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IRON ORE REDUCTION PROCESS

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2 Sheets-Sheet 1

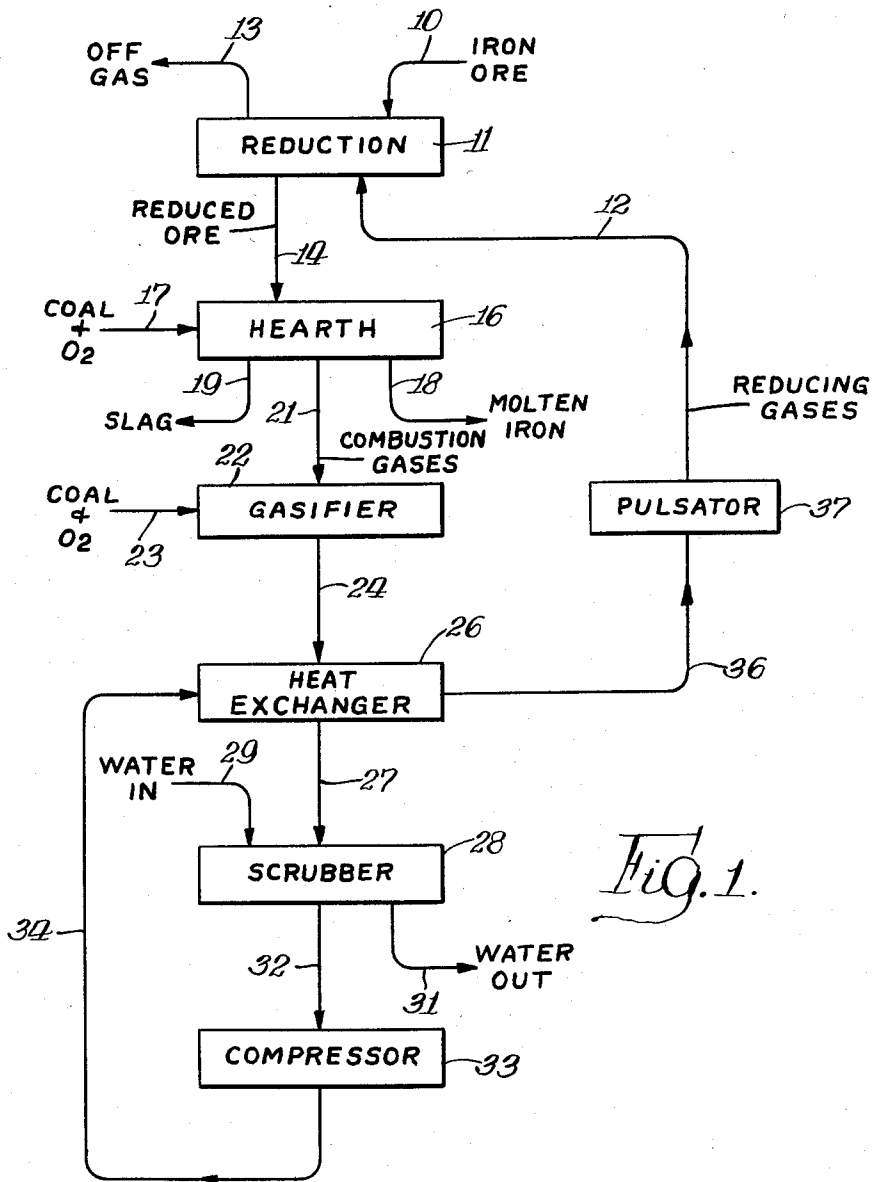


Fig. 1.

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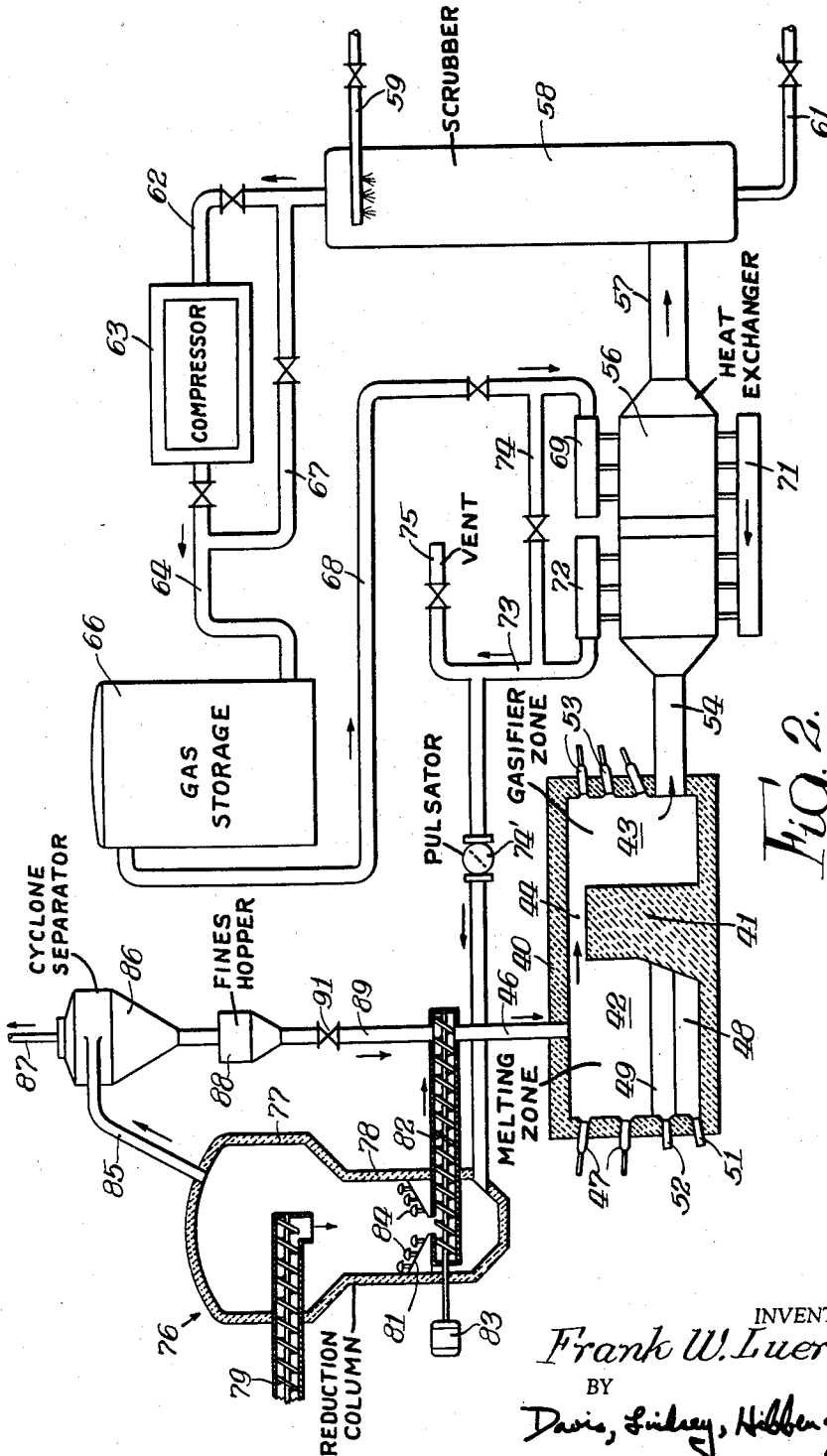
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6 Claims. (Cl. 75—34)

This invention relates to the direct reduction of iron oxide, particularly iron oxide ores, by the use of a reducing gas comprising principally carbon monoxide.

Many different processes have been thought of and tried for the production of metallic iron from iron ores. However, except for unusual raw material conditions, the use of the conventional blast furnace has been considered to be the most economical and commercially feasible technique for the production of pig iron. In spite of its widespread use, the blast furnace suffers from two serious handicaps associated with the raw materials which are charged to the blast furnace: (1) the iron ore used in the blast furnace must have a fairly large particle size so that it will not be blown out of the top of the furnace, and (2) the only really satisfactory fuel for use in the blast furnace is metallurgical coke which possesses sufficient strength to support a column of solids in the furnace.

Both of the aforementioned limitations on the blast furnace raw materials are becoming increasingly acute. For example, to mention only one aspect of the ore problem, various iron ore concentrating processes now in use produce a finely divided material which cannot be charged as such to the blast furnace but first must be agglomerated into larger pieces. Obviously, this necessary step adds considerably to the cost of operating the blast furnace. With respect to coke, under present day conditions the supply of good coking coals is rapidly decreasing and the cost of coke plants is increasing. As a result, the cost of coke today represents the greatest single item in the conversion of iron ore to pig iron in the blast furnace.

It has long been known that subdivided or granular iron ore can be converted to a mixture of metallic iron and gangue by treating it at an elevated temperature with suitable reducing gases. In fact, the prior art is replete with disclosures and suggestions of various types of direct reduction processes. However, for many different reasons, none of the suggested processes have been adopted on a commercial scale. My invention involves a unique process flow arrangement for the direct reduction of iron ore by means of reducing gases obtained by combustion of pulverized solid carbonaceous fuels such as coal or the like. As will hereinafter appear, the use of fuels such as coal results in the presence of coal ash in the combustion gases thereby introducing certain technical problems which must be overcome in order to realize an efficient and commercially feasible process.

Accordingly, a primary object of the invention is to provide a novel and improved process for the direct reduction of iron ore.

A further object of the invention is to provide a novel process of the foregoing character which utilizes a reducing gas obtained from the combustion of coal or like solid carbonaceous fuel.

Another object of the invention is to provide an iron ore reduction process utilizing a reducing gas obtained by the combustion of coal or the like and including novel means for overcoming coal ash and related problems which arise from such combustion.

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Other objects and advantages of the invention will become apparent from the subsequent detailed description taken in conjunction with the accompanying drawings; wherein:

5 Fig. 1 is a block flow diagram illustrating a preferred method of practicing the invention; and

Fig. 2 is a generally diagrammatic flow sheet illustrating the process in somewhat more detail.

10 Broadly speaking, my invention involves three principal process stages as applied to an integrated reduction and melting process: (1) a combined melting and gas generator zone wherein a fuel is burned in order to melt and separate the iron from the reduced ore and at the same time producing combustion gases, (2) an upgrading or gas enrichment zone wherein the combustion gases from the first zone are treated to increase their reducing capacity, and (3) a reduction zone wherein subdivided iron ore is contacted with the enriched reducing gases from the second zone and from which the resultant reduced iron ore is passed to the first zone.

20 Referring first to Fig. 1, subdivided or granular iron oxide ore is introduced through a line 10 to a reduction zone 11 and contacted with CO-rich reducing gases introduced to the zone 11 through a line 12. The ores which may be used in the process comprise any of the well known iron oxide ores including hematite, magnetite, and others which may contain at least about 5 wt. percent and as much as 45 wt. percent gangue materials, particularly silica and alumina. Other ores similar to iron ore such as iron-manganese ores may also be used. It is also within the scope of the invention to charge to the reduction zone 11 other iron oxide materials such as mill scale, etc. In the reduction zone 11 the well known reduction reactions of iron oxide with CO (and H₂) take place with the heat of reaction being supplied by the sensible heat of the reducing gases which are at a temperature of from about 900° F. to about 1800° F. as hereinafter described. Effluent reducing gases of depleted CO content are discharged through a line 13 from the reduction zone 11.

40 The resultant ore particles comprising reduced iron, gangue, and a certain amount of unreduced iron oxide pass from the reduction zone 11 through a line 14 to a hearth zone 16 which comprises a combined melting and gas generator zone. A solid carbonaceous fuel is introduced in admixture with a high oxygen content gas through a line 17 and combustion of the fuel with the oxygen takes place within the hearth zone 16. In order to supply the thermal requirements of the melting operation, the combustion of the fuel in the hearth zone 16 must be sufficiently complete so that an excess of CO₂ is produced with the result that the combustion gases in the hearth zone 16 are oxidizing to iron. The preferred solid carbonaceous fuel burned in the hearth zone 16 is coal such as anthracite, bituminous or sub-bituminous coal in pulverized form. The oxygen-rich gas introduced with the fuel must contain at least 85% oxygen, e.g. a commercial grade of straight oxygen which may be 98–99% pure, or oxygen enriched air. As mentioned above, the relative quantities of coal and oxygen and the other combustion conditions in the hearth zone 16 are regulated so that the exit combustion gases from the hearth zone are oxidizing in character with a CO₂:CO ratio of at least about 1. In this way, a temperature of from about 2900° F. to about 3500° F. is obtained in the hearth zone so that the heat of combustion is sufficient to insure melting of the iron in the reduced ore particles thereby separating the same from the gangue of the ore.

70 As a result of the combustion of solid carbonaceous fuel, particularly coal, in the hearth zone 16, there is an unavoidable introduction of non-combustible ash to the hearth zone which must be removed from the system. A

certain amount of the ash is removed by formation of slag in the hearth zone from the coal ash and also from the gangue in the ore. This fluid slag formed in the hearth zone acts as a protective layer or blanket overlying the molten iron in the hearth so as to protect the latter against reoxidation by the oxidizing combustion gases. Molten iron and slag may be tapped from the hearth as desired through lines 18 and 19, respectively. Excessive reoxidation of the molten iron in the hearth 16 is also retarded by the presence of extraneous carbon either in the form of coal fall-out during the combustion of coal in the hearth or as the result of direct addition of coal or other carbonaceous solid to the surface of the molten iron. In the case of coal fall-out, the combustion of coal with the oxygen-rich gas is controlled so that there is a certain amount of fall-out or deposition of excess burned coal which is rapidly assimilated in the liquid-slag metal system. The carbon content of the coal effects additional reduction of iron oxide in the hearth zone 16 and also effects carburization of the molten iron to the usual pig iron or hot metal level of from about 2 wt. percent to about 4 wt. percent.

Since the combustion gases produced in the hearth zone 16 contain an excess of CO_2 , they must be subjected to an upgrading or enrichment treatment before they can be used for ore reduction purposes. Preferably, the upgrading or enrichment of the combustion gases is effected by reducing the CO_2 content by reaction with carbon. Thus, the combustion gases pass from the hearth zone 16 through a line 21 to a gasifier zone 22 to which oxygen and an excess of coal are also supplied through a line 23. The carbon in part of the coal supplied at 23 reacts with the CO_2 in the combustion gases supplied at 21, and the endothermic heat requirements for the reduction reaction are furnished by the sensible heat in the combustion gases supplemented by additional heat evolved in the gasifier 22 by partial combustion of another part of the coal with the oxygen supplied through the line 23. The high oxygen content gas supplied at 23 may be straight commercial grade oxygen or may be an oxygen enriched gas containing at least 85% oxygen just as in the hearth 16. Although reaction of CO_2 with the carbon content of the coal to produce CO is the primary reduction reaction accomplished in the gasifier zone 22, it will also be appreciated that water vapor contained in the combustion gases fed to the gasifier zone 22 will be reduced by reaction with carbon to form H_2 .

For best results, it is desirable that the CO_2 content of the combustion gases from line 21 be decreased by reduction in the gasifier 22 to the extent that the exit gases removed through a line 24 contain not more than about 10% CO_2 with a $\text{CO}:\text{CO}_2$ ratio of at least about 7, and preferably not more than about 5% CO_2 with a $\text{CO}:\text{CO}_2$ ratio of at least about 10, in order to provide the necessary reducing capacity for use in the direct reduction of iron oxide ores.

At this point in the process, before the CO-rich gases discharged through the line 24 can be used for reducing purposes in the reduction zone 11, it is essential to overcome several technical problems introduced by reason of the procedures carried out in the hearth zone 16 and in the gasifier zone 22. First of all, the reduction reactions carried out in the gasifier zone 22 are highly endothermic so that there is obviously a certain minimum temperature level at which the gasifier zone 22 can be operated for commercially practical results. Generally speaking, the CO-rich gases leaving the gasifier through line 24 are at a temperature of from about 1900° F. to about 2400° F., and before these reducing gases can be employed in the reduction zone 11 they must be cooled to a somewhat lower temperature level. Of even greater importance, however, is the fact that the gases removed through the line 24 invariably contain substantial amounts of coal ash which must be removed. As previously mentioned, part of the ash introduced at the hearth

zone 16 by combustion of coal therein is converted to slag and removed through the line 19. However, a certain amount of ash particles are entrained in the combustion gases and pass to the gasifier zone 22 wherein additional coal ash is introduced by reason of the further combustion coal introduced at 23. Here again, part of the ash may be removed from the gasifier zone 22 as a semi-solid slag or ash deposit, but the operating conditions required in the gasifier zone 22 are such that a troublesome quantity of coal ash necessarily escapes from the gasifier zone 22 as entrained particles in the exit gases. Although a gas cleaner such as a cyclone separator or an electrostatic precipitator could be used to effect removal of ash from the high temperature gases in the line 24, I have found that the most effective and inexpensive removal may be effected by means of a liquid washing or scrubbing device such as a water scrubber, particularly a venturi scrubber. However, before the ash-containing gases can be scrubbed with a water or other wash liquid they must first be cooled sufficiently to avoid excessive volatilization and loss of the scrubbing liquid. Thus, the hot gases from the line 24 pass through a heat exchanger 26 where the gases are cooled to a temperature on the order to 500° F. to 600° F. and are thence introduced through a line 27 to a scrubber 28. Water or other liquid cleaning agent is introduced to the scrubber 28 through a line 29 and the effluent scrubbing liquid is removed through a line 31. Although water is quite effective for removing ash from the gases, the scrubbing liquid may also comprise a chemical reactant or solvent such as monoethanolamine or the like for selectively removing CO_2 from the gases.

Although there are some advantages in operating the hearth zone 16 at superatmospheric pressure, these advantages are outweighed by the practical considerations of simplified hearth construction and ease of access to the hearth. Accordingly, it is preferred that the hearth zone 16 be operated at atmospheric or substantially atmospheric pressure. Under such circumstances, a gas pump or compressor must be provided in the system for effecting passage of gases through the various stages of the process and particularly through the reduction zone 11. In the present instance, the effluent clean reducing gases from the scrubber 28 pass through a line 32 to a compressor 33 and thence through a line 34 to the heat exchanger 26 wherein the cooled and cleaned gases are reheated to a suitable reaction temperature of from about 900° F. to about 1800° F. by indirect heat exchange with the hot gases from the gasifier zone 22. The reheated reducing gases then pass through a line 36 to a pulsating device 37 which may impart a cyclic pulsating pressure to the gas stream for the purpose hereinafter described. From the pulsator 37 the reducing gases pass through the line 12 to the reduction zone 11 as heretofore described.

Although in its broadest scope the invention contemplates any suitable means of contacting subdivided iron ore with the reducing gases in the reduction zone 11, it is preferred to utilize a countercurrent non-fluidized moving bed operation wherein a column of ore solids moves downwardly in countercurrent contact with upwardly flowing reducing gases, the fluid velocity of the upwardly flowing gases being maintained below the threshold velocity for fluidization of the ore particles and preferably not in excess of about 80% of the required velocity for fluidization. To accommodate the preferred non-fluidized moving bed operation, the particle size of the subdivided or granular iron oxide material fed to the reduction zone 11 is preferably within the range of from about 1/2" to about 100 mesh which includes particles substantially smaller than anything suitable for use in the blast furnace. For the foregoing particle size range the fluid velocity of the reducing gases may be from about 130 ft./hr. to about 16,000 ft./min., it being understood of course that the higher gas velocities may

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be used with the larger particle sizes and vice versa. The pressure in the reducing column may be atmospheric or slightly above. Dependent upon the particle size of the iron ore and various other factors including the design of the reduction vessel, difficulties are occasionally encountered with mechanical bridging or blocking which impedes the orderly flow of the ore solids. In order to overcome this difficulty, the pulsator 37 described above is interposed at a suitable point downstream from the compressor 33 thereby imparting to the gas stream a pulsating effect caused by momentary increases and decreases in gas pressure. This scheme effectively avoids mechanical blocking difficulties in the non-fluidized moving bed operation whenever such difficulties are encountered.

Referring now to Fig. 2, a somewhat more detailed assembly of apparatus is shown for carrying out the invention as heretofore described. Thus, the hearth unit in this case comprises a unitary refractory lined enclosure 40 having an upright insulating wall or thermal barrier 41 which divides the enclosure into a melting and gas generation zone 42 at one side of the wall 41 and an upgrading or gasified zone 43 at the other side. A restricted gas passageway 44 is provided therebetween. Reduced iron ore particles are fed to the melting zone 42 through a line 46, and a plurality of coal-oxygen burners 47 are mounted at the outer end wall of the zone 42. The molten iron bath is designated at 48 with the supernatant slag layer being shown at 49, and metal and slag outlets from the hearth are illustrated at 51 and 52, respectively. The high temperature combustion gases (2900°-3500° F.) containing an excess of CO₂ flow through the gas passageway 44 and turbulently intermix with the relatively lower temperature flames and combustion gases from additional coal-oxygen burners 53 mounted in the outer end wall of the gasifier zone 43. The reduction reactions carried out in the gasifier zone 43 are at a significantly lower temperature than the high temperature melting which takes place in the zone 42, and the thermal wall 41 serves to maintain the temperature differential.

The CO-rich reducing gases at a temperature of from about 1900° F. to about 2400° F. pass through a line 54 and thence through the interior of a two-stage heat exchanger 56 and finally through an outlet line 57 to the base of a scrubber 58. Water or other scrubbing liquid is introduced at the top of the scrubber 58 through a line 59 for countercurrent contacting with the reducing gases to remove ash therefrom. The effluent scrubbing liquid is removed from the scrubber 58 through a line 61 for discard, recirculation, or recovery treatment as the case may be. Of course, additional cooling of the gases takes place in the scrubber 58 so that the effluent ash-free reducing gases may be removed from the top of the scrubber at a temperature of from about 100° F. to about 200° F.

The gases from the scrubber 58 pass through a line 62 to a compressor 63 and thence through a line 64 to a gas storage device or holder 66. A by-pass line 67 is provided around the compressor 63. The pressurized reducing gas is fed from the holder 66 through a valve controlled line 68 to a header 69 at the inlet side of the first stage of the heat exchanger 56, and the gases are reheated by passage through the first stage of the heat exchanger to a common header 71 and thence through the second stage of the heat exchanger to an outlet header 72 communicating with the discharge line 73. A by-pass line 74 is also provided around the heat exchanger 56, and a valve-controlled vent line 75 extends from the discharge line 73. The pulsating device is in this instance located in the line 73 and comprises a rotary valve mechanism 74' which is adapted to be opened and partially closed for cyclically restricting and enlarging the flow passage through the line 73 whereby to impart desired pulsations in the gas flow for the rea-

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sons heretofore described. Of course, it will be understood that the pulsator 74' may be operated either continuously or only as required when mechanical flow difficulties are encountered in the reduction zone.

The reducing gas stream from the line 73 is fed into the bottom of a refractory lined reduction column 76 having an upper portion 77 of relatively enlarged diameter and a lower portion 78 of reduced diameter. Subdivided ore particles are fed into the enlarged upper portion 77 of the reduction column through a screw conveyor mechanism 79 from a supply source or ore preheating source (not shown). As will be well understood by those skilled in the art, the ore thus introduced forms a downwardly moving or flowing bed which is supported at its lower end by a cone-shaped partition 81. The partition 81 communicates with one end of another screw conveyor 82 operated by a motor 83 for discharging the reduced ore from the reduction column 76 to the hearth feed line 46. The partition 81 is provided with suitable protected gas passages, such as the bubble cap elements 84, whereby the reducing gas introduced from the line 73 at the bottom of the column 76 may pass upwardly through the partition 81 and thence in countercurrent relation with the downwardly moving bed of ore particles.

The depleted or "spent" reducing gases pass from the top of the reduction column 76 through a line 85 to a cyclone separator 86 or like device wherein entrained fine ore particles are removed. The off gases are discharged through a line 87 and ore fines are collected in a hopper or storage vessel 88 which is connected by line 89 and a valve 91 to the discharge end of the screw conveyor 82 whereby the ore fines may be intermittently charged to the melting zone 42.

The invention as described above relates to an integrated reduction and melting process for making hot metal or molten iron. However, it will be appreciated that the heat exchange, ash removal, and related features of the invention are equally applicable to a simple reduction process wherein the end product is reduced solid iron ore which may be compressed into larger particles without melting and supplied as sponge iron or so-called synthetic scrap. Obviously, in such case only a gas generator zone for producing a CO-rich reducing gas is required in place of the melting-gas generation-gas enrichment arrangement described herein.

Although the invention has been described with reference to certain specific embodiments by way of illustration, it will be understood that various modifications and equivalents may be resorted to without departing from the scope of the invention as defined in the appended claims.

I claim:

1. In an iron ore reduction process wherein a CO-rich reducing gas is produced by means including the combustion of a pulverized solid carbonaceous fuel with an oxygen-rich gas containing at least about 85% oxygen, whereby the resultant reducing gas contains entrained ash particles, and wherein said reducing gas is contacted with subdivided iron oxide ore at a temperature of from about 900° F. to about 1800° F.; the method of preparing the reducing gas for use in the reduction step which comprises passing the ash-containing reducing gas at an elevated temperature through a heat exchange zone and therein cooling the reducing gas, removing entrained ash from the cooled reducing gas, and passing the ash-free cooled reducing gas through said heat exchange zone in indirect heat exchange relation with the elevated temperature ash-containing gas and thereby reheating the ash-free reducing gas to a temperature of from about 900° F. to about 1800° F.

2. The process of claim 1 further characterized in that entrained ash is removed from the cooled reducing gas by scrubbing the latter with a washing liquid.

3. The process of claim 1 further characterized in that

said ash-free cooled reducing gas is passed through said heat exchange zone by means of a compressor.

4. In an iron ore reduction process wherein a CO-rich reducing gas is produced by burning a pulverized solid carbonaceous fuel in a hearth zone at substantially atmospheric pressure with an oxygen-rich gas containing at least about 85% oxygen and controlling the combustion so that the CO₂:CO ratio in the resultant combustion gas is at least about 1 whereby the heat of combustion is sufficient for melting reduced iron and whereby said combustion gas is oxidizing to iron, and wherein the combustion gas from said hearth zone is thereafter passed through a gas treating zone in which the CO content of the gas is increased by reduction of CO₂ with carbon so that the treated gas is CO-rich and is strongly reducing to iron oxide, the effluent CO-rich reducing gas from said treating zone containing entrained ash particles, and wherein the CO-rich reducing gas is contacted with subdivided iron oxide ore in a reducing zone at a temperature of from about 900° F. to about 1800° F., and wherein the resultant reduced iron ore from said reducing zone is passed to said hearth zone in which the reduced iron is melted by the heat of combustion; the method of preparing the CO-rich reducing gas for use in the reduction step which comprises passing the ash-containing reducing gas at an elevated temperature from said treating zone to a heat exchange zone and therein cooling the reducing gas, removing entrained ash from the cooled reducing gas, and pumping the ash-free cooled reducing gas through said heat exchange zone in indirect heat exchange relation with the elevated temperature ash-containing reducing gas from said treating zone and thereby reheating the ash-free reducing gas to a temperature of from about 900° F. to about 1800° F.

5. The process of claim 4 further characterized in that entrained ash is removed from the cooled reducing gas by scrubbing the latter with a washing liquid.

6. In an iron ore reduction process wherein a CO-rich reducing gas is produced by burning pulverized coal in a hearth zone at substantially atmospheric pressure with an oxygen-rich gas containing at least about 85% oxygen and controlling the combustion so that the CO₂:CO ratio in the resultant combustion gas is at least about 1 whereby the heat of combustion is sufficient for melting reduced iron and whereby said combustion gas is oxidizing to iron, and wherein the combustion gas from said hearth zone is thereafter passed to a coal gasifier zone to which pulverized coal and an oxygen-rich gas containing at least 85% oxygen are also introduced so as to burn a portion of the coal introduced to the gasifier zone with the oxygen-

rich gas introduced thereto and so as to react the carbon in another portion of the coal introduced to the gasifier zone with the CO₂ contained in the combustion gas and thereby reduce CO₂ to CO so that the exit gas from the gasifier zone contains not more than about 10% CO₂ with a CO:CO₂ ratio of at least about 7, the effluent CO-rich reducing gas from said gasifier zone containing entrained ash particles, and wherein the CO-rich reducing gas is contacted with subdivided iron oxide ore in a reducing zone at a temperature of from about 900° F. to about 1800° F., and wherein the resultant reduced iron ore from said reducing zone is passed to said hearth zone in which the reduced iron is melted by the heat of combustion; the method of preparing the CO-rich reducing gas for use in the reduction step which comprises passing the ash-containing reducing gas at an elevated temperature from said gasifier zone to a heat exchange zone and therein cooling the reducing gas, scrubbing the cooled reducing gas with a washing liquid and thereby removing ash, and pumping the ash-free cooled reducing gas through said heat exchange zone in indirect heat exchange relation with the elevated temperature ash-containing reducing gas from said gasifier zone and thereby reheating the ash-free reducing gas to a temperature of from about 900° F. to about 1800° F.

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