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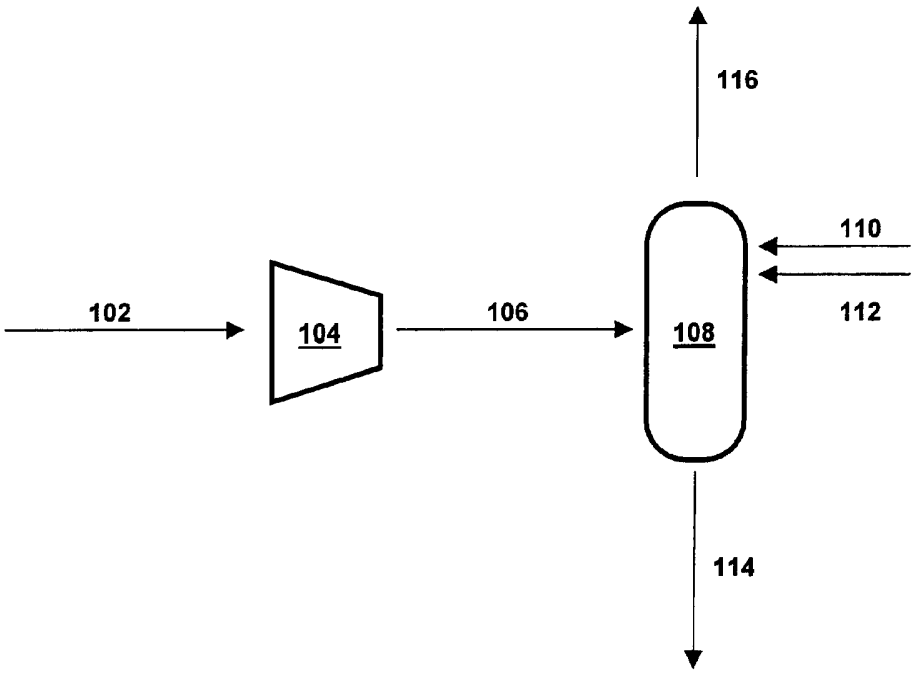
(54) **Title:** METHOD FOR OBTAINING AN AMMONIUM CARBAMATE SOLUTION FROM A GAS MIXTURE CONTAINING NH_3, H_2O AND CO_2



(57) **Abstract:** The invention relates to a method for obtaining an ammonium carbamate solution from a gas mixture that contains more than 40 wt.% NH_3, less than 50 wt.% CO_2 and less than 40 wt.% H_2O and has a pressure between 0.1 MPa and 4 MPa, comprising a compression step, in which the pressure of the gas mixture is increased to a pressure between 0.5 MPa and 25 MPa, and an absorption step.

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METHOD FOR OBTAINING AN AMMONIUM CARBAMATE SOLUTION FROM A GAS MIXTURE CONTAINING NH_3, H_2O AND CO_2

The invention relates to a method for obtaining an ammonium carbamate solution from a gas mixture containing NH_3, H_2O and CO_2.

Such a method is applied in processes for the preparation of melamine, such as in the Stamicarbon process as described in 'Melamine and Guanamines', par. 4.1.3 of Ullmann's Encyclopedia of Industrial Chemistry, Sixth Edition, 2001 Electronic Release. In the known process the gas mixture is obtained from a cooling zone in which the melamine-containing flow coming from a reactor is cooled. The gas mixture, which has a pressure of approximately 0.7 MPa, is subsequently subjected to an absorption step in which, besides an ammonium carbamate solution, a flow of gaseous NH_3 is also obtained. An ammonium carbamate solution is here and hereafter understood to mean an aqueous solution that, besides ammonium carbamate, can also contain free NH_3 and/or CO_2 and/or other compounds derived from NH_3 and CO_2, such as ammonium bicarbonate.

The flow of gaseous NH_3 recovered from the absorption step is used elsewhere in the melamine production process, as fluidization gas in the reactor. The Stamicarbon process further comprises a heat exchanger in which the gas mixture is partially condensed before it is subjected to the absorption step.

In many cases the ammonium carbamate solution obtained from the gas mixture is processed further for use as a raw material for the preparation of urea. The preparation of urea from ammonia and carbon dioxide via ammonium carbamate takes place at an elevated pressure, usually between 12.5 MPa and 25 MPa, via the following reaction scheme.

$2\ NH_3$	+	CO_2	$\leftrightarrow$	$H_2N\text{-}CO\text{-}ONH_4$	$\leftrightarrow$	$H_2N\text{-}CO\text{-}NH_2$	+	H_2O
ammonia		carbon dioxide		ammonium carbamate		urea		water

The further processing of the ammonium carbamate solution in particular relates to the release from the solution of a part of the NH_3, which is then recycled within the melamine production process, and to the reduction of the quantity of water in the solution.

A disadvantage of known processes such as the Stamicarbon process is that the aforesaid further processing requires much energy in the form of steam. Moreover, the further processing is technically complicated, which means it can only be realized at very high costs.

The object of the invention is to largely eliminate the said disadvantage.

This object is achieved in that the gas mixture, which contains more than 40 wt.% NH_3 less than 50 wt.% CO_2 and less than 40 wt.% H_2O and has a pressure between 0.1 MPa and 4 MPa, is compressed in a compression step prior to the absorption step, which involves increasing the pressure of the gas mixture to a pressure between 0.5 MPa and 25 MPa. Preferably the gas mixture has a pressure between 0.5 and 2.5 MPa before entering the compression step. The pressure of the gas mixture is increased in the compression step; preferably by at least 0.4 MPa, more preferably by at least 0.7 MPa. The pressure of the gas mixture is preferably increased to a pressure between 1 MPa and 5 MPa in the compression step, more preferably to a pressure between 1.5 MPa and 3 MPa, this in particular if the method according to the invention is incorporated in a process for the preparation of melamine. Alternatively, the pressure of the gas mixture is preferably increased to a pressure between 12.5 MPa and 25 MPa, this in particular if the method according to the invention is incorporated in a process for the preparation of urea.

Surprisingly, it has been found that in the method according to the invention the quantity of ammonia that is entrained via the ammonium carbamate solution, either as free ammonia or as ammonium ion, is lower than in the known method, so that no or at least less further processing is necessary. This advantage offsets the consumption of energy in the form of electricity or steam in the compression step.

A further advantage of the invention is that an ammonium carbamate solution, obtained from the gas mixture may contain a lower percentage of H_2O at a higher pressure without an – undesirable – solid phase being formed. The method according to the invention can therefore be applied to gas mixtures with a H_2O percentage that is so low that this would lead to problems due to the formation of solids

in the known processes. The gas mixture preferably contains more than 50 wt.% NH_3, between 10 and 30 wt.% CO_2 and less than 35 wt.% H_2O. The ammonium carbamate solution obtained from the gas mixture preferably has more than 10 wt. % water in order to prevent solid phase formation, in particular when the pressure of the solution is below 3 MPa.

Yet another advantage of the invention is that, if the pressure in the compression step is increased up to approximately 1.5 MPa, preferably 1.8 MPa or still higher, such as for example 2 MPa or more, the flow of gaseous NH_3 obtained from the absorption step can wholly or partially be condensed in a simple way because the condensation temperature of NH_3 at the said pressures has increased to above the temperature level at which cooling water circuits in plants are usually operated.

Yet another advantage of the method according to the invention is that, if it is part of a process for the preparation of melamine in which the gas mixture is obtained at approximately the pressure of the reactor, it is not necessary to increase the pressure in the reactor to obtain the ammonium carbamate solution at a higher pressure. As a result, existing reactors for example do not need to be adapted, and new reactors can be of simpler and thus cheaper design.

The compression step, the aim of which is to increase the pressure of the gas mixture, can be carried out in any way known to one skilled in the art, for example by means of a compressor. On account of the corrosive character of any ammonium carbamate formed upon condensation, the temperature of the compressor components that come into contact with the gas mixture is preferably so high that no condensation occurs. This can be achieved by for example heating the compressor, or by returning a part of the gas mixture whose pressure, and thus temperature, has been increased to the compressor inlet. In addition it may be advantageous to install a separation device before the compressor to trap any drops of liquid that are present. It may further be necessary or desirable to carry out the compression step by arranging several compressors in series.

The absorption step comprises a treatment of an incoming flow, in this method according to the invention consisting of the gas mixture whose pressure has been increased, with liquid NH_3 and optionally water or an aqueous solution of for example NH_3, ammonium carbamate or urea. This results in the formation of an ammonium carbamate solution. The absorption step can be carried out in various ways known to one skilled in the art, for example in a packed column or in a plate column. In the absorption step the liquid NH_3 absorbs the CO_2 and the H_2O from the gas mixture,

so that an ammonium carbamate solution is formed. The CO_2 will afterwards be present in the ammonium carbamate solution, for example as carbamate ion. The liquid NH_3 can be obtained wholly or partially by condensation of a part of the flow of gaseous NH_3 obtained from the absorption step. If the quantity of H_2O is so low that undesired formation of solids can take place, it is advantageous to feed water or an aqueous solution of, for example, NH_3, ammonium carbamate or urea.

In order to be able to carry out the absorption step in an efficient way it is advantageous when the flow that enters the absorption step has a dew point between 65°C and 140°C. More preferably the dew point lies between 80°C and 110°C.

In a preferred embodiment, a first partial condensation step is also done, in which the gas mixture is converted into a gas/liquid mixture. The first partial condensation step can be carried out after the first compression step. The first partial condensation step should be carried out prior to the absorption step. The first partial condensation step can be carried out in a way known to one skilled in the art, for example by means of a heat exchanger that cools the gas mixture so that it is partially condensed. During the first partial condensation step, already much of the gaseous CO_2 present enters the liquid phase, via the formation of for example ammonium carbamate in aqueous solution. The quantity of gaseous CO_2 that enters the absorption step is therefore lower compared to the embodiment not comprising the first partial condensation step, so that the flow of gaseous NH_3 is obtained more efficiently from the absorption step, which implies that less liquid NH_3 is required and/or a higher throughput is possible.

The said further processing of the ammonium carbamate solution to reduce its water content is technically complicated, comprises many process steps and is therefore expensive. For this reason it is a further objective of the invention to provide a method that can obviate the need for said further processing, because a concentrated ammonium carbamate solution is obtained from a gas mixture containing NH_3, H_2O and CO_2 that can be used in the production of urea without further processing to reduce its water content. In this context concentrated means that the mass flow of H_2O has been substantially lowered compared to the gas mixture; preferably the mass flow of H_2O is reduced by more than 25 w%. This method comprises:

- a first partial condensation step, in which the gas mixture is converted into a gas/liquid mixture;
- a first separation step, in which the gas/liquid mixture is separated into an

ammonium carbamate recyclate solution and a concentrated gas mixture, which contains more than 60 wt.% NH_3, less than 30 wt.% CO_2 and less than 10 wt.% H_2O;

- an absorption step,

with at least one of the following two mixtures:

- the gas mixture
- the concentrated gas mixture

being compressed to a pressure between 0.5 MPa and 7.5 MPa in a compression step.

In the first partial condensation step more water than NH_3 or CO_2 is condensed. Upon condensation, ammonium carbamate in solution is formed. It is an advantage of the method according to the invention that the water content in the concentrated gas mixture can be reduced to such an extent that the concentrated ammonium carbamate solution subsequently obtained from the absorption step can directly be applied in the production of urea. The liquid flow recovered from the first separation step according to the invention is defined here and hereafter as an ammonium carbamate recyclate solution. If the method is applied in a process for the preparation of melamine it is advantageous to use the ammonium carbamate recyclate solution as a direct coolant for cooling of the flow leaving the reactor. This has the advantage that the presence of ammonium carbamate during cooling can prevent undesirable reactions of melamine to form compounds such as ammeline and ammelide.

The first partial condensation step can be carried out in a way known to one skilled in the art, for example by means of a heat exchanger that cools the gas mixture so that it is partially condensed.

The aim of the first separation step is to separate the liquid present after the first partial condensation step from the gas phase. The first separation step can be carried out in a way known to one skilled in the art. It is possible, for example, to combine the first separation step with the first partial condensation step, for example in a heat exchanger constructed so that the liquid phase can be collected and can be separated. It is also possible to carry out the first separation step in a so-called knockout drum. This a vessel in which gravity is used to carry out the first separation step: The gas/liquid mixture is fed to the knock-out drum, after which the liquid flows down along the wall and is thence removed and the gas is removed from the middle of the vessel via the top.

As indicated above, the gas mixture and/or on the concentrated gas mixture is subjected to the compression step. In a preferred embodiment the gas mixture is compressed. The compression step also leads to an increase in the temperature of the gas mixture. This has the advantage that the first partial condensation step can be carried out at an elevated temperature, so that higher-quality steam can be generated from the heat released during condensation. More preferably the concentrated gaseous mixture is then subjected to a second partial condensation after the first separation step, in which a second gas/liquid mixture is formed as feed flow for the absorption step. During the second partial condensation step already much of the CO_2 present has entered the liquid phase, via the formation of for example ammonium carbamate. The quantity of gaseous CO_2 that enters the absorption step is therefore lower, so that the flow of gaseous NH_3 is obtained more efficiently from the absorption step, which implies that less liquid NH_3 is required and/or a higher throughput is possible. Even more preferably a second separation step is added to this embodiment, in which step, carried out between the second partial condensation step and the absorption step, a first concentrated ammonium carbamate solution is separated from the second gas/liquid mixture. The advantage of this is that a smaller flow goes to the absorption step, reducing the load on the absorption step. A further advantage of this embodiment is that the composition of the first concentrated ammonium carbamate solution differs somewhat from that of the concentrated ammonium carbamate solution recovered from the absorption step, so that these two solutions can be used for different purposes. If desired the two concentrated ammonium carbamate solutions can also be combined for further processing.

In another preferred embodiment according to the invention it is not, as described above, the gas mixture that is subjected to the compression step, so prior to the first partial condensation step and the first separation step, but the concentrated gas mixture leaving the first separation step. As a result the flow that enters the compression step is smaller than when the gas mixture is already subjected to the compression step, the ammonium carbamate recyclate solution already having been separated. More preferably the concentrated gaseous mixture is then subjected to a second partial condensation step between the compression step and the absorption step, resulting in the formation of a second gas/liquid mixture as feed flow for the absorption step. As described above it is advantageous when, as a consequence of the second partial condensation step, much of the CO_2 present has already entered the liquid phase, via the formation of for example ammonium carbamate. This results in a

lower quantity of gaseous CO_2 entering the absorption step, so that the flow of gaseous NH_3 can be obtained from the absorption step in a more efficient way, which implies that less liquid NH_3 is required and/or a higher throughput is possible. In addition the heat released in this second partial condensation step can be used to supply steam. Even more preferably a second separation step is added to this embodiment, carried out between the second partial condensation step and the absorption step, in which a first concentrated ammonium carbamate solution is separated from the second gas/liquid mixture. The advantage hereof, as described above, is that a smaller flow goes to the absorption step, reducing the load on this step. A further advantage of this embodiment is that the composition of the first concentrated ammonium carbamate solution differs somewhat from that of the concentrated ammonium carbamate solution recovered from the absorption step, so that these two solutions can be used for different purposes. It is of course also possible for the two concentrated ammonium carbamate solutions to be combined for further processing. It may be advantageous to additionally, and prior to the first partial condensation step, increase the pressure of the gas mixture to between 0.3 MPa and 7.5 MPa in a precompression step. This is particularly advantageous if this method according to the invention is part of a process for the preparation of melamine in which the reactor is operated at a low pressure, for example atmospheric pressure.

When applying those embodiments of the method according to the invention in which a concentrated ammonium carbamate solution is obtained, this solution generally contains between 25 and 50 wt.% NH_3, between 25 and 50 wt.% CO_2 and between 1 and 30 wt.% H_2O. From the gas mixture then a quantity of water has been removed which, as stated earlier, is particularly advantageous if the concentrated ammonium carbamate solution is used for the preparation of urea. In addition, gaseous NH_3 has been recovered from the gas mixture, which can be put to any desired use. The gaseous NH_3 can for example, if the method according to the invention is applied to a gas mixture obtained in a process for the production of melamine, be fed to the melamine reactor to serve as fluidization gas.

The industrial applicability of the present invention is, as indicated earlier, not limited to incorporation in processes for the preparation of melamine. In particular, incorporation in processes for the preparation of urea is foreseen. For example, in conventional processes for the preparation of urea, gas mixtures are present from which an ammonium carbamate solution should be obtained. Conventional urea processes are understood to be those processes where the effluent

of the synthesis zone is reduced in pressure prior to the execution of subsequent process steps such as separation of unreacted NH_3 and CO_2 and/or decomposition of ammonium carbamate.

If the present invention is applied to a process for the preparation of urea, the gas mixture is preferably raised to a pressure at or slightly above the pressure in the urea synthesis zone, i.e. to a pressure between 12.5 MPa and 25 MPa, more preferably to a pressure between 14 MPa and 22 MPa. The advantage of this is that the water content of the stream returning to the urea synthesis zone is reduced, leading to a higher conversion rate in the synthesis zone as a result of which the recovery of unreacted components can be achieved by a reduced technical effort (i.e. reduced heat or steam consumption, reduced stripping agent consumption).

The method according to the invention is explained on the basis of the following drawings.

In the drawings Figure 1 shows an embodiment with a compression step, followed by an absorption step;

Figure 2 an embodiment with a first partial condensation step and a separation step, with a compression step taking place either prior to the first partial condensation step or between the separation step and the absorption step;

Figure 3 an embodiment in which, besides a compression step, a first partial condensation step, a separation step and an absorption step, also a second partial condensation step takes place, this step being carried out between the first separation step and the absorption step;

Figure 4 an embodiment in which, compared with Figure 3, a second separation step has been added, between the second partial condensation step and the absorption step;

Figure 5 an embodiment in which, compared with Figure 3, the compression step is carried out after the separation step;

Figure 6 an embodiment in which, compared with Figure 5, a second separation step has been added, which is carried out between the second partial condensation step and the absorption step;

Figure 7 an embodiment in which the gas mixture is subjected to a precompression step, followed by either a first partial condensation step, separation step, compression step and absorption step, or a series of process steps as in Figure 6;

Figure 8 an embodiment according to the prior art.

The first digit of the numbers in the figures is the same as the number of the figure. If the last two digits of the numbers of different figures are the same, they refer to the same element.

In Fig. 1 the gas mixture, which for example comes from the cooling zone of a process for the preparation of melamine, is fed via line **102** to compressor **104**. The gas mixture of which the pressure has been increased is then fed via line **106** to absorber **108**. Absorber **108** also receives liquid ammonia via line **110** and water or an aqueous solution of for example ammonium carbamate via line **112**. Lines **110** and **112** can optionally be combined to form one line. Via line **114** the ammonium carbamate solution leaves absorber **108** and via line **116** the flow of gaseous NH_3.

Fig. 2 shows condenser **218**, in which the gas mixture is subjected to the first partial condensation step. A coolant, for example water, is supplied via line **220** and removed via line **222**. The gas/liquid mixture then goes via line **224** to separator **226**. From separator **226** on the one hand the ammonium carbamate recyclate solution is obtained via line **228**, while on the other hand the concentrated gas mixture becomes available that goes via line **230,** to absorber **208**, which further operates as described under Figure 1. It is possible for condenser **218** and separator **226** to be combined into one apparatus. In Figure 2, two possible locations for compressor **204** are shown by means of dotted lines: either upstream of condenser **218**, so that the gas mixture supplied via line **202** is subjected to the compression step, or upstream of absorber **208**, so that the concentrated gas mixture coming from separator **226** is subjected to the compression step.

Fig. 3 shows an embodiment in which the gas mixture entering compressor **304** via line **302** is subjected to the compression step, after which the gas mixture of which the pressure has been increased enters, via line **328,** condenser **318**, where the first partial condensation takes place. The gas/liquid mixture then goes via line **324** to separator **326.**The concentrated gas mixture leaving separator **326** is fed via line **334** to a next condenser **336**, in which the second partial condensation step takes place. Condenser **336** is cooled by means of a coolant, for example water, that is supplied via line **338** and removed via line **340**. The second gas/liquid mixture is then fed via line **342** to absorber **308**.

Fig. 4 builds on the embodiment of Fig. 3 by feeding the second gas/liquid mixture, after the second partial condensation step, carried out in condenser **436,** via line **444** to separator **446,** in which the second separation step is carried out. The first concentrated ammonium carbamate solution leaves separator **446** via line

448, after which the remaining flow is fed via line **450** to absorber **408**.

The method as shown in Fig. 5 builds on that of Fig. 2; in this case compressor **504** is installed downstream of the first partial condensation step (in condenser **518**) and the first separation step (in separator **526**). The compressor is fed with the concentrated gas mixture via line **552.** The compressed flow from compressor **504** is fed via line **554** to condenser **536**; here the second partial condensation step takes place. The second gas/liquid mixture is then fed via line **542** to absorber **508**.

Fig. 6 shows how the design as shown in Fig. 5 can be extended further according to the invention by installing second separator **656**, in which the second separation step takes place. Second separator **656** is fed via line **642** with the second gas/liquid mixture. The second gas/liquid mixture is separated into the first concentrated ammonium carbamate solution, which is removed via line **658**, and a strongly concentrated gas mixture, which is fed to absorber **608** via line **660.**

Fig. 7 shows the situation in which a precompression step is carried out in compressor **762**, after which the precompressed gas mixture is fed via line **764** to condenser **718,** where the first partial condensation step is carried out. The compression step itself then follows after the first partial condensation step and the first separation step (carried out in separator **726**). Optionally, and indicated by dotted lines, a second partial condensation can still be carried out in condenser **736** and afterwards optionally a second separation in separator **756** before the absorption step takes place in absorber **708**.

Below, the invention is elucidated on the basis of the following example, with also a comparative experiment being given.

Example I

In Example I use is made of the configuration as shown in Fig. 5. The compositions of the various flows are shown in Table 1.

Table 1

Flow	**502**	**524**	**552**	**528**	**554**	**542**	**514**	**510**	**512**	**516**
NH_3 (wt.%)	63	63	74	36	74	74	41	100	0	100
CO_2 (wt.%)	18	18	17	20	17	17	39	0	0	0
H_2O (wt.%)	19	19	9	44	9	9	20	0	100	0
T [°C]	111	70	70	70	215	105	100	45	33	45
P [MPa]	0.6	0.6	0.6	0.6	2	2	2	2	2	2
Wt. % gas	100	71	100	0	100	55	0	0	0	100
Total [t/h]	86	86	61	25	61	61	28	12	0.5	45

Comparative experiment A

See Fig. 8. In comparative experiment A a gas mixture is fed via line **802** to condenser **818**, where partial condensation takes place. The gas/liquid mixture coming from condenser **818** is fed via line **866** to absorber **808**. Flows **802** and **866** are not subjected to a compression step. Absorber **808** also receives a liquid NH_3 flow via line **810** and an aqueous flow via line **812**. An ammonium carbamate solution and a gaseous NH_3 flow leave absorber **808**, via line **814** and line **816**, respectively. Compositions of the said flows are given in Table 2 below.

Table 2

Flow	**802**	**866**	**814**	**810**	**812**	**816**
NH_3 (wt.%)	59	59	36	100	0	100
CO_2 (wt.%)	13	13	19	0	0	0
H_2O (wt.%)	28	28	45	0	100	0
T [°C]	119	73	72	45	33	5
P [MPa]	0.6	0.6	0.6	2	0.6	0.6
Wt.% gas	100	46	0	0	0	100
Total [t/h]	81	81	53	12	1	41

As is evident from Example I and Comparative experiment A the concentrated ammonium carbamate solution **514** of Example 1 contains less ammonia (approx. 11 t/h, or 41 wt.% of 28 t/h) than the ammonium carbamate solution **814** of Comparative experiment A (approx. 19 t/h, or 36 wt.% of 53 t/h), despite the fact that flow **802** in the comparative experiment has a somewhat lower initial ammonia concentration (59 wt.% versus. 63 wt.% for flow **502**). In addition the concentrated ammonium carbamate solution **514** of Example 1 contains a lower percentage of water (20 wt.%, almost 6 t/h) than the ammonium carbamate solution **814** of Comparative experiment A (45 wt.%, almost 24 t/h). As a result, the concentrated ammonium

carbamate solution **514** can be used without further processing to reduce its water content in the preparation of urea, which is not possible in an efficient way for ammonium carbamate solution **814**.

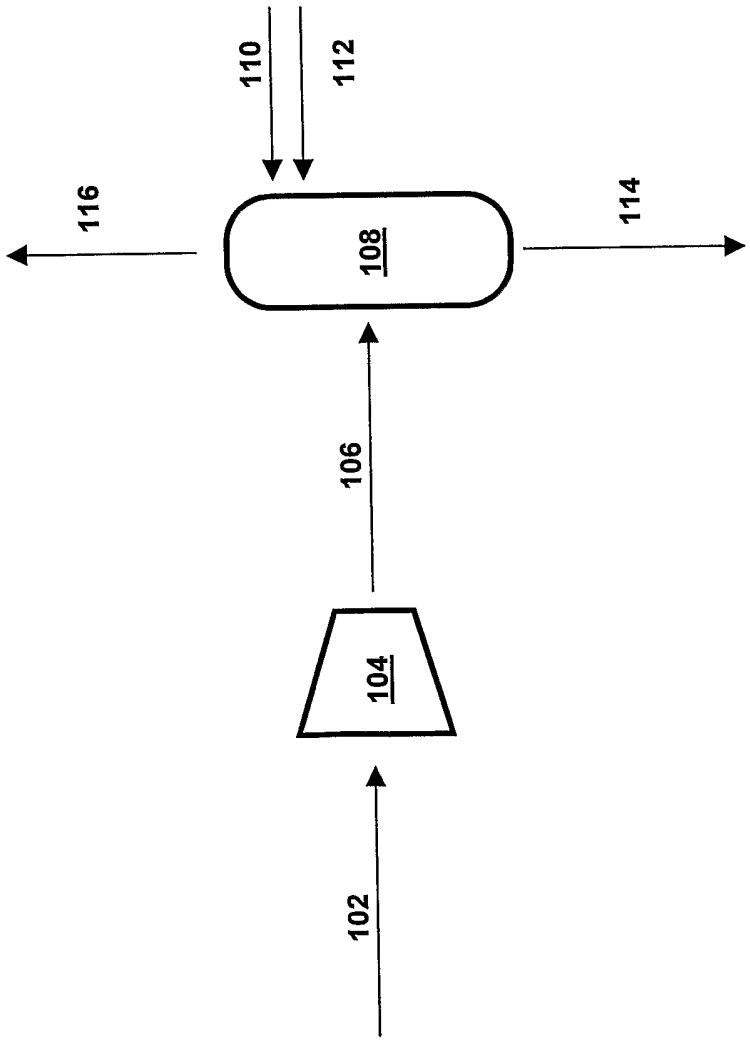
CLAIMS

1. Method for obtaining an ammonium carbamate solution from a gas mixture that contains more than 40 wt.% NH_3, less than 50 wt.% CO_2 and less than 40 wt.% H_2O and has a pressure between 0.1 MPa and 4 MPa, comprising a compression step in which the pressure of the gas mixture is increased to a pressure between 0.5 MPa and 25 MPa, and an absorption step.
2. Method according to claim 1, further comprising, after the compression step and prior to the absorption step, a first partial condensation step, in which the gas mixture is converted into a gas/liquid mixture.
3. Method for obtaining a concentrated ammonium carbamate solution from a gas mixture that contains more than 40 wt.% NH_3, less than 50 wt.% CO_2 and less than 40 wt.% H_2O and has a pressure between 0.1 MPa and 4 MPa, comprising:
   - a first partial condensation step, in which the gas mixture is converted into a gas/liquid mixture;
   - a first separation step, in which the gas/liquid mixture is separated into an ammonium carbamate recyclate solution and a concentrated gas mixture that contains more than 60 wt.% NH_3, less than 30 wt.% CO_2 and less than 10 wt.% H_2O;
   - an absorption step,

   with at least one of the following two mixtures:
   - the gas mixture
   - the concentrated gas mixture being compressed in a compression step to a pressure between 0.5 MPa and 25 MPa.
4. Method according to claim 3, in which the gas mixture is compressed.
5. Method according to claim 4, in which after the first separation step the concentrated gaseous mixture is subjected to a second partial condensation step in which a second gas/liquid mixture is formed as feed flow for the absorption step.
6. Method according to claim 5, in which a first concentrated ammonium carbamate solution is separated from the second gas/liquid mixture in a second separation step, carried out between the second partial condensation step and the absorption step.

7. Method according to claim 3, in which the compression step is carried out after the first separation step.
8. Method according to claim 7, in which the concentrated gaseous mixture is subjected to a second partial condensation step, carried out between the compression step and the absorption step, in which a second gas/liquid mixture is formed as feed flow for the absorption step.
9. Method according to claim 8, in which a first concentrated ammonium carbamate solution is separated from the second gas/liquid mixture in a second separation step, carried out between the second partial condensation step and the absorption step.
10. Method according to any one of claims 7 – 9, in which, prior to the first partial condensation step, the pressure of the gas mixture is increased to between 0.3 MPa and 7.5 MPa in a precompression step.
11. Method according to any one of claims 3 – 10, in which the concentrated ammonium carbamate solution comprises between 25 and 50 wt.% NH_3, between 25 and 50 wt.% CO_2 and between 1 and 30 wt H_20.
12. Method according to any one of claims 1 – 11, in which the gas mixture has been obtained in a process for the production of melamine.
13. Method according to any one of claims 1 – 11, in which the gas mixture has been obtained in a process for the production of urea.

FIG. 1



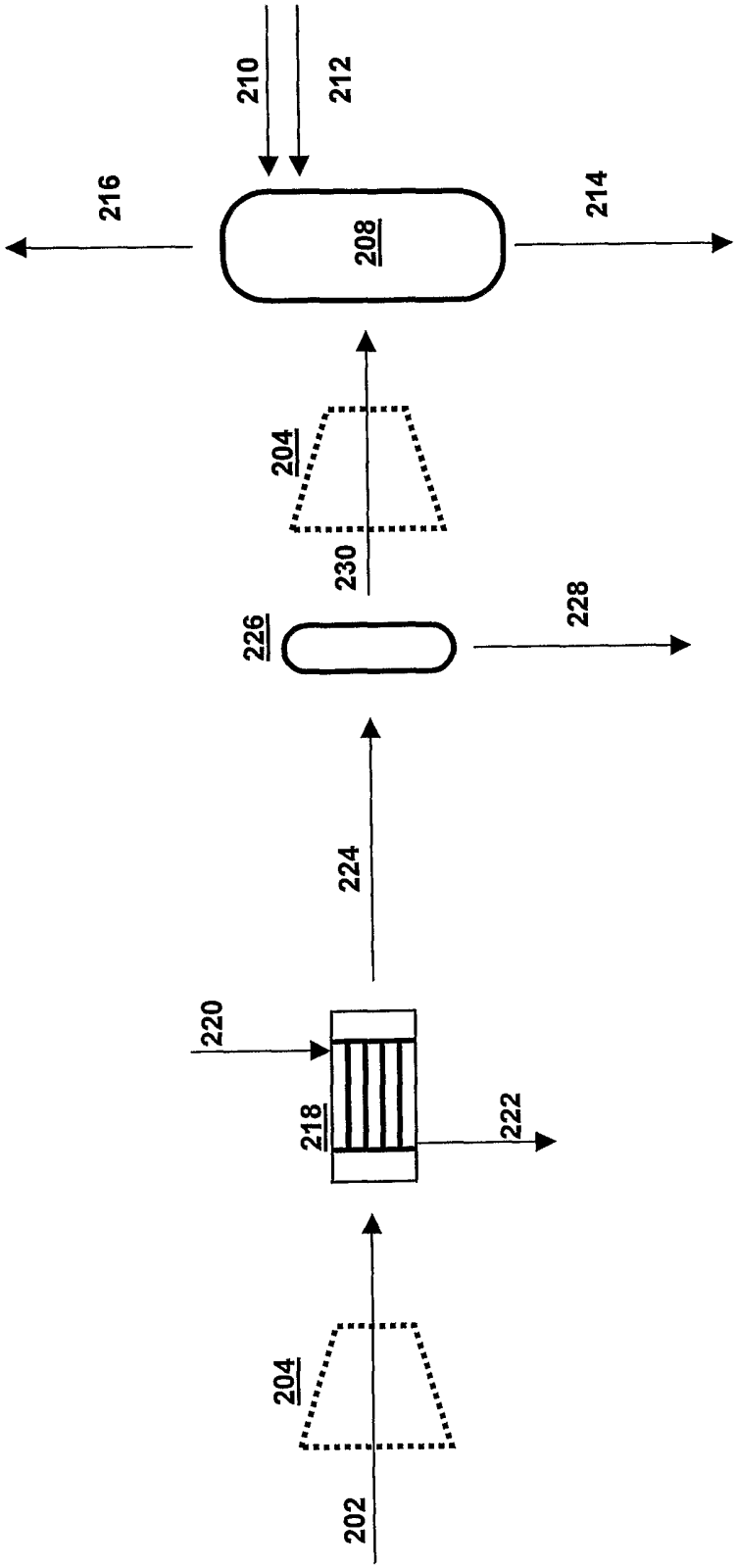


FIG. 2

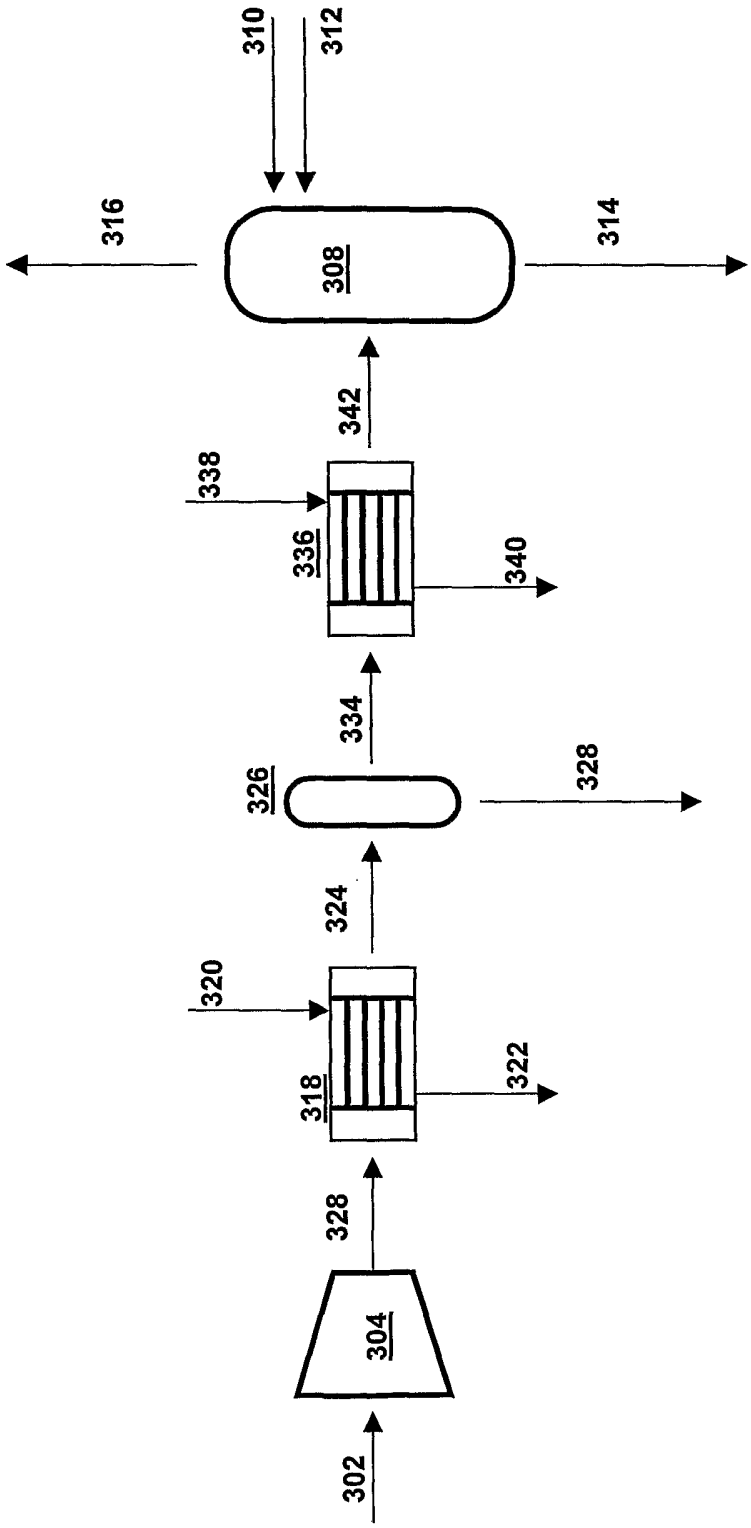

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FIG. 3

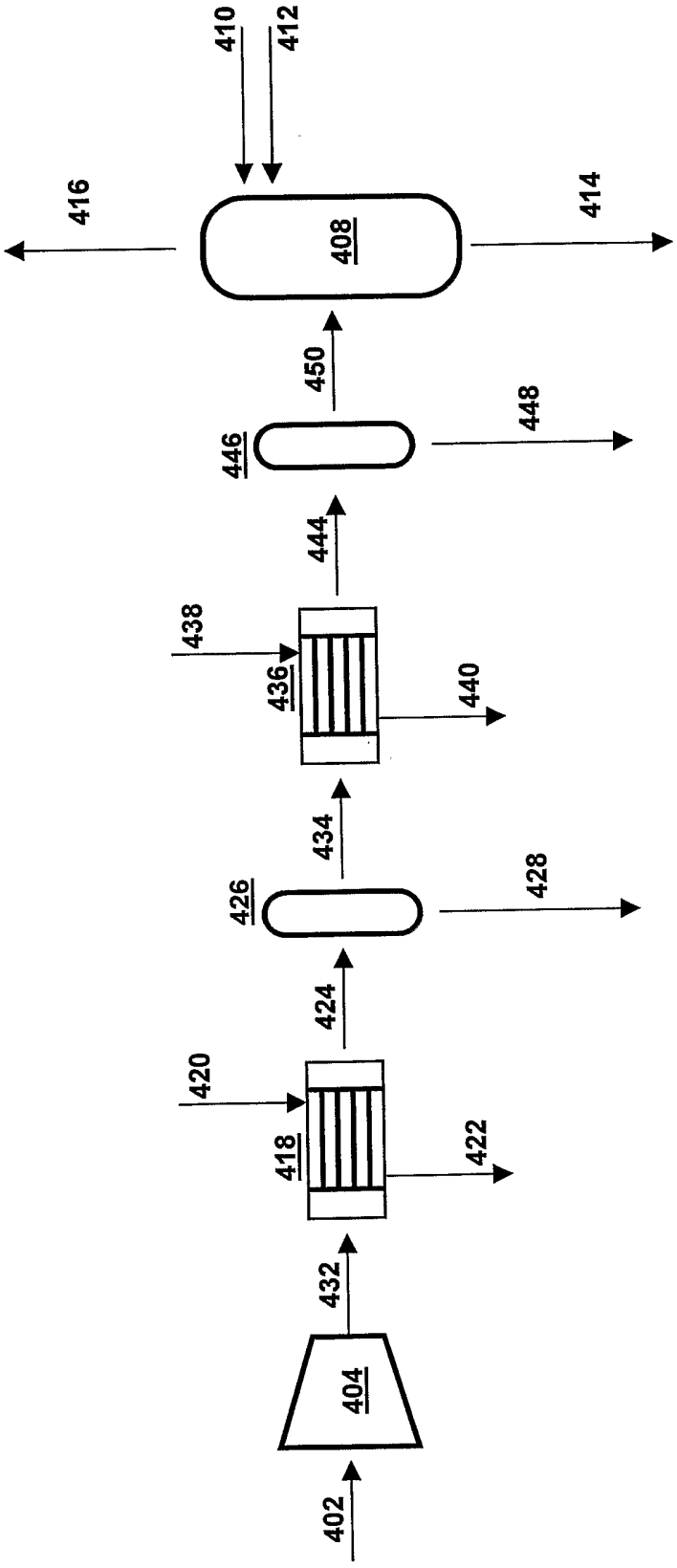
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FIG. 4

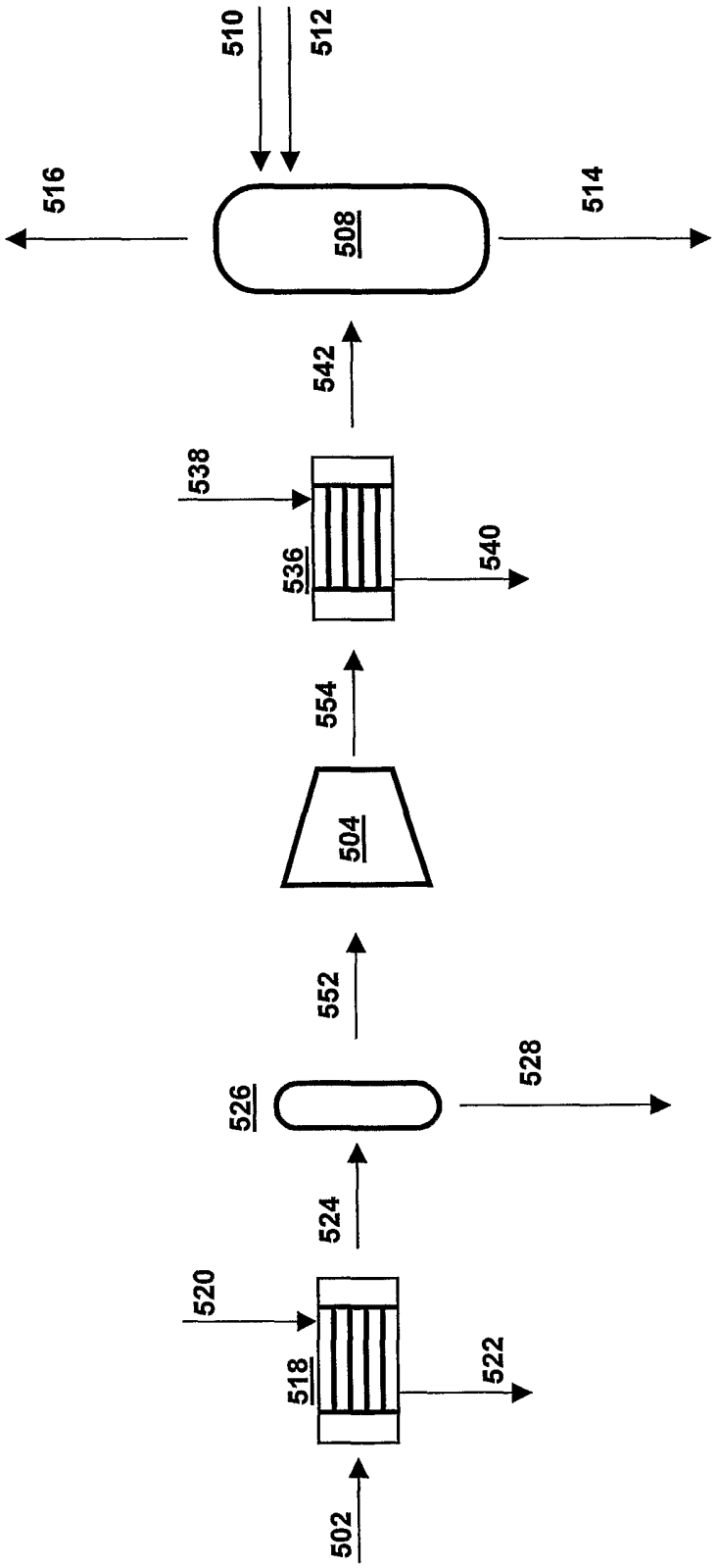
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FIG. 5

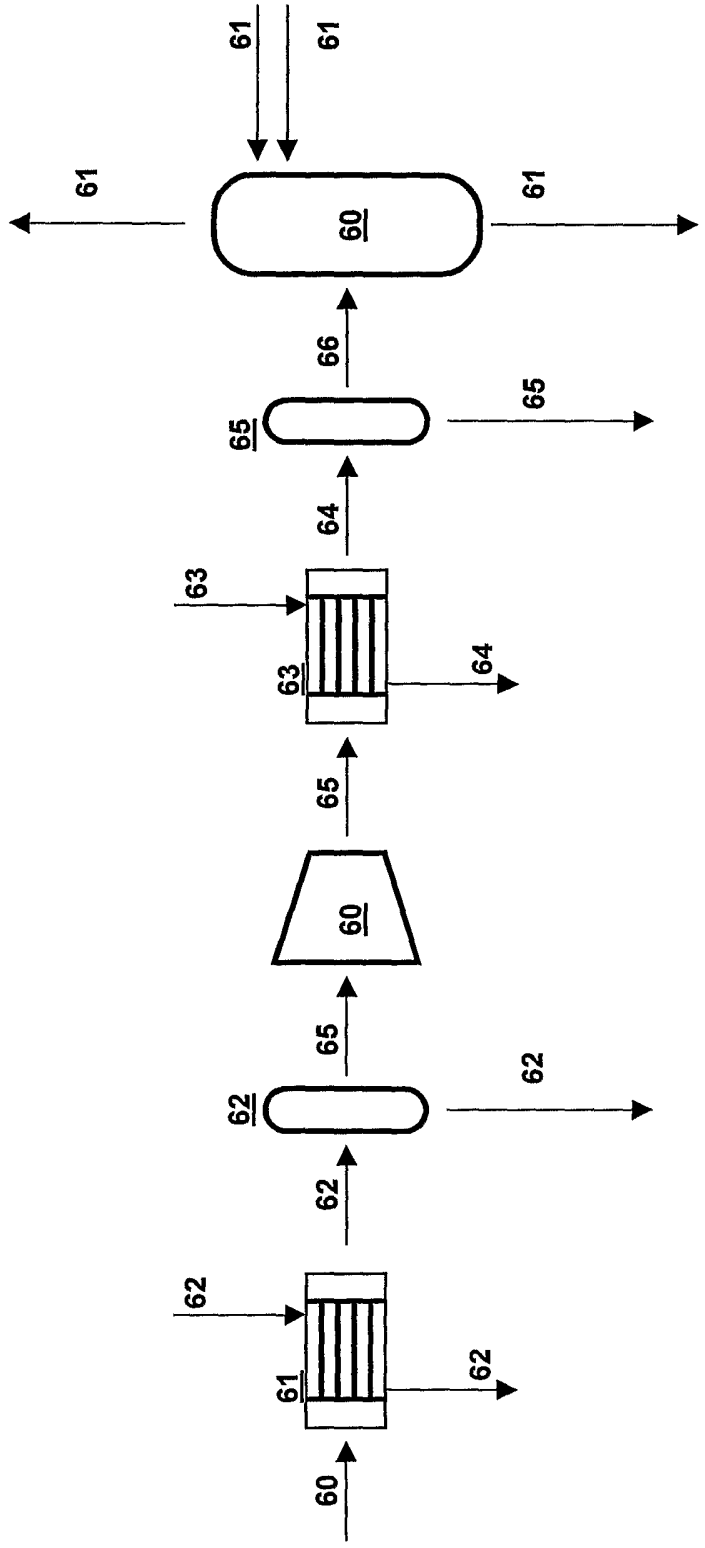

FIG. 6

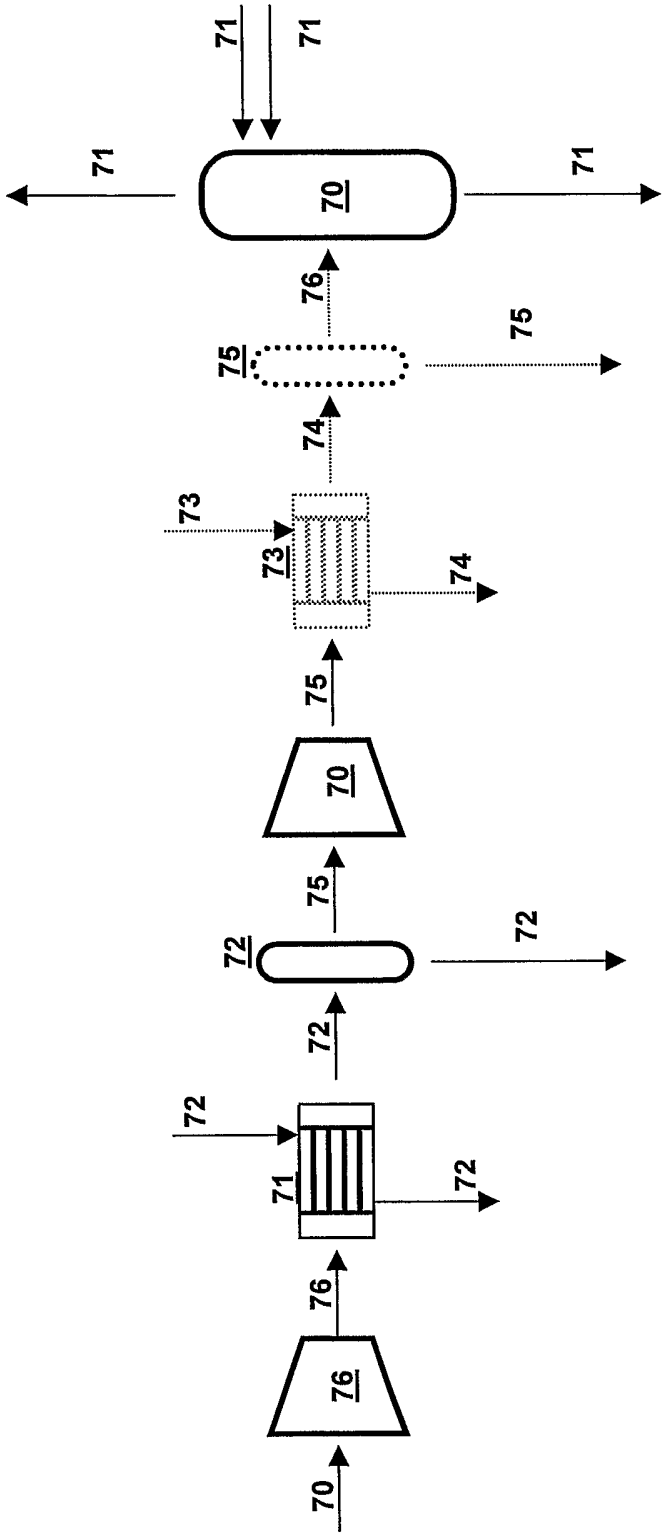

FIG. 7

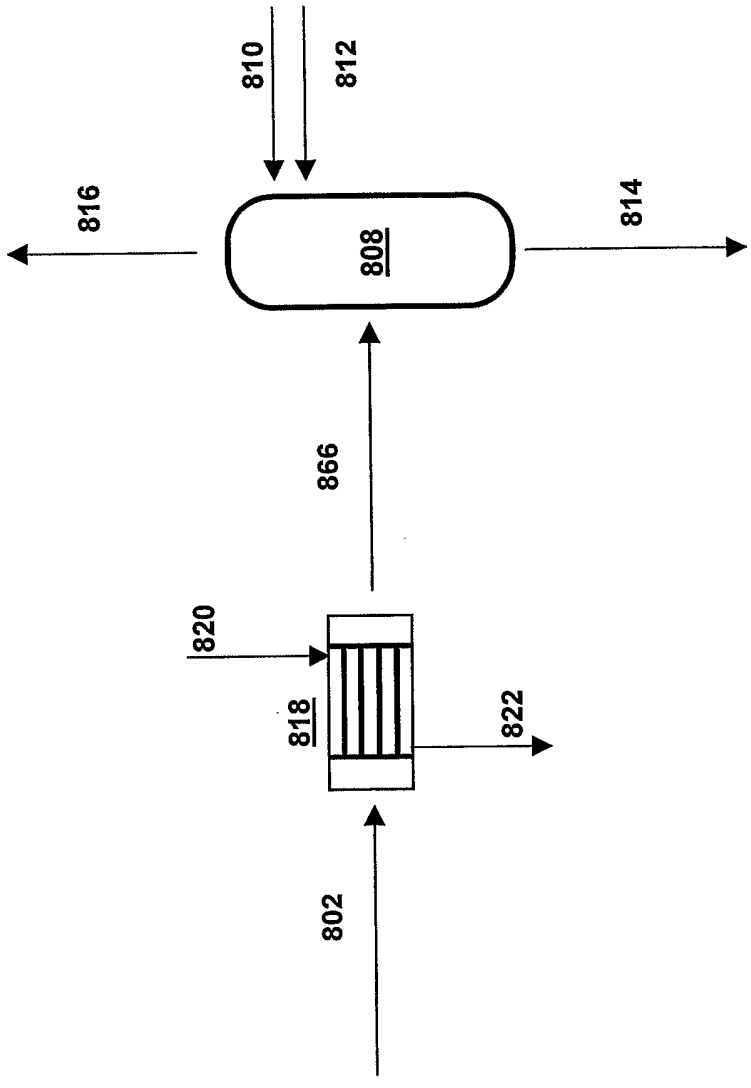

FIG. 8

INTERNATIONAL SEARCH REPORT

PCT/NL 02/00433

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 C01B21/12 C07D251/60 C01C1/10

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 C01B C07D C01C

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)
WPI Data, PAJ, CHEM ABS Data, EPO-Internal

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 3 708 536 A (HILLENBRAND E) 2 January 1973 (1973-01-02) column 3, line 44 - line 58; claims; figure	1,12,13
A	US 3 310 558 A (OELE) 21 March 1967 (1967-03-21) column 3, line 59 -column 4, line 25; claims; figure 1	1-11
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[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

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Date of the actual completion of the international search	Date of mailing of the international search report
20 September 2002	27/09/2002

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Zalm, W

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

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