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#### (54) METHOD AND APPARATUS FOR THE DIRECT REDUCTION OF IRON ORES UTILIZING SYNGAS

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### (57) **ABSTRACT**

A direct reduction plant for the production of a metallized product (DRI) by the reduction of iron ores in lump or pellet form, wherein the reducing gas utilized in the DRI reactor contains acid gases such as sulphur compounds and carbon dioxide. The make-up reducing gases are typically produced by partial oxidation of hydrocarbons (syngas) at a high pressure while the DRI reactor is usually operated at a lower pressure. The pressure level of the reducing gas effluent from the DRI reactor (top gas), after cooling and dewatering, is increased to the pressure level of the syngas and the resulting recycle reducing gas is then combined with the make-up syngas and treated in a single acid-gas absorption unit forming a combined stream of clean upgraded reducing gas which thereafter is expanded in a turbine for lowering combined reducing gas pressure to the pressure level of the DRI reactor and is then heated to a temperature preferably above 950° C. and used in the DRI reactor for producing said DRI. The compressor for raising the pressure of the initially cool recycle gas uses energy from the expansion turbine (used for lowering the pressure of the high-pressure combined reducing gas).





Figure 1

#### METHOD AND APPARATUS FOR THE DIRECT REDUCTION OF IRON ORES UTILIZING SYNGAS

#### FIELD OF THE INVENTION

**[0001]** The invention relates to the direct reduction of iron ores in a reduction system comprising a direct reduction process for producing direct-reduced-iron (DRI) and a gasification process for producing hydrogen and carbon monoxide produced by partial oxidation of a hydrocarbon source.

#### BACKGROUND OF THE INVENTION

**[0002]** Direct reduction of iron ores for producing prereduced 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 and that the metallic iron is produced in solid form with low sulfur and silicon content and that the DRI may be easily melted in electric-arc furnaces. The reducing agents utilized in the direct reduction plants are hydrogen and carbon monoxide, most typically 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.

**[0003]** Hydrogen and carbon monoxide (syngas) can also be produced by partial oxidation of hydrocarbons other than natural gas, for example heavier gases such as propane, butane, etc, oil and coal. One of the disadvantages of utilizing heavy hydrocarbons is the need of sulfur and other acid gases removal and also that the gasification processes normally operate at relatively higher pressure levels as compared with the pressure of the direct reduction processes.

[0004] U.S. Pat. Nos. 5,744,067; 6,033,456 and 6,149,859 to Jahnke et al. describe an integrated process for supplying the reducing gases to a direct reduction plant. In this proposed solution, high-pressure syngas is fed to the direct reduction process by lowering the pressure through expansion in a turbine. This utilizes the energy from said expansion to drive the recycle gas compressor. The integrated process of Jahnke et al comprises two acid gas absorber units, one operating at the high pressure level of the hydrocarbons gasifier and the other operating at the low pressure level of the direct reduction plant. Jahnke teaches that the integrated process is improved by utilizing a single solvent regeneration unit (stripper) common to both acid-gas absorbers. The regenerated acid-gas-loaded solvent effluent from the common stripper may be utilized in a parallel or series arrangement with respect to the absorption units.

**[0005]** Jahnke however does not teach nor even recognize the possibility of having a single absorption unit wherein the acid-gas content is removed from a combined stream of both the syngas produced in the gasifier and the recycle reducing gas from the reduction reactor as in the present invention.

**[0006]** The direct reduction process of the present invention operates with a single absorption unit at the pressure range of the partial oxidation process and by this offers a number of previously unrecognized advantages over the prior art.

**[0007]** 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**

**[0008]** It is therefore an object of the present invention to provide a process and apparatus for producing DRI utilizing a reducing gas which is prepared by partial oxidation of hydrocarbons.

**[0009]** It is another object of the invention to provide a method and apparatus whereby a direct reduction plant 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.

**[0010]** It is a further object of the invention to provide a method and apparatus for increasing the efficiency of energy utilization in a dire& reduction system for producing DRI wherein some energy of the high pressure syngas from a hydrocarbon gasification plant is utilized for driving compressors used in the reduction plant.

#### SUMMARY OF THE INVENTION

**[0011]** The objects of the invention are generally achieved by providing a method for producing DRI utilizing a synthesis gas produced preferably by partial oxidation of hydrocarbons, wherein the pressure level of the expended reducing gas effluent from the DRI reactor (top gas) is increased by a compressor to the pressure level of the syngas and the recycle top gas is combined with the make-up syngas and treated in a single acid-gas absorption unit thus forming a combined stream of cleaned reducing gas which is expanded in a turbine for lowering its pressure to the pressure level of the DRI reactor and using the turbine to drive the compressor and thereafter the combined stream is then heated to a temperature preferably above 950° C. and used in the DRI reactor for producing said DRI.

#### BRIEF DESCRIPTION OF THE DRAWINGS

**[0012]** FIG. 1 is a schematic process diagram of the combination of a hydrocarbon gasification plant and a direct reduction plant (having a moving bed reduction reactor) according to a preferred embodiment of the invention.

#### DETAILED DESCRIPTION OF PREFERRED EMBODIMENTS OF THE INVENTION

[0013] Referring to the attached FIG. 1, the integrated system for reduction of iron oxides comprises a hydrocarbon gasification plant 10 and a direct reduction plant 12. The reduction plant 12 comprises a reduction reactor 14 having an upper reduction zone 16 and a lower discharge zone 20. Particulate solid iron ores 18 in the form of lumps or pellets are contacted in the reduction zone 16 with a high-temperature reducing gas from pipe 64). This first stream of good quality high-temperature reducing gas in pipe 64 mainly is comprised of hydrogen and carbon monoxide which react with the iron ores to produce direct reduced iron (DRI) 22. The DRI is discharged from said reactor 14 through the lower discharge zone 20. 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 14, 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 **20** of reactor **14** may optionally have means, well known in the art, for circulating a stream of cooling gas for cooling down said DRI to a temperature level below about  $100^{\circ}$  C. before its discharge from said reactor. See for example the cooling/discharge zone **14** with an associated cooling gas loop shown in U.S. Pat. No. 4,524,030.

[0014] Spent reducing gas exits as an effluent from the reduction zone 16 at a temperature in the range from about 300° C. to about 500° C. via pipe 24 (for upgrading in a recycle circuit and return back to the reduction zone 16). Such recycle reducing gas initially passes (as the second stream) through a heat exchanger 26 (where its sensible heat is used to preheat the downstream upgraded portion of the reducing gas just prior to being recycled back as said first stream into the reduction zone 16). The spent reducing gas, now partially cooled, flows on through pipe 28 into a cooler/scrubber 30, and is there cleaned and cooled down to ambient temperature by direct contact with water 32. The spent reducing gas effluent from the reduction zone 16 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 18). The upgrading of the reducing gas effluent begins in the cooler/scrubber 30, where the water produced by the hydrogen reduction reaction condenses and is extracted from the system through pipe 34 along with the cooling water 32.

[0015] A minor portion of the cleaned and dewatered spent gas is purged from the recycle circuit through pipe 82 having a pressure control valve 84 (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 in burner 86 for the gas heater 58 and optionally if needed may also be supplemented with some syngas from gasifier 90 via pipes 46 and 45. The remaining portion of the cleaned and dewatered reducing effluent gas is then transferred to compressor 38 through pipe 36 (as the third stream), wherein its pressure is raised to a level suitable for further treatment prior to its ultimate recycling to reactor 14.

**[0016]** Depending on the operating pressure level of the gasification unit 90, an additional recycle gas compressor 42, in serial arrangement with compressor 38, may be required to increase the pressure of the recycle reducing gas in pipe 44 up to the pressure level of the  $CO_2$  removal column unit 48. Such supplemental compressor 42 may be driven by a motor 43.

[0017] According to the invention, the  $CO_2$  removal unit 48 simultaneously serves both the normally relatively low pressure recycle reducing gas and also the syngas necessarily produced at high pressure by the gasifier 90.

**[0018]** In the gasifier **90**, fossil fuels such as coal, refinery residues and biomass are partially oxidized with oxygen **92** and steam **94**. The resulting syngas contains  $H_2$ , CO, CO<sub>2</sub>,  $H_2O$ , CH<sub>4</sub>, and S (in the form of  $H_2S$  and/or COS, CS<sub>2</sub>, mercaptans), and dust. This syngas exits through pipe **96** and is cooled and cleaned in a dedicated scrubbing system **98**. The gasification waste by-products exit the gasifier **90** by discharge pipe **112**.

**[0019]** After quenching and cleaning in scrubber **98**, the composition of the syngas exiting via pipe **100** may optionally be adjusted to increase the hydrogen content by a gas shifting reaction to obtain a  $H_2/CO$  ratio of 2 to 3 (measured by % volume) in a shifter unit **102**. Steam may be supplied as

the reactant for the shift reaction through pipe **104** in amounts metered by a flow control valve **106**.

[0020] The shifted syngas is then passed through pipe 108 to another cooler/scrubber 110.

[0021] The clean syngas, shifted or not, then flows through pipe 46 while still at a gasification system pressure typically in the range of 20 to 40 bars absolute. This high pressure syngas, being supplied as make-up reducing gas, combines with the recycle reducing gas at like pressure in pipe 44. The resulting supplemented recycle reducing gas flows via pipe 47 (as the fourth stream) of for subsequent further upgrading in a  $CO_2$  absorption unit 48 (where acid-gases, e.g. sulphur compounds and carbon dioxide, are removed from the combined reducing gas stream). The CO<sub>2</sub> removal unit preferably is a conventional amine solution-based absorption system, comprising an absorption column 49 and stripping column 72 with related facilities, such as heat exchangers and steam re-boiler 74 (where steam 76 heats the amine solution received from the absoption column 49 via pipe 70 to eliminate CO<sub>2</sub> and sulphur-containing gases by discharge through outlet pipe 78). The regenerated amine solution is returned via pipe 80 and reused in the absorption column 49.

[0022] The upgraded recycle reducing gas stream flowing through pipe 50 is then depressurized in a gas expansion turbine 52, which is used to drive the compressor 38 (thus achieving an overall mechanical energy synergy between the gasification and the reduction systems). The upgraded reducing gas stream, now typically at a lower pressure in the range of 6 to 10 bars absolute, passes on via pipe 54 (as the fifth stream) to be preheated in heat exchanger 26 and then further passes on through pipe 56 into gas heater 58; where its temperature is raised to levels above about 800° C. As is known in the art, higher temperatures are desirable for faster and more efficient reactions, but are limited by undesirable agglomeration of the ores and DRI preventing the free flow of particles in and from the reactor 14. Temperatures of as high as at least 1020° C. have been used, but require special selection of temperature tolerant ores and/or special coating of the ore particles. Optionally, as an aid to obtaining reducing gas temperatures at the higher end of the range, an oxygen-containing gas 62, at a rate regulated by flow control valve 63, is mixed with the hot reducing gas from pipe 60 to carry out a controlled partial combustion to raise the temperature of the reducing gas to a higher desired level, prior to the upgraded recycle reducing gas being fed by pipe 64 (as the first stream) into the reduction zone 16 for effecting the direct reduction of iron ores 18.

**[0023]** The reducing gas entering the reduction zone 16 preferably has a composition characterized by a ratio of  $H_2/CO$  in the range from 1.5 to 4.0 in volume percent and at a pressure in the range from about 2 to about 7 bars absolute and at a temperature in the range from about 830° C. to about 1020° C.

**[0024]** The process of the invention offers a number of advantages over the prior art. For example, it requires less steam (thermal energy) for regenerating the amine solution utilized in the absorption unit **48**. By operating the stripper column **72** at a significantly lower relative pressure, separation of the carbon dioxide and other acid-gases out from the amine solution is improved. The steam can be generated by using waste energy from the syngas process in lieu of being produced in external boilers. The mechanical energy in the DR process is particularly improved, since the pressurized syngas from the gasifier is used to drive the recycling of the

reducing gas. This is in addition to the known practice of preheating the recycle reducing gas using available energy from the top gas in a heat exchanger, before being further heated up in the gas heater. Alternatively, the thermal energy of the top gas can be used for low-pressure steam generation, which can be used in the  $CO_2$  removal system.

**[0025]** 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 in a reduction reactor utilizing make-up reducing gases containing sulphur compounds and/or carbon dioxide from a high-pressure source at a pressure level higher than the mean pressure level of operation of the DRI reactor, said process comprising reacting in the reactor at a temperature above about 800° C. lumps or pellets of iron ore with a first stream of recycle reducing gas that has been upgraded to be effective to produce said DRI, the great majority of which stream comprises hydrogen and carbon monoxide; withdrawing as a second stream the depleted reducing gas from said reduction reactor for recycling and subsequent upgrading; scrubbing and cooling said second stream for removing water thus producing a third stream of cold and dewatered recycle reducing gas; increasing the pressure of at least a portion of said third stream and combining such portion with the make-up reducing gases from the high-pressure source to form a fourth stream of high-pressure recycle reducing gas; removing acid gases including said sulphur compounds and carbon dioxide from said fourth stream to form a fifth stream of clean upgraded recycle reducing gas; lowering the pressure of said fifth stream by expansion in a turbine and using energy derived from such expansion to achieve the increase in pressure of said third stream; and increasing the temperature of said fifth stream to above about 800° C. to form said first stream.

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**2**. A process according to claim **1**, wherein said reduction reactor is a moving bed reactor and the DRI is continuously discharged.

3. A process according to claim 2, wherein the temperature of said fifth stream is raised above  $830^{\circ}$  C.

**4**. A process according to claim **3**, wherein said reducing gases containing sulphur and/or carbon dioxide are produced by partial oxidation of hydrocarbons.

5. A process according to claim 4, wherein said hydrocarbon is a liquid derived from oil.

6. A process according to claim 4, wherein said hydrocarbon is coal.

7. A process according to claim **4**, wherein the pressure level of the source of said reducing gases is in the range from 20 to 40 bars absolute.

**8**. A process according to claim 7, wherein the pressure level in the reduction reactor is in the range of 2 to 7 bars absolute.

**9**. A process according to claim **8**, wherein the pressure level of said fifth stream is in the range of 6 to 10 bars absolute.

10. A process according to claim 9, wherein temperature ranges from about  $950^{\circ}$  C. to about  $1020^{\circ}$  C.

11. A process according to claim 10, wherein the composition of the first stream has a composition with a ratio of  $H_2/CO$  in the range from 1.5 to 4.0 in volume percent.

12. A process according to claim 3, wherein said reducing gases containing sulphur and/or carbon dioxide are produced as a syngas by partial oxidation of composition chosen from the group consisting of fossil fuels and biomass.

13. A process according to claim 2, wherein the composition of the make-up reducing gases when combined with the third stream has a composition with a ratio of  $H_2/CO$  in the range from 2 to 3 in volume percent.

14. A process according to claim 1, further comprises cooling DRI in a cooling zone of said reduction reactor.

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