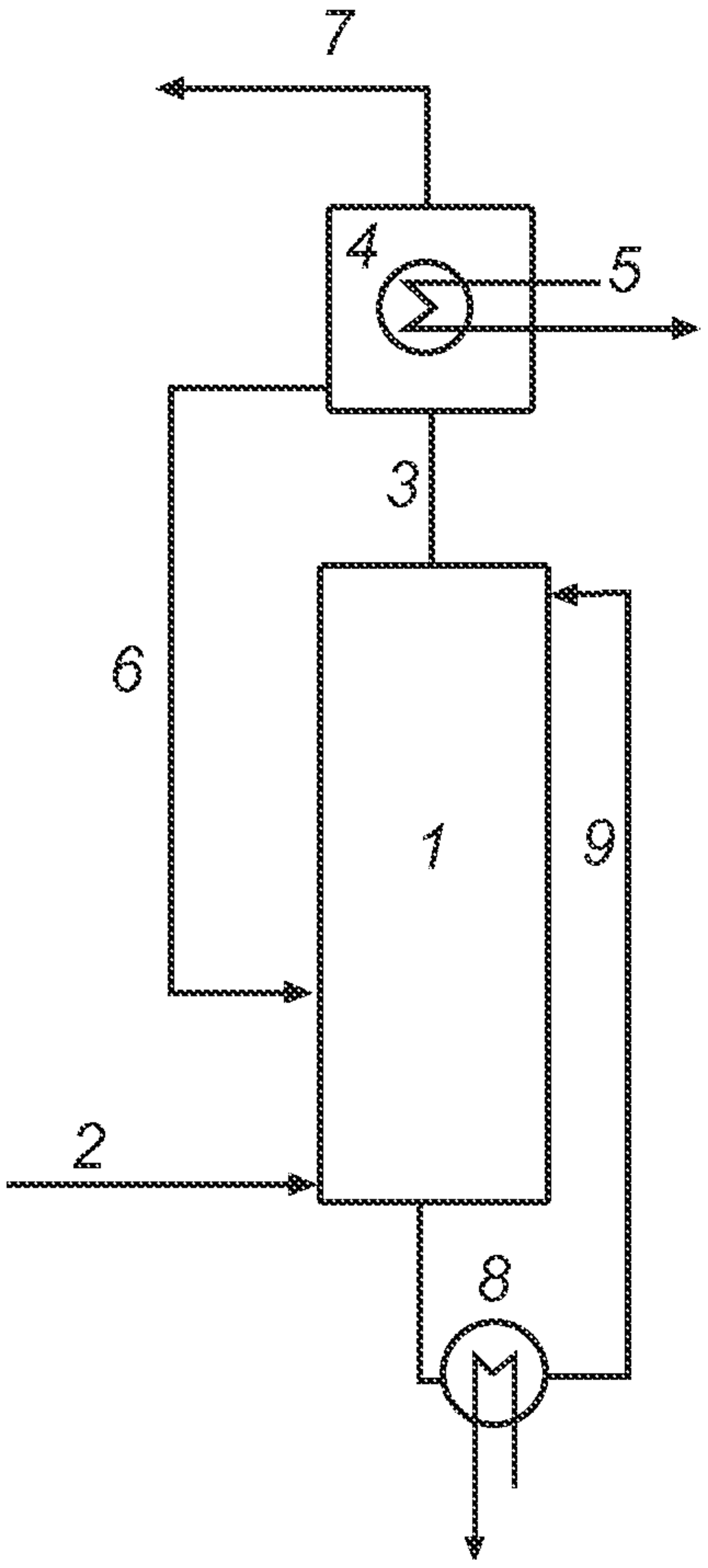

Fig. 1a

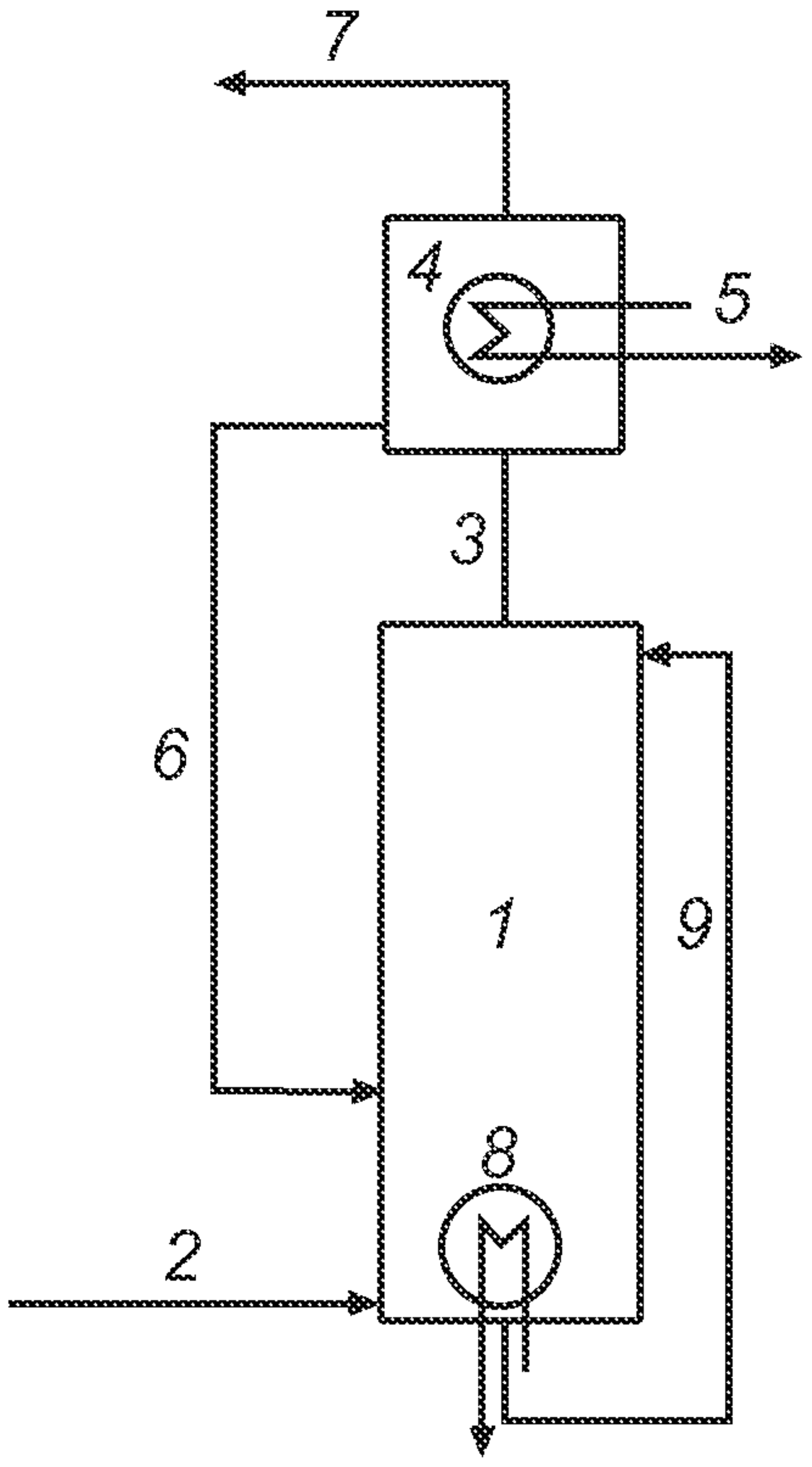

Fig. 1b

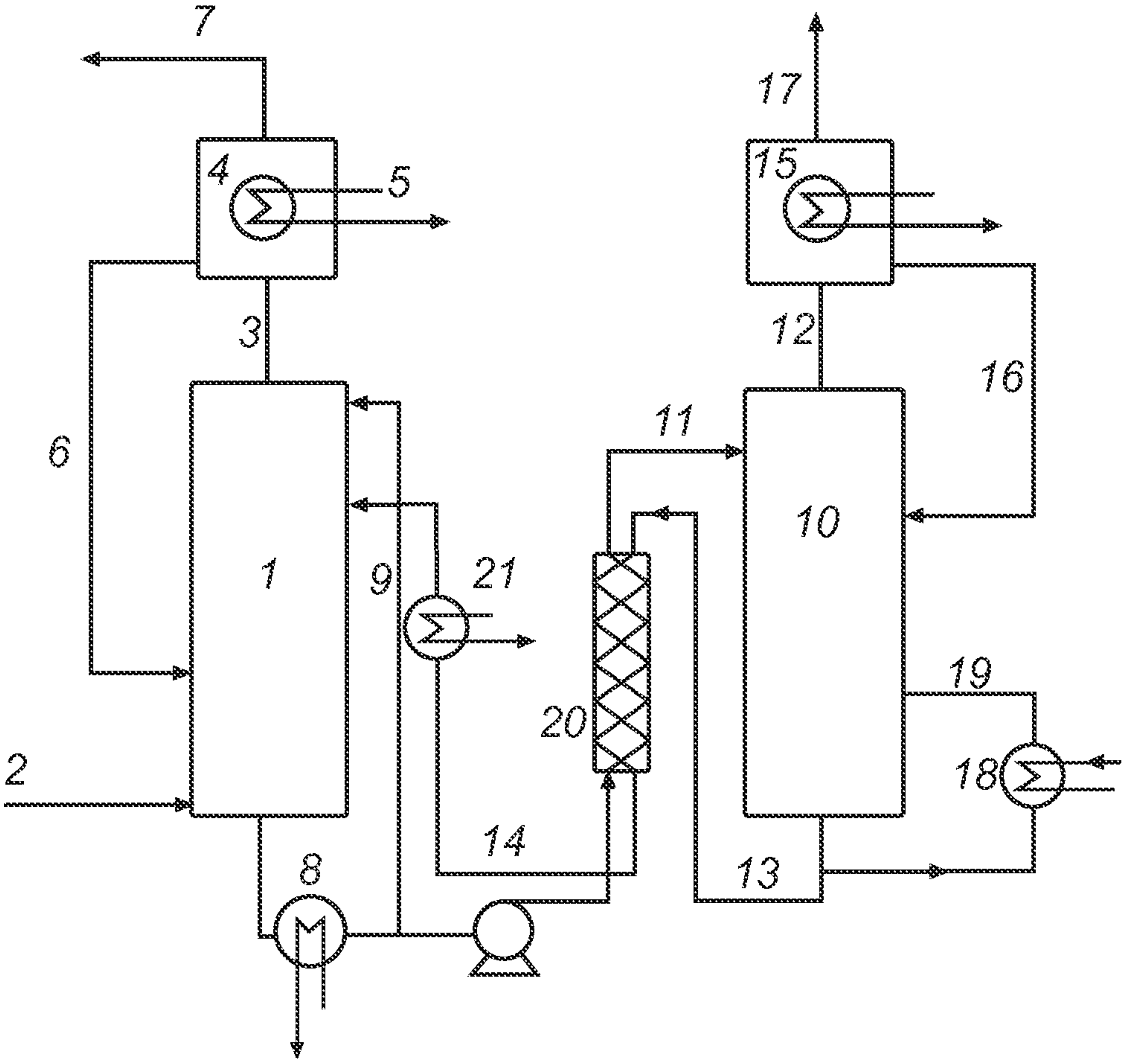

Fig. 2

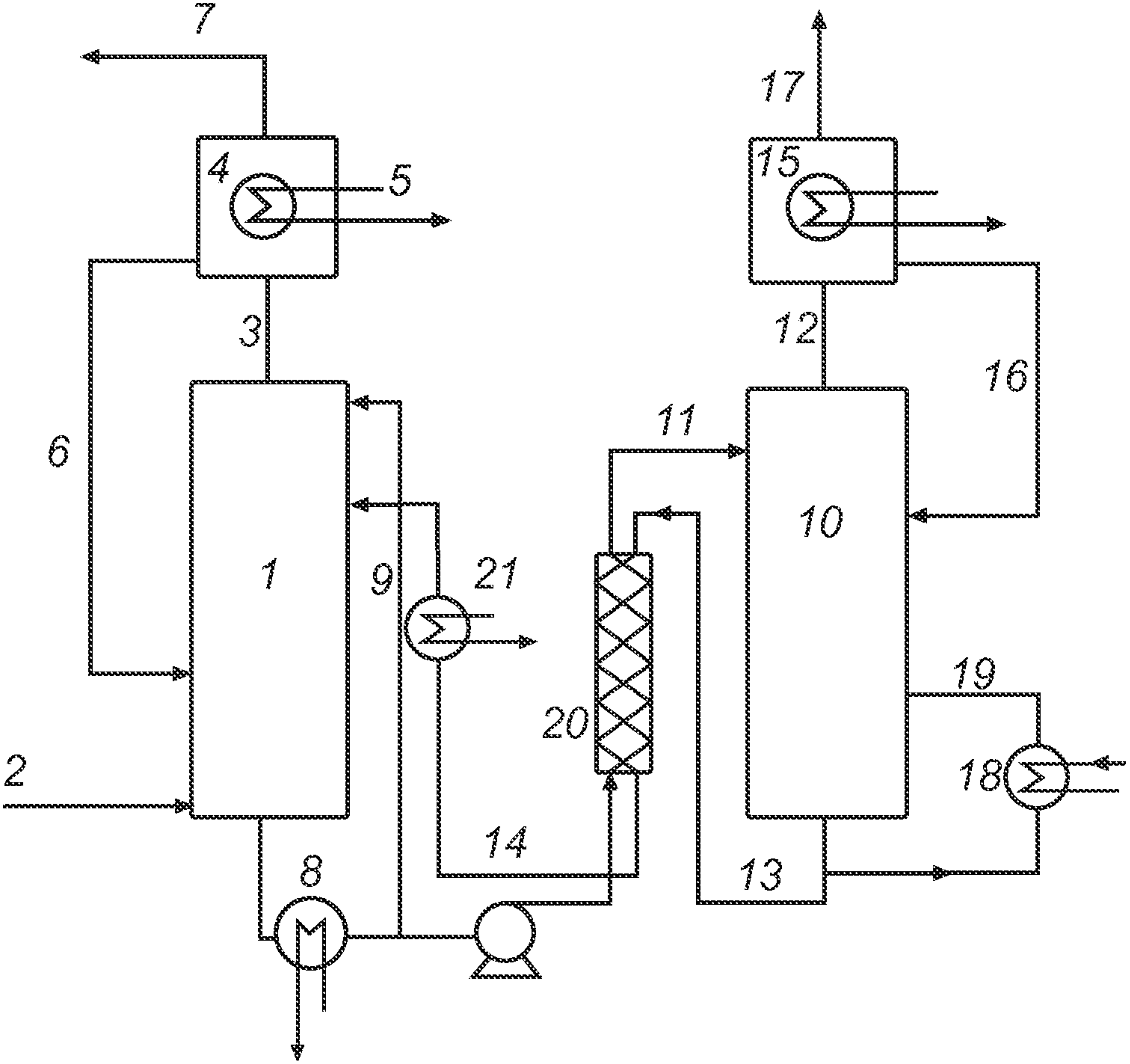

Fig. 2