(19) Europäisches Patentamt
European Patent Office
Office européen des brevets

(11) Publication number: **0 092 856**
A2

(12) EUROPEAN PATENT APPLICATION

(21) Application number: 83200434.5

(22) Date of filing: 28.03.83

(51) Int. Cl.^{3}: C 10 J 3/46
C 10 J 3/84

(30) Priority: 26.04.82 NL 8201715

(43) Date of publication of application:
02.11.83 Bulletin 83/44

(84) Designated Contracting States:
BE DE FR GB IT

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(54) A process for the gasification of a solid carbon-containing fuel.

(57) Coal is ground, dried and subsequently subjected to a partial combustion with oxygen or oxygen-enriched air, the oxygen having been obtained from an air separation plant. Hot synthesis gas produced by the partial combustion is first cooled to 100-500°C and then further cooled to 25-250°C by means of indirect heat exchange with a nitrogen-rich gas stream obtained in the air separation plant. Preferably, the heated nitrogen-rich gas stream is used to dry the coal.

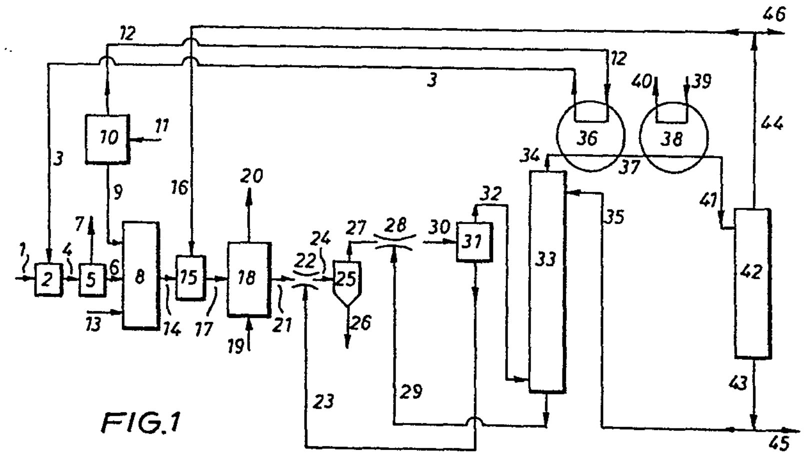

FIG.1

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A PROCESS FOR THE GASIFICATION OF A SOLID CARBON-CONTAINING FUEL

The invention relates to a process for the gasification of a solid carbon-containing fuel, characterized in that

a) the fuel is ground fine and dried,

b) the fuel is subsequently partially combusted to synthesis gas by means of oxygen or oxygen-enriched air, which oxygen originates from an air separation plant.

c) the synthesis gas is cooled to a temperature of 100-500°C, and

d) the cooled synthesis gas is further cooled to a temperature of 25-250°C by means of indirect heat exchange with a nitrogen-rich gas originating from the air separation plant.

In the gasification (partial combustion) of a solid carbon-containing fuel synthesis gas is formed substantially consisting of carbon monoxide and hydrogen. Suitable solid fuels are coal, brown coal, coke, peat, wood, etc.

A solid fuel is generally less reactive than a gaseous or a liquid fuel. In order nevertheless to obtain a rapid reaction the solid fuel is ground fine. The ground fuel is subsequently transported to a gasification reactor. A fuel containing a relatively large quantity of water may agglomerate during transport and thus cause blockages in the transport line. In order to prevent this the solid fuel is dried before being transported to the reactor. After drying the moisture content of the fuel is preferably 0-10% by weight. The solid fuel is partially combusted with oxygen or with oxygen-enriched air, since the reaction then proceeds more rapidly than with air. Further, the synthesis gas formed now contains less nitrogen than when it is gasified with air. This simplifies the subsequent purification of the synthesis gas. Moreover, synthesis gas prepared by means of oxygen is more suitable for certain syntheses, for example that of methanol or hydrocarbons.

The oxygen required as reactant is prepared in a plant where air is separated into an oxygen-rich and at least one nitrogen-rich gas. To this end use is made of cryogenic distillation.

The partial combustion preferably takes place in the presence of a moderator. The moderator has a moderating effect on the temperature in the reactor by endothermic reaction with the reactants and/or the partial oxidation products. Suitable moderators are steam and carbon dioxide.

After the fuel has reacted with the oxygen, the synthesis gas formed leaves the reactor at a temperature of 1200°C-1700°C. Apart from carbon monoxide and hydrogen the gas may also contain, inter alia, carbon dioxide, water vapour, sulphur compounds, methane and small amounts of hydrogen cyanide and ammonia. Moreover, it entrains slag droplets. The slag droplets may present problems when they cool down. The fact is that they have no melting point but a melting range that can be hundreds of degrees centigrade. Since they are sticky in the melting range, they may cause blockages. If coal is used as fuel, the slag is usually sticky in the temperature range of 900-1500°C. Solid slag is no longer sticky. Therefore, the hot gas is preferably rapidly cooled to a temperature of 700-900°C by injecting cold gas or a cold liquid. As a result of the rapid cooling the slag droplets quickly solidify to solid particles. Suitable coolants are recycled synthesis gas, water and/or steam. The cooled synthesis gas is subsequently further cooled to 100-500°C, preferably in a waste heat boiler in which useful high-pressure steam is generated. Subsequently, the solid slag particles are preferably removed from the gas.

After the solid slag particles have been separated from the gas, the synthesis gas is further processed. For example, the other impurities are then removed from the synthesis gas. To this end it is advantageous to reduce the temperature of the impure synthesis gas to 25-250°C. This has so far been performed by means of air or water coolers. In this manner, however, the residual heat of the synthesis gas was not applied usefully.

The cryogenic distillation of air yields a nitrogen-rich gas which is generally discharged into the atmosphere. It is also possible that, in addition to an oxygen-rich gas, pure or substantially pure nitrogen and a stream of waste nitrogen is produced. The pure nitrogen is preferably used in the process, for example as carrier gas for the ground fuel during the transport to the reactor. Another application is in the synthesis of ammonia. The waste nitrogen stream, which still contains some per cents of oxygen, is usually discharged into the atmosphere. The temperature of the nitrogen-rich gas that is discharged is generally chosen about 10°C lower than the temperature of the ambient air.

In the process according to the present invention the relative cold of the nitrogen-rich gas is used. The synthesis gas is further cooled to a temperature of 25-250°C by means of indirect heat exchange with the nitrogen-rich gas.

The above-mentioned indirect heat exchange can take place immediately after the passage of the synthesis gas through the waste heat boiler. However, the solid slag particles are preferably at least partly first removed from the synthesis gas. This is effected in a suitable separator, such as a cyclone, bend separator, filter, etc. The heat exchange between the synthesis gas and the nitrogen-rich gas preferably takes place after the gas has at least partly been freed from the solid slag particles in the separator. Although at least the greater part of the solid slag particles has been separated from the synthesis gas in the separator, solid slag particles can remain behind in the gas and these remaining slag particles constitute a risk of fouling of the cooler. The synthesis gas is therefore more preferably not further cooled by means of indirect heat exchange with the nitrogen-rich gas until after it has at least once been scrubbed with water. The washing step results in an aqueous suspension of solid slag particles and synthesis gas that has substantially been purified of slag particles. The aqueous suspension is separated from the synthesis gas and is advantageously at least partly recycled into the system. Suitable scrubbers are Venturi scrubbers and gas scrubbers in which gas and water are countercurrently contacted with each other.

In the scrubbing step the synthesis gas is already cooled down. By means of the indirect heat exchange with the nitrogen-rich gas the synthesis gas is further cooled preferably to a temperature of 40-140°C.

As a result of the scrubbing step the synthesis gas contains much water vapour. The gas is preferably dried. This is most simply effected by cooling the synthesis gas to below the dew-point, as a result of which part of the water vapour condenses. If it is cooled to far below the dewpoint, the greater part of the water vapour condenses. Subsequently, the dry gas is separated from the condensate. The synthesis gas is preferably cooled to far below the dewpoint in a cooling treatment taking place after the indirect heat exchange with the nitrogen-rich gas. During said cooling the synthesis gas is advantageously cooled to a temperature of 10-75°C. The cooling can be carried out both with air and with water. Since the synthesis gas has already been cooled to 25-250°C by means of indirect heat exchange with the nitrogen-rich gas, a relatively small cooler will suffice.

The nitrogen-rich gas heated according to the invention is preferably applied usefully for drying the solid fuel. Drying takes place before the fuel is fed into the gasification reactor. If the solid fuel is supplied as not too coarse lumps, drying can be performed before it is passed to the grinding mill. If the fuel consists of large lumps, then it is more efficient to dry it during and/or after grinding. If the fuel has a high water content, the hot nitrogen-rich gas, optionally after further heating, can be used to dry the fuel entirely or partly. In the latter case drying to the required water content takes place afterwards in another manner or with another hot gas. It is possible to use many types of grinding mills. Depending on the type of grinding mill drying takes place during or after grinding. For example, if a ball mill or a roller mill is used, the hot nitrogen-rich gas is introduced into the mill and already exercises its drying effect during grinding. The hot nitrogen-rich gas is subsequently used to discharge the ground fuel from the grinding mill and dries the fuel particles during transport.

Before the drying of the solid fuel the nitrogen-rich gas is preferably heated to a temperature in the range of 50 to 400°C, depending on the water content of the fuel. For the greater part of the fuels a temperature of 90-150°C is suitable. The lower temperature limit is such that just enough expelling power is available to remove the water present from the fuel. The upper temperature limit is determined by economic motives. The heat content of the synthesis gas is such that the required quantity of the nitrogen-rich gas can be heated to a temperature up to 400°C.

As already stated before, the nitrogen-rich gas is preferably the waste nitrogen stream formed in the cryogenic distillation of air. The invention is not limited thereto. Use can be made of any nitrogen-rich gas originating from the air separation plant. If a nitrogen-rich gas having a relatively high oxygen content is used to dry the ground fuel, there is a risk of an explosive combustion of the fuel with the oxygen. Therefore, the nitrogen-rich gas preferably contains less than 12% by volume of oxygen, more preferably less than 10% by volume. The waste nitrogen stream complies with said requirements.

The invention is now further illustrated with reference to the Figures to which the invention is otherwise by no means limited. Auxiliary means, such as compressors, pumps, valves, etc., are not shown in the diagrammatic figures.

In Fig. 1 a carbon-containing solid fuel is introduced into a grinding mill 2 via line 1. Via a line 3 a stream of hot nitrogen-rich gas is introduced into the grinding mill 2, where the gas dries the finely ground fuel. Ground and dried fuel, together with the gas mixture of inter alia nitrogen and water vapour, is conducted to a separator 5 through a line 4. Suitable separators are for example, bend separators, cyclones, filters etc. In the separator 5 the ground fuel is separated from the gas. The gas mainly consisting of nitrogen and water vapour is vented through a line 7. The separated fuel particles are passed to a reactor 8 through a line 6. (Since the gasification reactor 8 is preferably operated at elevated pressure, the fuel is brought to the desired pressure by means of compressors, supply vessels, locks etc. which are not shown in the Figure.) An oxygen-rich gas originating from an air separation plant 10 is also introduced into the reactor 8 via a line 9. Air is fed into the air separation plant 10 via a line 11. In plant 10 are formed an oxygen-rich gas stream that is passed to the reactor 8 via the line 9 and a substantially pure nitrogen stream, which can at least partly be used in the transport of the fuel to the reactor through the line 6. (Said stream is not shown in the Figure.) The plant 10 also produces a waste nitrogen stream which is discharged via a line 12. The gasification of the carbon-containing fuel with the oxygen and a moderator (steam or CO_2) supplied via a line 13 takes place in the reactor 8. The resulting synthesis gas loaded with slag droplets is passed via a line 14 to a cooling zone 15 where it is cooled down by injecting a cooled and purified recycled synthesis gas that is supplied via line 16. In the cooling zone 15 all slag droplets in the hot synthesis gas solidify. Via a line 17 a mixture of synthesis gas and solid slag particles is discharged from the cooling zone 15 and passed into a waste heat boiler 18 where it is cooled indirectly with water that is supplied via a line 19 and discharged as steam via a line 20. From the waste heat boiler 18 the still warm mixture of synthesis gas and solid slag particles is passed to a Venturi tube 22 via a line 21. There it is contacted with a suspension of solid slag particles in water that is conducted to the Venturi tube 22 via a line 23. In the Venturi tube 22 all the water of the suspension evaporates and a mixture of synthesis gas, water vapour and solid slag particles is passed via a line 24 to a cyclone 25 where the greater part of the solid slag particles is separated from the gas mixture and is discharged from the installation via a line 26. The remainder of the solid slag particles is passed, together with the gas mixture, via a line 27 into a Venturi scrubber 28 where it is contacted with an aqueous suspension of solid slag particles supplied via a line 29. The mixture of synthesis gas, water vapour, drops of water and solid slag particles formed in the Venturi scrubber 28 is conducted to a separator 31 via a line 30. Here an aqueous suspension of solid slag particles is separated from the gas mixture and discharged via the line 23 through which line the aqueous suspension is passed to the Venturi tube 22. The mixture of synthesis gas and water vapour still containing a small quantity of solid slag particles is introduced via a line 32 into the lower part of a gas scrubber 33, where it is countercurrently contacted with water that is passed to the upper part of the column 33 via a line 35. In the column 33 the last residues of solid slag particles are removed from the gas mixture owing to which an aqueous suspension of solid slag particles is formed that is passed from the column 33 to the Venturi scrubber 28 via the line 29. The gas mixture that is now practically free from solid slag particles is conducted via a line 34 to a cooler 36 where the synthesis gas mixture is further cooled by indirect heat exchange with the cold waste nitrogen stream from the line 12 and the waste nitrogen stream is heated. The resulting hot stream is passed from the cooler 36 to the grinding mill 2 via the line 3. If the synthesis gas mixture is cooled to below the dewpoint in the cooler 36 a mixture of synthesis gas, water vapour and water is conducted via a line 37 to an air cooler 38 to which air is supplied via a line 39 and discharged via a line 40. Here cooling to far below the dewpoint takes place, owing to which substantially the whole quantity of water vapour condenses. If the synthesis gas mixture is cooled in the cooler 36 to a temperature above the dewpoint of the gas mixture, the line 37 contains exclusively synthesis gas and water vapour. A mixture of a synthesis gas, condensation water and a small quantity of water vapour is passed from the cooler 38 to a separator 42 via a line 41. In the separator 42 the mixture is separated into condensate that is discharged via a line 43 and a substantially dry synthesis gas that is discharged via a line 44. Part of the condensate is recycled to the column 33 via the line 35. The remaining part is discharged from the apparatus via a line 45. Part of the substantially dry synthesis gas is recycled to the cooling zone 15 via the line 16. The remaining part of synthesis gas is discharged as final product from the apparatus via a line 46 for further processing.

Fig. 2 shows another embodiment of the process according to the invention. Said process is particularly suitable to be used for solid fuels containing a relatively large quantity of water. Immediately after leaving the waste heat boiler 18 the synthesis gas is passed via the line 21 to a heat exchanger 101 where it is cooled by means of a preheated stream of nitrogen-rich gas supplied to the heat exchanger 101 via a line 102 and discharged via a line 103 to the grinding mill 2. The cooled synthesis gas is passed via a line 105 to a Venturi tube 106. Here it is contacted with a water stream via a line 107. All the water introduced evaporates in the Venturi tube 106. The gas mixture is passed via line 108 to a bag filter 109. Here the solid slag particles are separated from the gas mixture. The separated solid slag particles are discharged from the apparatus via a line 110. The gas mixture is passed via a line 111 to the cooler 36 from which it is conducted to the cooler 38 via the line 37. Water vapour condenses in the coolers 36 and 38. After separation of the condensation water from the synthesis gas in the separator 42 part of the water is conducted to the Venturi tube 106 via the line 107. The cold nitrogen-rich gas stream is introduced into the cooler 36 via the line 12 and after heating it is discharged via the line 3. The somewhat warm gas stream can be further heated by indirect heat exchange with superheated steam in a heat exchanger 104 to which the steam is supplied via a line 112 and from which it is discharged via a line 113. To this end use can, for example, be made of steam obtained in the line 20. Instead of indirect heat exchange with steam it is also possible to use steam injection into the nitrogen-rich gas stream in order to raise the temperature of the gas stream. The warm nitrogen-rich gas stream is passed to the heat exchanger 101. Here it cools the synthesis gas further and is further heated itself. The hot gas stream is subsequently passed to the grinding mill 2 via the line 103 in order to dry the fuel.

EXAMPLE 1

By a process, as described in Fig. 1, 45.8 tons per hour (t/h) of coal having the following composition is passed to the mill 2:

C	67.3	% by wt.
H	4.4	"
N	1.5	"
O	5.9	"
S	2.9	"
ash	12.0	"
water	6.0	"

A quantity of 123.3 t/h of a nitrogen-rich gas stream of 120°C is added thereto. The gas has the following composition:

N_2+Ar	95	mole %
O_2	4	"
H_2O	1	"

The ground coal is separated from the gas in a filter 5. A quantity of 43.93 t/h of coal powder is passed to the reactor 8 via the line 6. 125.17 t/h of the nitrogen-rich gas with water vapour is vented via the line 7. The gas has a temperature of 70°C and the following composition:

N_2+Ar	92.8	mole %
O_2	3.9	"
H_2O	3.3	"

The water content of the coal powder is still 2.0% by weight. The temperature of the synthesis gas generated in the reactor 8 is 360°C in the line 21.

After the washing step with water the temperature is still 130°C. In the cooler 36 a quantity of 176.5 t/h of synthesis gas is cooled with 123.3 t/h nitrogen-rich gas of 10°C. The temperature of the resulting mixture of synthesis gas, water vapour and water is 122°C; the nitrogen-rich gas is heated to 120°C.

In the cooler 38 the temperature of the synthesis gas-containing mixture is reduced to 50°C by air cooling, owing to which a total quantity of 19.1 t/h of water vapour condenses. After the separation of the mixture in the separator 42 the line 45 discharges 1.55 t/h of water and the line 46 discharges 84.5 t/h of synthesis gas of the following composition from the apparatus:

CO	62.6	mole %
H_2	27.6	"
CO_2	2.0	"
H_2S	1.0	"
H_2O	0.6	"
N_2	5.6	"
Ar	1.0	"

EXAMPLE 2

In a process as described in Fig. 2 a quantity of 50 t/h of brown coal is passed to the mill 2. The brown coal has the following composition:

C	39.6	% by wt.
H	3.0	"
N	0.7	"
O	10.3	"
S	0.7	"
ash	10.8	"
water	34.9	"

135 t/h of nitrogen-rich gas of 370°C having the following composition is added thereto via the line 103:

N_2+Ar	95	mole %
O_2	4	"
H_2O	1	"

After separation in filter 5 35.8 t/h of brown coal powder is conducted to the reactor 8 via the line 6 and 149.2 t/h of nitrogen-rich gas with water vapour is vented via the line 7. The vented gas has a temperature of 90°C and the following composition:

N_2+Ar	81.6	mole %
O_2	3.4	"
H_2O	15.0	"

The water content in the brown coal powder is still 9% by weight.

The temperature of the synthesis gas generated in reactor 8 is 400°C in the line 21. Said temperature is reduced to 220°C in the heat exchanger 101, while the nitrogen-rich gas in the line 102 is heated from 180°C to gas of 370°C, which is passed to the mill 2 via the line 103. The gas leaving the bag filter 109 still has a temperature of 180°C. 103.1 t/h of synthesis gas is cooled in the cooler 36 to 98°C with 135 t/h of cool nitrogen-rich gas of 10°C. In this cooling step the nitrogen-rich gas is heated to 160°C. Said gas is further heated to 180°C with steam in the heat exchanger 104, after which it is passed to the heat exchanger 101 via the line 102. The mixture of synthesis gas, water vapour and water formed in the cooler 36 is cooled to 50°C in the air cooler 38, owing to which a total quantity of 7.1 t/h of water vapour condenses. After the separation of the mixture in the separator 42 the line 45 discharges 5.4 t/h of water and the line 46 discharges 58.0 t/h of synthesis gas of the following composition from the apparatus:

CO	59.1	mole %
H_2	28.1	"
CO_2	6.1	"
H_2S	0.5	"
H_2O	0.6	"
N_2	4.4	"
Ar	1.2	"

CLAIMS

1. A process for gasifying a solid carbon-containing fuel, characterized in that:

a) the fuel is ground fine and dried,

b) the fuel is subsequently partially combusted to synthesis gas by means of oxygen or oxygen-enriched air, which oxygen originates from an air separation plant,

c) the synthesis gas is cooled to a temperature of 100°C-500°C,

d) the cooled synthesis gas is further cooled to a temperature of 25-250°C by means of indirect heat exchange with a nitrogen-rich gas originating from the air separation plant.

2. A process as claimed in claim 1, characterized in that the synthesis gas is further cooled by means of indirect heat exchange with the nitrogen-rich gas after the solid slag particles have at least partly been removed from the synthesis gas.

3. A process as claimed in claim 2, characterized in that the synthesis gas is further cooled to a temperature of 40-140°C by means of indirect heat exchange with the nitrogen-rich gas.

4. A process as claimed in any one or more of the preceding claims, characterized in that after the indirect heat exchange with the nitrogen-rich gas the synthesis gas is further cooled to a temperature of 10-75°C in a cooler.

5. A process as claimed in any one or more of the preceding claims, characterized in that the nitrogen-rich gas is heated to a temperature in the range of 50-400°C.

6. A process as claimed in any one or more of claims 1-5, characterized in that the nitrogen-rich gas thus heated is used to dry the solid fuel.

7. A process as claimed in claim 1, as described hereinbefore, with special reference to the Figure.

8. Synthesis gas, as far as obtained by the process according to one or more of the preceding claims.

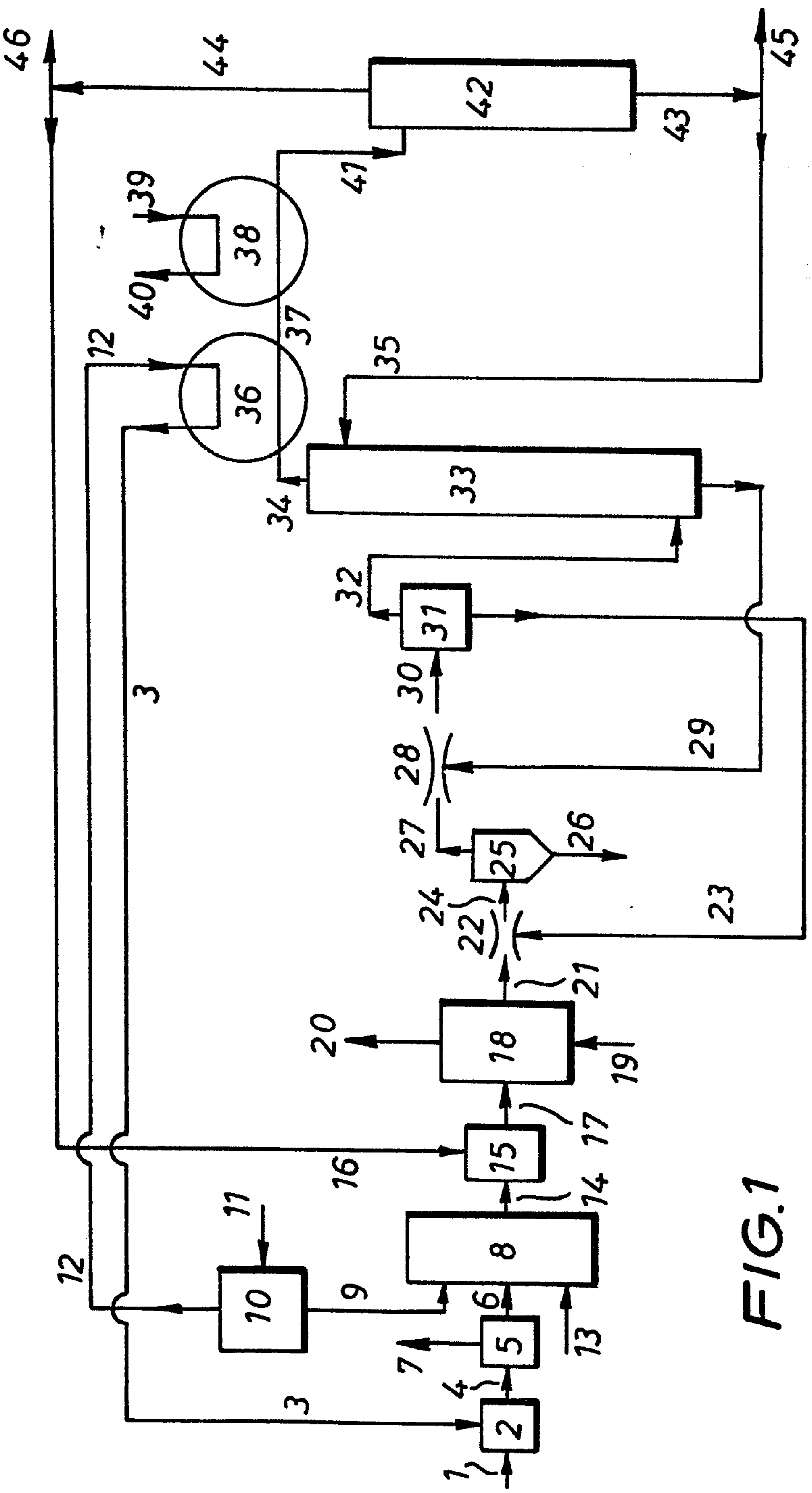

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FIG.1

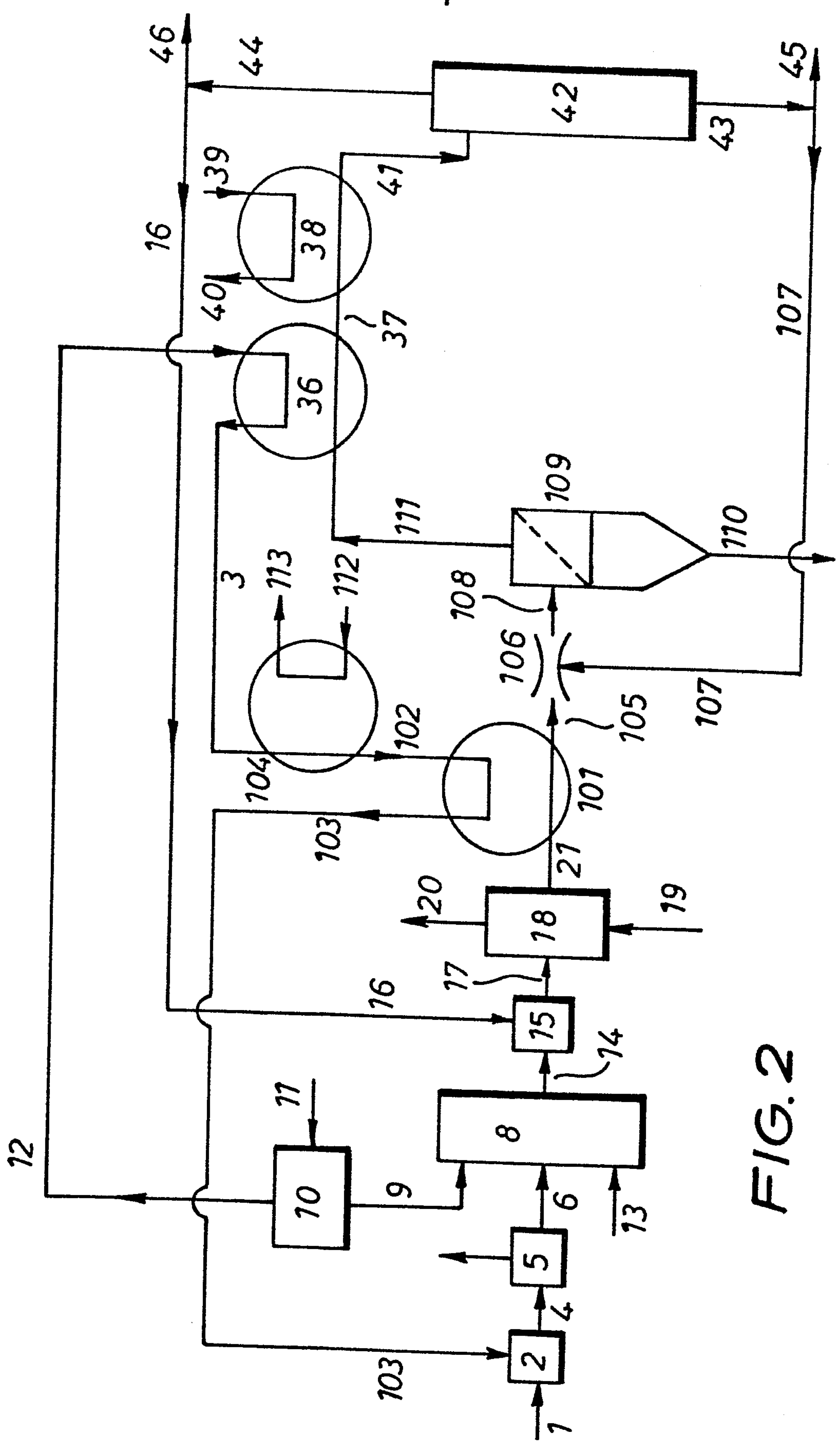
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FIG. 2