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(54) **Title:** METHOD AND APPARATUS FOR THE DIRECT REDUCTION OF IRON ORES UTILIZING GAS FROM A MELTER-GASIFIER

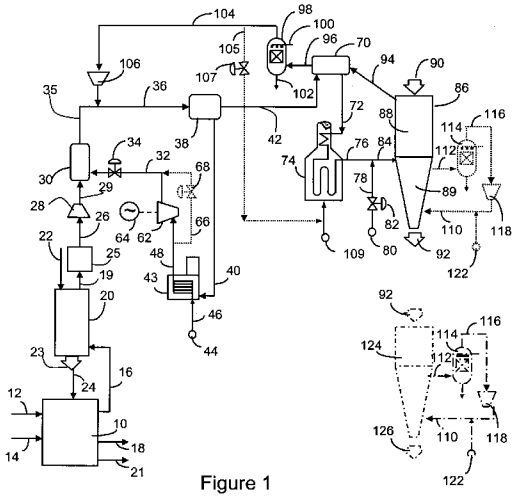

Figure 1

(57) **Abstract:** A method and apparatus for producing a metallized product (DRI) in a downstream direct reduction reactor utilizing the reducing gas effluent either from an upstream reduction reactor associated with a melter-gasifier or from the melter-gasifier itself (in the absence of such upstream reduction reactor), wherein said reducing gas is reacted with water in a shifter and then CO_2 is removed from a combined stream of said shifted gas with recycled gas effluent from said downstream reactor in an adsorption CO_2 removal unit. Separation of CO_2 produces a lean gas stream and a CO_2 laden gas stream. The CO_2 lean gas stream is utilized for producing DRI in said downstream reduction reactor and the CO_2 laden gas stream is used as fuel in a boiler for producing steam which serves then at least to react with carbon monoxide in said shifter to produce hydrogen and preferably also for power generation with resulting overall increased plant efficiency.

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Title of the Invention

Method and Apparatus for the Direct Reduction of Iron Ores Utilizing Gas from a Smelter Furnace

Field of the Invention

The invention relates to the direct reduction of iron ores in a reduction system comprising a smelter furnace, which may be for example a hydrocarbon melter-gasifier wherein coal or other hydrocarbon is converted into a reducing gas mainly composed of hydrogen and carbon monoxide that can be used for direct reduction of iron ore. For example, this reducing gas may be utilized in a first (upstream) direct reduction furnace, and the excess gas effluent from said first reduction furnace is then used for producing direct-reduced-iron (DRI) in a second (downstream) reduction furnace. The invention is an improvement over the prior art whereby the energy per ton of iron produced is decreased as compared with known prior art systems.

Background of the Invention

Smelter furnaces used for melting iron require reducing atmospheres at least to protect the metallic iron being melted and also to reduce the iron oxides usually present in the iron-bearing charge. The hydrocarbon fuel needed to melt the metal in the smelting process generates an excess of reducing gas that is advantageously used elsewhere such as in the direct reduction of iron ore.

Direct reduction of iron ores for producing pre-reduced metallized materials useful for the production of steel is becoming more and more widespread in the steel industry. Some of the advantages of direct reduction plants are that the production capacity may be relatively small as compared with pig iron production in coke-fed blast furnaces, that the metallic iron is produced in solid form with low sulfur and silicon content, and that the resulting DRI may be easily melted in electric-arc furnaces. The reducing agents utilized in the direct reduction plants are hydrogen and carbon monoxide, produced by reformation of natural gas and therefore, these plants have been built in areas where natural gas is available and at relatively low price. A direct reduction plant is also an attractive alternative for utilizing sources of energy available in the form of gases from coke ovens, blast furnaces, coal and/or oil gasification, natural gas, etc.

A particular source of reducing gas, which is inherent to a coal metallurgical plant, is the excess gas produced in the combination of a coal melter-gasifier and a direct reduction reactor, known in the industry with the tradename Corex. Corex plants produce gasified coal

by partial combustion with oxygen-containing gas in the melter-gasifier vessel. The reducing gas generated therein is transferred to the reduction reactor where iron ore lumps or pellets are reduced to varying degrees of metallization. The prereduced iron lumps or pellets are then fed to the melter-gasifier where hot molten metal is produced as a precursor material for steelmaking.

U.S. Patent No. 5,238,487 to Hauk et al. discloses a process for producing pig iron comprising a melter-gasifier 2, a first reduction reactor 1 and a second reduction reactor 15 wherein DRI is produced using reducing gas effluent from said first reactor 1. Hauk does not teach or suggest using a shifter for adjusting the composition of the gas stream effluent from said first reactor 1.

U.S. Patent No. 5,989,308 to Kepplinger et al. describes a plant for the production of pig iron and/or sponge iron comprising a melter-gasifier 8, a first direct reduction furnace 12 for lumpy ores and at least one fluidized bed reactor 19 to process fine ores utilizing reducing gas effluent from said first reduction furnace. Kepplinger however fails to disclose any shifter for increasing the reduction potential of the effluent gas from said first reduction furnace.

U.S. Patent No. 6,251,162 to Eichberger et al. shows a method for producing liquid pig iron wherein a melter-gasifier provides reducing gas to a first reduction furnace 12 and a second reduction reactor 21. This patent does not teach or suggest using a shifter for adjusting the composition of the reducing gas effluent from said first reduction reactor.

U.S. Patent No. 5,958, 107 to Greenwalt discloses a system comprising a melter-gasifier 12, a first reduction reactor 10, and a second reduction reactor 44. The gas effluent from the first reduction reactor is scrubbed, compressed and treated in a shifter wherein CO is converted into H_2 with the purpose of decreasing its CO concentration so that the potential of carbon deposition in the gas heater 42 decreases. Greenwalt's system is inefficient because he does not teach or suggest to combine the recycled gas from the second reduction reactor 44 with the reducing gas effluent from the first reduction reactor 10 prior to CO_2 removal, therefore, the reducing potential of the recycled gas to reactor 44 is not advantageously improved. Greenwalt also fails to disclose the utilization of thermal energy in the system for producing steam used in the shifter 28.

U.S. Patents 5,997,608 and 5,997,609 to Diehl et al. describe a process for producing sponge iron wherein gas recycled from a second reduction reactor 27 is combined with a gas stream effluent from a first reduction reactor 1 and then the combined streams are stripped of CO_2 in a CO_2 removal unit 15. No shifter is suggested by Diehl.

U.S. Patents Nos. 5,676,732 and 5,882,579 to Viramontes-Brown et al. disclose a method and an apparatus for producing direct reduced iron utilizing a reducing gas produced in a melter-gasifier. The system described in these patents comprises a process wherein the water content of exhausted reducing gas effluent from a first reduction reactor 12 (containing a high content of carbon dioxide) is increased and then the humidified gas is treated in a shifter 36 for converting a carbon monoxide to carbon dioxide according to the water gas shift reaction: CO + H2O → CO2 + H2. Carbon dioxide is then withdrawn from the shifted gas in a CO_2 removal unit 42, thus considerably improving its reducing potential. This regenerated gas is utilized in a second reduction reactor 52 for production of direct reduced iron (DRI) 56. The system of these patents provides a useful and advantageous way of improving the DRI production using excess gas from a melter-gasifier.

The present invention further improves this system by integrating the energy available in the plant for producing steam which in turn is used for power generation, and then the resulting low-pressure steam is used in the shifter.

Documents cited in this text (including the foregoing listed patents), and all documents cited or referenced in the documents cited in this text, are incorporated herein by reference. Documents incorporated by reference into this text or any teachings therein may be used in the practice of this invention.

Objects of the Invention

It is therefore an object of the present invention to provide an improved process and apparatus for producing DRI utilizing a reducing gas which is prepared by partial oxidation of hydrocarbons in a melter-gasifier.

It is a another object of the invention to provide an improved method and apparatus whereby a second direct reduction reactor for producing DRI is advantageously combined with a hydrocarbon gasification plant allowing for the utilization of direct reduction in locations where natural gas is not readily available or its utilization for DRI production is not economically attractive.

It is a further object of the invention to provide a method and apparatus for increasing the efficiency of energy utilization in a direct reduction system comprising a melter-gasifier, a first (upstream) DRI reduction reactor and a second (downstream) DRI reduction reactor for increasing the capacity of DRI production, wherein according to the present invention this increased efficiency is accomplished by utilizing the thermal energy available in a gas stream effluent from a CO_2 separation unit for generating steam which is in turn used for power

generation utilized in the reduction plant and for carrying out the shift reaction of said gas stream.

Summary of the Invention

More particularly, the objects of the invention are generally achieved by providing a process for producing DRI utilizing reducing gases containing a high concentration of carbon monoxide from a liquid-metal production system comprising a smelter furnace producing a primary reducing gas; and a downstream reduction reactor wherein DRI is produced by utilizing upgraded reducing gas derived from said primary reducing gas, said process comprising subjecting reducing gas derived from said primary reducing gas to a scrubber cooler yielding a cooled derived reducing gas having from about 15% to about 30% by volume of hydrogen and at least about 40% by volume of carbon monoxide; adding water to humidify said cooled derived reducing gas; passing the humidified derived reducing gas through a shifter to react the carbon monoxide with water to yield more hydrogen as well as carbon dioxide; passing the shifted gas, together any with cooled recycle gas from said downstream reduction reactor through a CO2 adsorption unit whereby a CO2 lean gas stream and a CO2 laden gas stream are formed; producing DRI in said downstream reduction reactor by heating the CO2 lean gas stream and utilizing that as the upgraded reducing gas; using said CO2 laden gas stream as fuel to produce steam; and using said steam as the water for the reaction of carbon monoxide in said shifter.

In this process the primary gas can be fed directly to the scrubber cooler.

Alternatively, the smelter furnace can be a melter-gasifier further comprising an upstream reduction reactor wherein said primary reducing gas is utilized for DRI production therein and wherein a secondary gas comprising gas effluent from said upstream reduction reactor is the reducing gas derived from said primary reducing gas which is fed to the scrubber cooler.

Brief Description of the Drawings

Figure 1 is a schematic process diagram of the combination of a hydrocarbon gasification plant of the melter-gasifier type and two direct reduction reactors improved according to a preferred embodiment of the present invention.

Figure 2 is a schematic process diagram of the combination of an iron smelter furnace and a direct reduction reactor according to another preferred embodiment of the present invention.

Figure 2a is a schematic process diagram of a separate optional cooling structure that may optionally be added to the downstream flow of hot DRI from the discharge zone of the downstream DRI reduction reactor of Figure 2.

Detailed Description of Preferred Embodiments of the Invention

Referring to the attached Figure 1, the integrated system for reduction of iron oxides comprises a smelter furnace in the form of a melter-gasifier 10 fed with a hydrocarbon, preferably any suitable type of coal 12 and oxygen 14 for the partial combustion of said coal. Partial combustion of coal in the iron melt produces slag 18 and molten iron metal 21 which are withdrawn from the melter-gasifier 10 by known means. The primary effluent gas from the melter-gasifier 10 is a reducing gas containing carbon monoxide and hydrogen. This reducing is fed through pipe 16 to an upstream reduction reactor 20. Iron ore lumps or pellets or mixtures thereof 22 are introduced into said upstream reactor 20, wherein they are reduced into metallic iron (DRI) 23 and are discharged through conduit 24 for melting in melter-gasifier 10. A secondary (expended) reducing gas 19 from an upstream reduction reactor 20 is cooled down in a scrubber cooler 25 (typically also condensing and removing the oxidant water from the gas) and is then fed via pipe 26 to compressor 28 where its pressure is increased in order to be fed on into the reduction system of a downstream reduction reactor 86. This gas stream 26 typically contains from about 15% to about 30% by volume of hydrogen and at least about 40% by volume of carbon monoxide. The chemical reducing potential of such cooled expended gas leaving the compressor 28 via pipe 29 is still low, because it contains high concentrations of carbon dioxide and lower concentrations of carbon monoxide and hydrogen after having reacted with iron oxides in reactor 20 according to the following chemical reactions (not balanced):

$$Fe_xO_y + H_2 \rightarrow Fe + H_2O \qquad (a)$$

$$Fe_xO_y + CO \rightarrow Fe + CO_2 \qquad (b)$$

Therefore, it is necessary to increase the ratio of reducing agents H_2 and CO to oxidant agents H_2O and CO_2 so that these reducing gases may be efficiently utilized in the downstream reduction reactor 86. Water is condensed when cooling these reducing gases before their compression. The gases are then treated in a catalytic shifter 30 according to the water-gas shift reaction:

$$H_2O + CO \rightarrow CO_2 + H_2 \qquad (c)$$

After having transformed carbon monoxide to hydrogen through reaction (c) by feeding water to shifter 30 through pipe 32 provided with regulating valve 34, CO_2 is removed from the reducing gases by the CO_2 removal unit 38. This unit may be of the type

of pressure-swing adsorption, known as PSA, whereby CO_2 is concentrated in a gas stream 40, and a second gas stream 42 is recycled to reduction reactor 86 as an improved reducing gas (by reason of having a lower CO_2 concentration). The reducing potential of the reducing gases making up stream 42 is thereby significantly upgraded and can be effectively utilized in the downstream reduction reactor 86. The CO_2 removal unit 38 is sized for removing CO_2 from the two combined streams that flow into pipe 36 (namely, from (1) the depleted recycled effluent gas from the downstream reduction reactor 86 flowing through pipe 104 and from (2) the CO_2 high-concentration effluent gas flowing through pipe 35 from shifter 30.

The reducing gas leaving the CO_2 removal unit 38, improved to a high reducing potential, flows through pipe 42, is preheated in heat exchanger 70, and then flows on through pipe 72 to a direct-fired gas heater 74 where its temperature is raised to levels above about 830°C. Optionally, an oxygen-containing gas 78 (from a source 80), at a rate regulated by flow control valve 82, is mixed with the hot reducing gas from pipe 76 to carry out a controlled partial combustion to raise the temperature of the reducing gas to a higher desired level prior to being fed by pipe 84 into the reduction zone 88 for effecting the direct reduction of iron ores 90.

The reducing gas entering the reduction zone 88 preferably has a composition characterized by a ratio of H_2/CO in the range from 1.5 to 4.0 in volume percent; a pressure in the range from about 2 to about 7 bars absolute and a temperature in the range from 800°C to 950°C.

The reduction reactor 86 has an upper reduction zone 88 and a lower discharge zone 89. Particulate solid iron ores 90 in the form of lumps or pellets are contacted within the reduction zone 88 with a high-temperature upgraded reducing gas from pipe 84 comprising hydrogen and carbon monoxide and thereby producing direct reduced iron (DRI) 92. The DRI is discharged from said reactor 86 through the lower discharge zone 89. Depending on the type of subsequent utilization of the DRI, it may be discharged hot or cold.

If discharged at high temperature from said reactor 86, it can be subsequently briquetted for further storage and handling, or it can be hot-fed directly into a steel-making furnace.

If cold DRI is to be produced, the lower discharge zone 89 of reactor 86 may optionally have cooling means, well known in the art, (shown in dotted lines in Figure 1 indicating that it is an optional feature of the system) for circulating a stream of cooling gas from pipe 110 for cooling down the DRI in the discharge zone 89 to a temperature level below about 100°C before its discharge from said reactor 86. The heated cooling gas is

withdrawn from the discharge zone 89 of the reactor 86 via pipe 112 and is cooled down in water scrubber 114 and then is recycled back to the discharge zone 89 of the reactor 86 by means of pipes 116 and 110 through compressor 118. Cooling gas make-up is fed to the cooling gas loop from a suitable source 122. This make-up cooling gas 122 may, for example, be reducing gas from the melter-gasifier, reducing gas from the upstream reduction reactor 20, or natural gas.

In a modification of these embodiments of the invention (see the dash-dot structure in Figures 1 & 2), DRI 92 having been produced at high temperature may be cooled down to ambient temperature in a separate DRI cooling vessel 124 with a cooling gas system similar to the cooling gas system illustrated in dotted lines for the discharge zone 89 of the downstream reduction reactor 86. This modification may, for example, be used when there is interest in having the flexibility to produce hot DRI for its immediate melting or briquetting at a remote point and yet also have the capability to safely discharge the DRI cold for storage for later utilization.

Spent reducing gas exits as an effluent from the reduction zone 88 at a temperature in the range from about 300°C to about 500°C via pipe 94 and is upgraded in a recycle circuit and returned back to the reduction zone 88. Such recycle reducing gas initially passes through a heat exchanger 70 (where its sensible heat is used to preheat the downstream upgraded portion of the reducing gas, from pipe 42, just prior to being recycled back into the reduction zone 88). The spent reducing gas, now partially cooled, flows on through pipe 96 into a cooler/scrubber 98, and is there cleaned and cooled down to ambient temperature by direct contact with water 100. The spent reducing gas effluent from the reduction zone 88 contains significant amounts of water and carbon dioxide (produced as by-products from the reactions of hydrogen and carbon monoxide with the iron oxide content of the iron ore 90). The upgrading of the reducing gas effluent begins in the cooler/scrubber 70, where the water produced by the hydrogen reduction reaction condenses and is extracted from the system through pipe 102 along with the hot effluent from the cooling water 100.

A minor portion of the cleaned and dewatered spent gas is purged from the recycle circuit through pipe 105 having a pressure control valve 107 (for pressure control of, and for maintaining a N_2 concentration below 13% by volume in, the recycle circuit). The purged gas may be advantageously utilized as fuel for the gas heater 74 and optionally, if needed, may also be supplemented with some natural gas from a source 109 or with gas from the melter-gasifier 10. The remaining portion of the cleaned and dewatered reducing effluent gas is then transferred to compressor 106 through pipe 104, wherein its pressure is raised to a

level suitable for its ultimate recycling to the reduction zone 88 of the reactor 86 (after CO_2 removal and heating, as previously described, in units 38 and 74 etc.).

The gas in pipe 40, with its high concentration of CO_2, (derived from the CO_2 removal unit 38), is not effective for reduction of iron oxides in reactor 86. However, it still contains useful calorific value. Consequently, according to this embodiment of the present invention, this gas is used to flow from pipe 40 into boiler 43 to provide the heat for generation of steam. Water from a suitable source 44 is supplied via pipe 46 to boiler 43, and the steam produced is utilized to drive turbine 62 and its attached generator 64 to produce electric power. Preferably, this electric power generation, using the high-pressure steam produced in the boiler 43, makes for full utilization of the heating value of the CO_2 laden stream (when combined with the subsequent use of the steam after discharge from the turbine 62 via pipe 32 in the shifter 32). However, in an alternative embodiment, some plants may only produce a low-pressure steam in boiler 43 for direct use of the steam in the shifter 30. In the illustrated preferred embodiment, the same effect can be achieved by use of a by-pass pipe 66 and a control valve 68 to by-pass the turbine and direct the steam directly to the shifter 30 in case of shut down of the turbine 62 for maintenance, temporary diminished power needs, malfunction, or the like.

Referring now to Figure 2, (showing another embodiment of the invention and wherein same numerals shown in Figure 1 represent equivalent elements in Figure 2) a smelter furnace 10 is charged with iron ore materials 22 and/or prereduced iron materials 23, and coal 12 and oxygen gas 14 as described in connection with Figure 1. Partial oxidation of coal 12 with oxygen 14 produces heat for melting said iron materials and for carrying out or completing the reduction of the iron-containing ore charge to said smelter furnace. Molten iron metal 21 and slag 18 are withdrawn from smelter 10 in a manner known in the art. Gases produced by the partial combustion of coal or other hydrocarbon in the smelter 10 are extracted as the primary reducing gas through pipe 16 and cooled and cleaned in the scrubber cooler 25. The cooled and clean gases then flow through pipe 26 to compressor 28 for further processing and ultimate utilization in the reduction of iron ores in reduction zone 88 of the reactor 86 as described above with reference to Figure 1.

The process of the invention offers a number of advantages over the prior art. For example, it requires less energy (thermal energy) in the overall system for producing one ton of DRI. By separating the carbon dioxide and other acid-gases in a PSA unit 38, the CO_2 containing stream 40 is used as a source of fuel gas for steam generation in external boilers

which is used for power generation and for providing the necessary water for the shifter 30 and
/or 124. This is in addition to the known practice of preheating the recycle reducing gas using available energy from the top gas effluent from the reactor 86 in a heat exchanger, before being further heated up in the gas heater 74.

Another advantage of the invention over the prior art is the flexibility to produce high quality DRI from any source of gas such as Natural gas, Reformed gas, Coke oven gas, Syngas, etc. while maintaining the same process configuration.

Still a further advantage of the invention is the minimized reducing gas requirements and complete control of the gas chemistry entering the DR Reactor, e.g. the gas temperature is controlled at levels above 950°C by a controlled addition of oxygen. The high temperature operation entails higher process efficiency. The pressure level of the system of the invention may be from about 1 to about 10 bars absolute.

Providing a shifter 30 operating with steam produced by utilization of thermal energy available in the reduction system, permits adequate control of the gas composition from the upstream reduction reactor, so that the H_2 / CO ratio is higher than 1, and results in a better reduction of iron ores, a lower disintegration of the ore during the reduction process, and the possibility of heating the reducing gas to temperatures above about 900°C, without the need of oxygen (from source 80) and thus assuring a higher quality of the upgraded reducing gas in pipe 84.

It is of course to be understood that in this specification only some preferred embodiments of the invention have been described for illustration purposes and that the scope of the invention is not limited by such described embodiments but only by the scope of the appended claims.

What is claimed is:

1. A process for producing DRI utilizing reducing gases containing

a high concentration of carbon monoxide from a liquid-metal production system comprising a smelter furnace producing a primary reducing gas; and

a downstream reduction reactor wherein DRI is produced by utilizing upgraded reducing gas derived from said primary reducing gas,

said process comprising

subjecting reducing gas derived from said primary reducing gas to a scrubber cooler yielding a cooled derived reducing gas having from about 15% to about 30% by volume of hydrogen and at least about 40% by volume of carbon monoxide;

adding water to humidify said cooled derived reducing gas;

passing the humidified derived reducing gas through a shifter to react the carbon monoxide with water to yield more hydrogen as well as carbon dioxide;

passing the shifted gas, together any with cooled recycle gas from said downstream reduction reactor through a CO_2 adsorption unit whereby a CO_2 lean gas stream and a CO_2 laden gas stream are formed;

producing DRI in said downstream reduction reactor by heating the CO_2 lean gas stream and utilizing that as the upgraded reducing gas;

using said CO_2 laden gas stream as fuel to produce steam; and

using said steam as the water for the reaction of carbon monoxide in said shifter.

2. A process according to claim 1, wherein said primary gas is fed directly to the scrubber cooler

3. A process according to claim 1, wherein said smelter furnace is a melter-gasifier further comprising an upstream reduction reactor wherein said primary reducing gas is utilized for DRI production therein and wherein a secondary gas comprising gas effluent from said upstream reduction reactor is the reducing gas derived from said primary reducing gas which is fed to the scrubber cooler.

4. A process according to any one of claims 1 to 3, wherein said steam is generated in a boiler fed with said CO_2 laden gas stream as fuel, further comprising using said steam for power generation.

5. A process according to any one of claims 1 to 4, further comprising hot discharging DRI produced in said downstream reduction reactor at high temperature above about 400°C.

6. A process according to any one of claims 1to 4, further comprising discharging DRI produced in said downstream reduction reactor at a temperature below about 100°C by circulating a cooling gas through said DRI before it is discharged from said downstream reduction reactor.

7. A process according to any one of claims 1 to 4 or 6, further comprising cooling said DRI produced in the downstream reduction reactor in a cooling vessel separate from said downstream reduction reactor.

8. A process according to any one of claims 6 or 7, further comprising using natural gas as cooling gas.

9. A process according to any one of claims 1 to 8, wherein the pressure level of said downstream reduction reactor is in the range from about 1 to about 10 bars absolute.

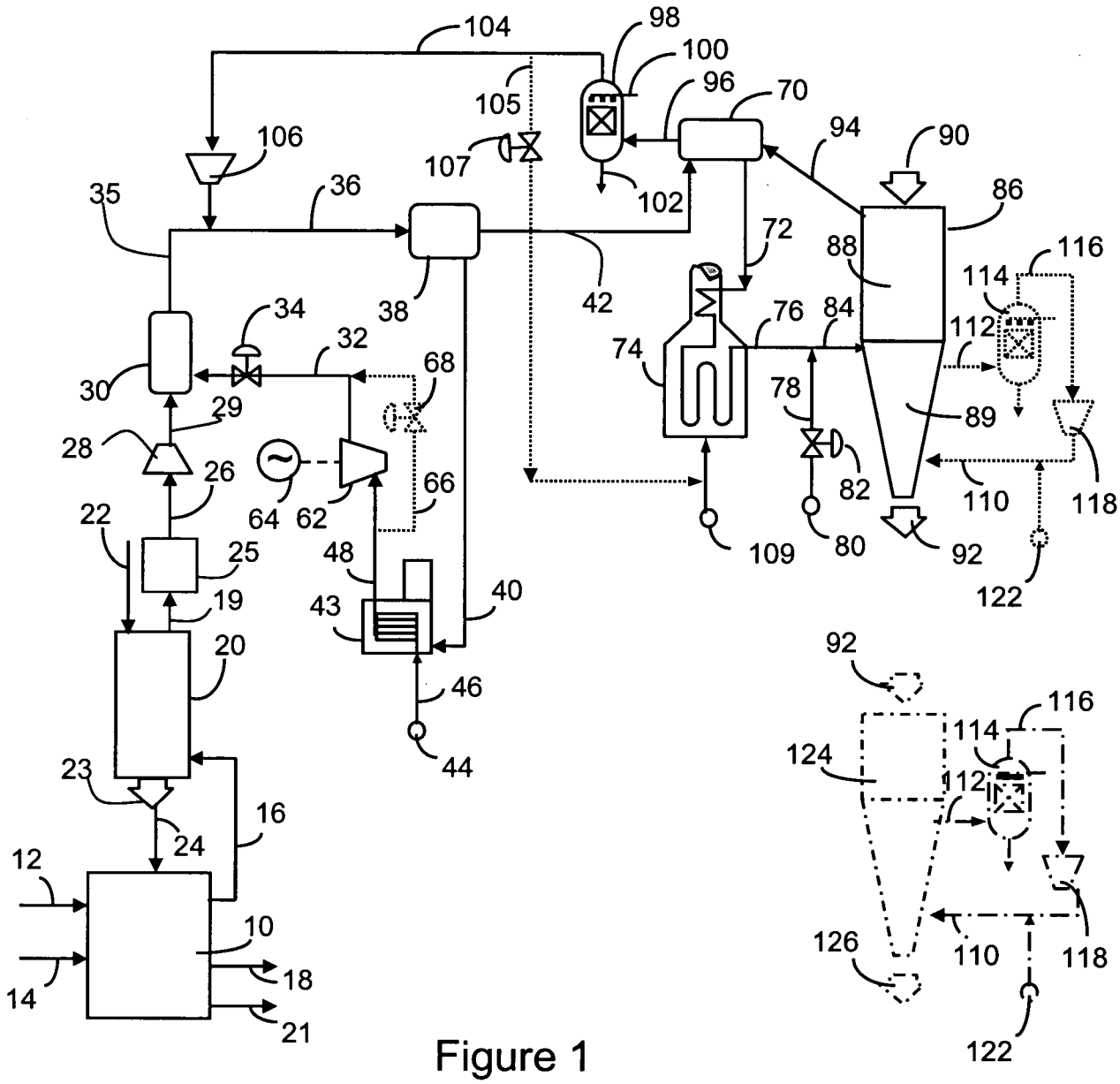
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Figure 1

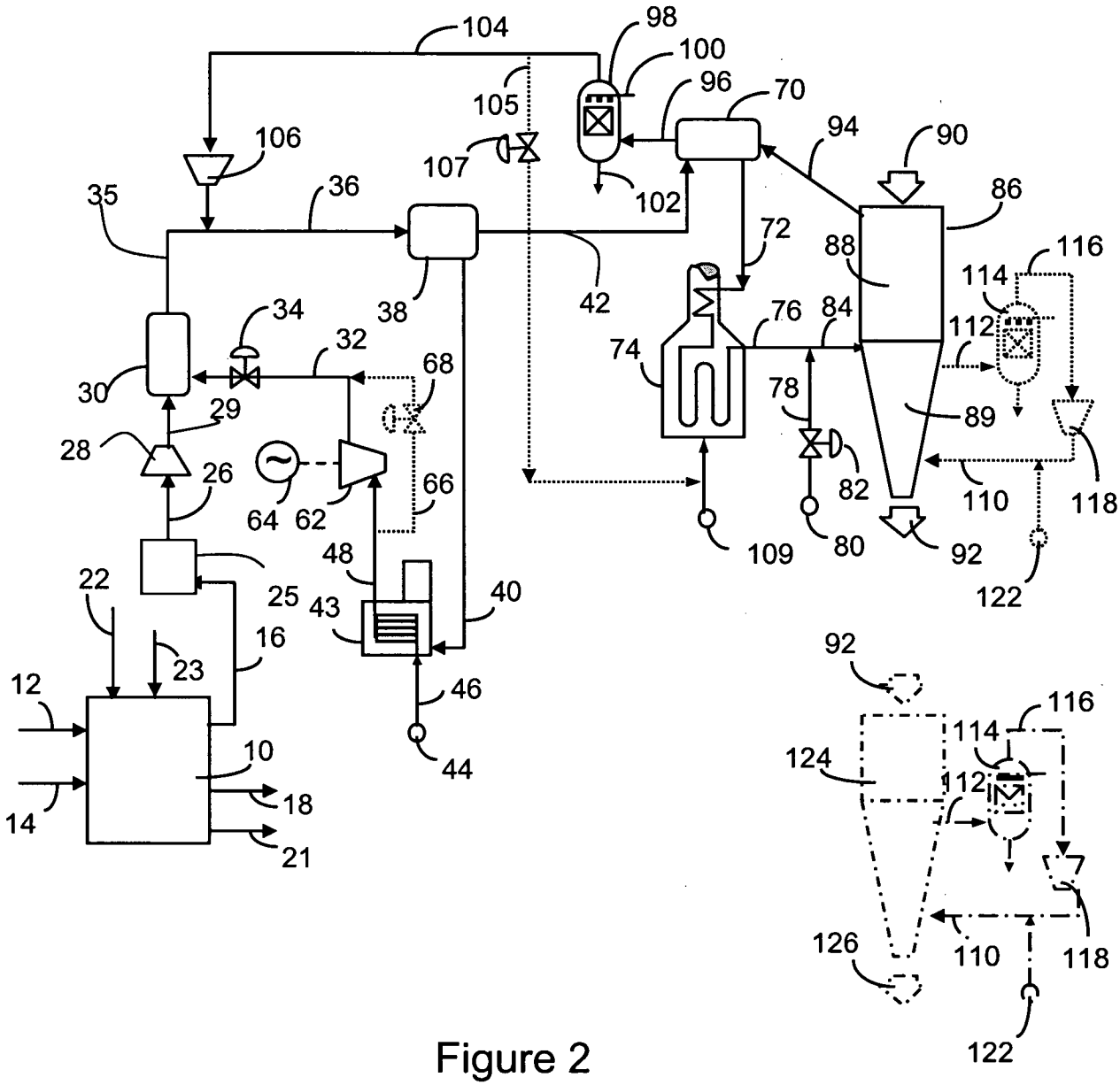
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Figure 2