

Feb. 8, 1938.

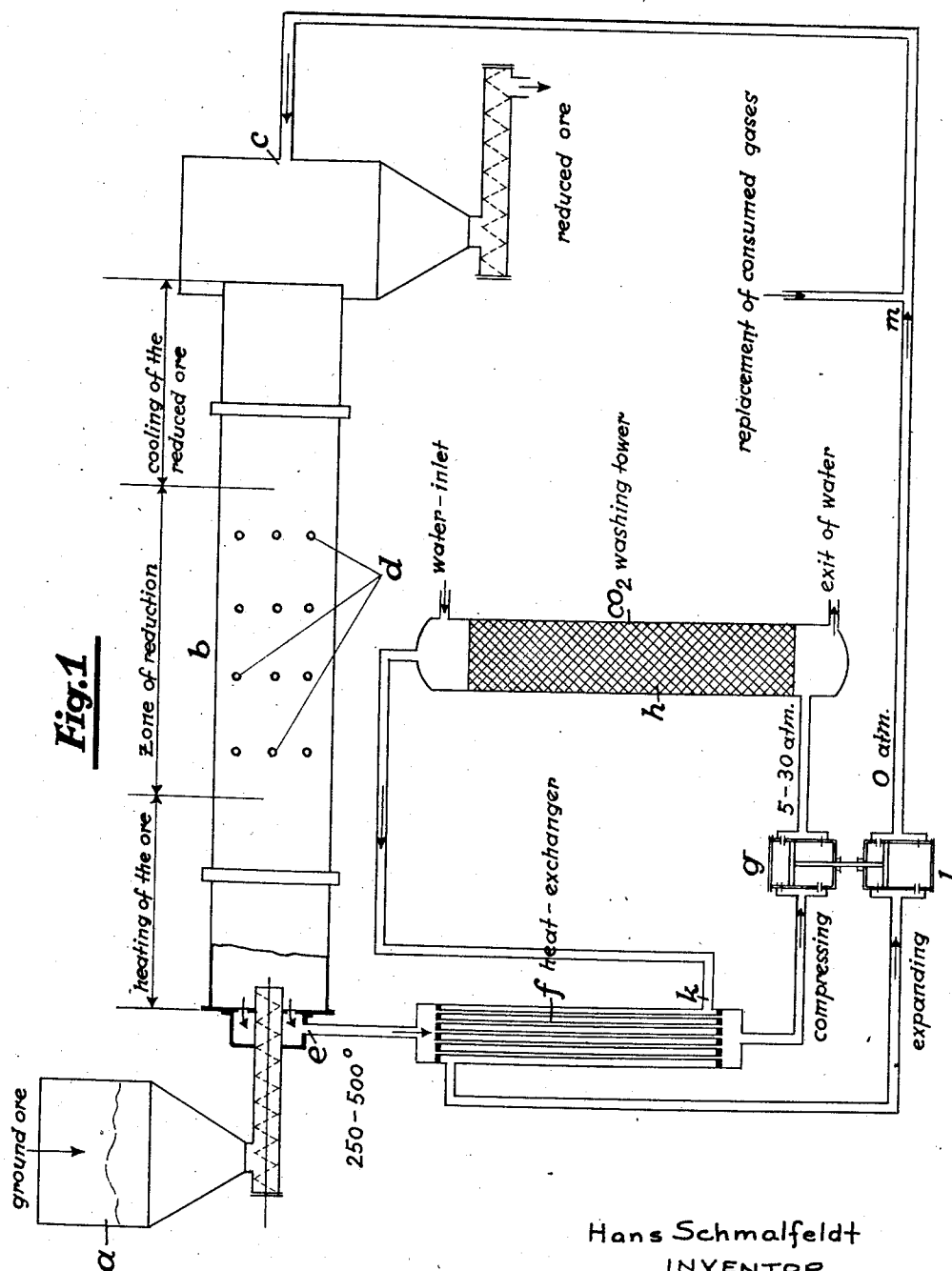
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2,107,549

PROCESS FOR THE REDUCTION OF FINE IRON ORES

Filed Dec. 11, 1934

2 Sheets-Sheet 1



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2 Sheets-Sheet 2

Fig. 2

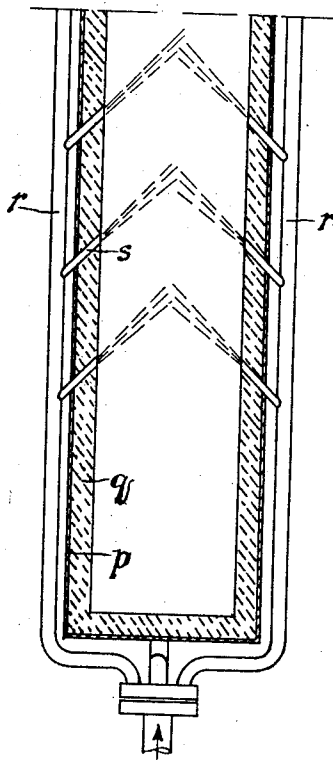


Fig. 5.

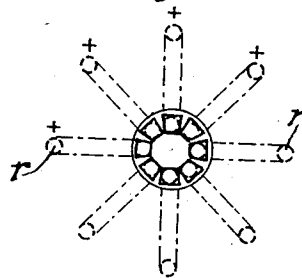


Fig. 6.

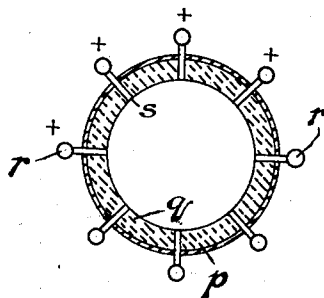


Fig. 3

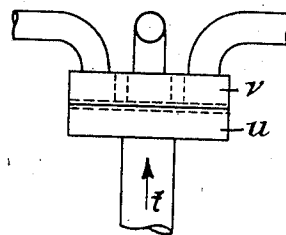


Fig. 4

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UNITED STATES PATENT OFFICE

2,107,549

PROCESS FOR THE REDUCTION OF FINE
IRON ORES

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Application December 11, 1934, Serial No. 756,968
In Germany December 12, 1933

9 Claims. (Cl. 75-35)

This invention relates to a process for the reduction of fine iron ores.

It is well known to reduce iron ores on a commercial scale in blast furnaces. Usually the furnace is charged with lumps of ore and coke, or when fine iron ores are to be reduced by the same process, the ore portion of the charge is usually introduced into the furnace in the form of briquettes. Since the process reduces the iron to a molten condition, it is not suitable for the reduction of fine ores which are to remain in powder form after reduction.

All possible processes and apparatus have been proposed for the purpose of undertaking the reduction of pulverized ores to iron powder. The reduction in a rotating drum through which the reducing gases are passed has been the only one which has proved practical. Here it is suitable to work in countercurrent, so that the fresh gases, which act as a reducing agent, cool the reduced ore and on the other hand, the used escaping gases preheat the freshly supplied ore. The processes which do not work in rotating drums were not successful because, in the shaft furnaces which then come into consideration the ore lies nearly or quite still and thereby sintering or caking takes place. No pulverized iron, therefore, can be obtained, for mechanical reasons alone, if a drum is not used. Added to this, the passage for the gas is blocked by the sintering and the ore does not receive a sufficient supply of gas. Therefore, even in cases where sintered material is required, a drum is also to be preferred.

It has now been found that the heavy ore gives rise to relatively little loss in dust, so that the process can also be carried out in a rotating reduction drum even with very fine ores.

While therefore the reduction of fine ores with gases in a rotating drum is in itself possible, it was however found that there could be no question of its working economically, for the following reasons:

1. The reducing gases can only be utilized to a small extent. During the reduction with e. g. carbon monoxide, only about a third of the carbon monoxide can be converted, while the remaining two thirds remain unattached; the same is true with hydrogen. In order to obtain a sufficient speed of reaction, it is necessary to keep sufficiently remote from the equilibrium composition of the gases which lies within about 63-68% CO. The reaction with carbon monoxide, for instance, takes place in accordance with the equation $\text{FeO} + \text{CO} \rightleftharpoons \text{Fe} + \text{CO}_2$. The reaction is

reversible and in equilibrium if about 60% of the present gases is CO. In the equilibrium state the reaction works neither from the left side to the right or vice versa: the equilibrium is obtained. In order to start a reaction from the left to the right side of the equation which, of course, is desired, it is necessary to use a larger amount of CO, say about 90%. This means that only 90% minus 60% equals 30% CO really reduces, whereas the remaining part is lost as far as its heat value is concerned. The escaping gases which take with them about two thirds of the heat value, cannot be used again without further treatment, as the reaction products carbon dioxide and steam oppose further decomposition.

2. The reduction of iron ores is endothermic, i. e. heat must be supplied for the reduction. It becomes all the more endothermic when this ore is also mixed with finely divided carbon in order to save reducing gas. But it is not only the endothermic reaction of the reduction process itself which absorbs the heat, but there are also additional losses in heat by radiation and conduction in the rotating drum and also by the sensible heat in the gases which are escaping and in the completely reduced ores. The reaction heat can naturally be supplied in small units by heating the drum from outside. As however the reduction temperature lies between 750 and 900° C. external heating already causes considerable difficulty, even on a laboratory scale, as the reduction drum becomes red hot thereby. On a large scale the supply of heat can no longer be undertaken through the drum, not only on account of difficulties of the material but also because the proportion of drum surface to drum content or output becomes increasingly unfavourable, it is therefore compulsory to use internal combustion, i. e. it is therefore necessary to introduce air into the reduction gas during the process. By this means a portion of the reduction gas is burnt, so that the reaction heat and loss of heat are balanced. The direct internal heating is of course bound up with the great disadvantage that the reduction capacity of the reduction gas is diminished as, by the combustion of the air the same reaction end products are formed as those which are formed during the reduction of the iron, namely carbon dioxide and steam.

It has now been found by practical experiments that the weakening of the reduction power of the reducing gases can be so great under some circumstances that a reduction to the required ex-

tent does not take place. This is above all the case when the reduction gases are unsuitable for the reduction. The reduction gases which contain many hydrocarbons are, first and foremost, unsuitable, as the hydrocarbons must split first into carbon monoxide and hydrogen, before the reduction process by means of carbon monoxide and hydrogen starts. Now the splitting of the hydrocarbons is also a strongly endothermic reaction.

The reaction heat for the reaction must naturally also be produced by the burning by means of air in the reducing drum. Hereby carbon dioxide and steam which oppose to the further reduction of iron ore, are again obtained as reaction products.

The idea now occurs to free the reaction gases, only a third of which has been used up, from water vapour and carbon dioxide, after it has passed through the drum and again to circulate it. A short consideration however shows that by the continual internal combustion—even in the most favorable case—so much nitrogen is introduced that the reduction gases are considerably diluted by the nitrogen and thereby rendered valueless. Even if the equilibrium cannot be shifted by the dilution with nitrogen, the speed of reaction will, however, be considerably decreased. On the other hand the speeds of the gas stream in the drum are increased, so that the gas has less time to react with the iron ore. Consequently the output is still further diminished and under some circumstances only a fraction of the output which is obtained when fresh gases are used, can be obtained.

I have found that the previous difficulties can be avoided by the new process and apparatus described in the following specification and illustrated in the accompanying drawings in which—

Fig. 1 is a diagrammatic view of the apparatus for carrying out the process of the present invention;

Fig. 2 is a sectional elevational view of the reduction drum shown in Fig. 1;

Fig. 3 is a transverse sectional view of the drum;

Fig. 4 is an elevational view of the control head;

Fig. 5 is a transverse section of the rotary part of the control head; and

Fig. 6 is a transverse section of the stationary part of the control head.

The process according to the present invention is carried out in the following way:

The reaction heat is not produced by the introduction of air but by the introduction of pure oxygen, so that no dilution with nitrogen takes place. The escaping gases, only a third of which has been used up, are then freed from carbon dioxide and steam and, after being mixed with fresh gases, returned into the drum. The washing out takes place by passing the escaping gases through carbonic acid absorbing liquids, such as solutions of caustic soda, or by means of compressing the escaping gases and washing out the carbonic acid by means of water at low temperatures of 10 to 20° C., preferably under pressures of 5 to 30 atmospheres. According to a further improvement of the invention such reducing gases are used in the reduction drum which contain either no or practically no hydrocarbons (less than 3%), in order to save the heat for cracking the hydrocarbons. The new process is therefore characterized by a combination of the following features:

1. The use of gases practically free from hydrocarbons.

2. Internal combustion in the drum with pure oxygen.

3. Re-use of the residual gases after they have been freed from carbon dioxide and steam.

The process can be advantageously carried out both by the combination of the features 2 and 3 and also by the combination of the features 1 and 2, but the best results are obtained by combining 1, 2, and 3.

The re-use of the residual gases would be impossible when burning with air itself if the above indicated disadvantages of slower reaction speed and small efficiency are taken into consideration, as a considerable portion of the residual gases must always be removed in order to maintain the equilibrium of nitrogen in the drum. Even if it were desired to return the gases, it would only be possible to return a small portion and the residue must be given up as lost. During the process of combustion with pure oxygen, definite quantities of nitrogen are, naturally, introduced with the burning gas. By choosing a suitable combustion gas, this quantity of nitrogen can, however, be considerably reduced. If water-gas is used, for instance, then only about 5% of nitrogen is introduced; with cracked coke furnace gas under some circumstances even less.

The freeing of the residual gases from carbon dioxide is preferably carried out in the following manner:

The cool residual gases are compressed in a small compressor and passed through a water tower in a known manner with a pressure of about 10–15 atmospheres, in which the carbon dioxide is washed out with water. The steam formed has already been condensed during the cooling; the last quantities are condensed with a large quantity of water during the further cooling of the gases by the washing. The gases freed from carbon dioxide and steam are again heated by heat exchange with the hot gases leaving the drum and are then expanded in an expanding machine for the purpose of recovering mechanical work. The compression and also the expansion can be undertaken in piston engines as well as in turbines. Preferably both machines are connected to one shaft. Any suitable power engine desired may provide the energy still necessary. If they are sufficiently preheated by the gases coming from the drum, it is possible so to arrange that the compressed preheated gases give nearly as much energy as is required for the compression of the cold gases. The washing water is forced into the wash tower in a known manner by means of a high pressure pump and after leaving the tower re-expanded by means of a freely radiating turbine or a Francis turbine, for the recovering of energy. Here also it is best to arrange the pump and turbine together with the motor, which supplies the residual energy, on one shaft.

The usual reduction gases come into consideration as reduction gases. Examples for these reduction gases are: distillation gases of mineral coal, gas of coke oven, gas obtained by slowly burning carbonaceous material, natural gas, gas from oil cracking plants, gas obtained by the decomposition of oil; gases free from hydrocarbons: water gas, cracked distillation gases, such as cracked coke oven gas, i. e. gases in which the hydrocarbons have been decomposed.

The oxygen necessary for the internal combustion is produced in the usual manner, for instance, by electrolysis of water or by decomposition of air.

Examples

I. A high per cent iron ore with about 90% Fe_2O_3 , in so far as it is not finely granular, is ground and then placed in the feeding hopper (a) (see Fig. 1) of the reduction drum. The material is fed into the reducing drum (b) by a worm or any other feeding device. The ore is first heated by the escaping gases and then reaches zones of such high temperatures that the reduction begins. After the reduction has been completed, the ore is cooled by the cold reduction gases introduced at (c) and then falls into a delivery casing, from which it is removed by a worm. The reduction temperature is obtained by the introduction of oxygen into the reducing drum, according to a preferred form of construction (see below) by means of a large number of nozzles (d). The supply of oxygen to the drum takes place through a kind of sliding head.

The used reduction gases escape at (e) with temperatures of about 250 to 500° C., according to the quality of the ore and reach a heat exchanger (f). After cooling in the heat exchanger and, if necessary after a final cooling, the gas is sucked by a compressor (g) and compressed to a pressure of about 5 to 30 atmospheres, preferably 12 atmospheres and forced into the carbon dioxide washing tower (h) under this pressure. The gas cooled and freed from carbon dioxide passes through (k) back into the said heat exchanger and reaches, in a warm condition, a pressure reducing machine (l), where the pressure is reduced to atmospheric pressure. The compression and the pressure reducing machines may be both piston machines or also turbines. The gases freed from carbon dioxide and steam arrive back in the drum through (c) after that portion of the gases which has been used up by the reduction and the internal combustion has been replaced through (m).

II. An ore poor in iron is to be reduced with the object later of separating, by a magnetic process, the reduced ore from the gangue. In this case the process is carried out similarly to I but the heat exchanger (f) is omitted as the gases will probably emerge too cold to make heat recovery pay. In this case the emerging gases are therefore immediately washed and cooled, they then reach the compressor, are forced from this into the carbon dioxide washing chamber, are expanded again by means of a pressure reducing machine, so that a part of the compression energy is recovered and then flow back into the drum.

The reduction is carried out at temperatures of 750 to 900° C., preferably at temperatures of 800-830° C.

The rotary drum may be lined with any material generally used for the lining of furnaces, such as stones of silicate or fire brick. There is no need here to pay much attention to the nature of the gangue of the ores as the temperatures in the drum are not high, and there is no need to fear the formation of molten liquids.

It has already been proposed to use gases free from hydrocarbons for the reduction of iron ores. But, on the one hand, these gases were only mentioned as alternatives or by way of example, i. e. it was not recognized that the freedom of the reduction gases from hydrocarbons was an essential condition for successful reduction. Besides, in all these previous proposals the other characteristics of the newer combination are lacking. In some cases the process was carried

out with external heating, in some cases with the use of air. Moreover the special nature of the regeneration proposed according to this invention was always lacking.

It is also already known to use, as reduction gases for the reduction of iron ores, the gases resulting during the obtaining of by-products which are relatively poor in hydrocarbons; such gases, however, always contain 25-40% hydrocarbons.

The advantages of the new process relatively to what was known are the following:

The new process can be carried out simply and practically and works with a minimum cost for gas, as owing to the special kind of the process, the gas used is used up completely. With the new process it is possible to produce iron powder directly. The degree of reduction obtained, amounts to about 85%, i. e. 85% of the total iron present is recovered as metallic iron, the rest as FeO . It is however possible to drive the degree of reduction still higher.

As deposits may be formed, under some circumstances at those places in the drum at which the oxygen is introduced, as over-heatings may easily occur there and the material begin to cake, it is advisable to remove this disadvantage by providing a number of nozzles for the introduction of the oxygen and only to introduce the oxygen by the nozzles in question when the nozzles are in their upper positions, so that the iron ore or iron being blown on directly by the hot oxygen flame does not come into question, but the gas only is heated. The connection is, for example, so chosen that the oxygen is only introduced in that part of the drum which is disposed above the middle plane of the drum. Each nozzle which is fastened to the periphery of the drum and receives its oxygen from outside and blows it into the drum, receives the supply of oxygen during that portion of the revolution which lies above the middle plane. For example, if the revolution is followed from the top point back to the top point again, the oxygen is supplied on the top point. The introduction of the oxygen still continues while the drum rotates through 90°, i. e. until the intersection of the path of the nozzle and the middle plane. Then the addition of oxygen from the nozzle in question is interrupted during the period of a revolution of 180° and only taken up again after the second passage through the middle plane.

This is obtained constructively by connecting the nozzles which lie approximately in the same line of the jacket, by pipes which connect with a control head which is fastened onto the drum. The control head provides for the relevant connection.

The advantage obtained by this form of construction is that a direct heating of the iron is avoided, and further, that the nozzles can cool during the time in which they are not burning, whereby the heating of the nozzles to too high a temperature is avoided.

It is preferable to arrange the nozzles in such a way that the emerging oxygen lies either entirely or partly in an axial direction. (The blowing direction of the nozzles must not be purely radial but must be slightly inclined in order that the blowing direction receives an axial component. The purpose of this device is to give the flames a longer path so that they cannot meet the opposing wall so easily.) This avoids some of the material to be reduced dropping into the nozzles.

Finally according to the invention it is possible to avoid the inconvenient washing out of the carbon dioxide from the residual gases, that is after the reduction. For this purpose the mixture of carbon monoxide and hydrogen, which is to be used for the reduction, is first converted, as regards the carbon monoxide by means of steam at temperatures of 380–550°, preferably about 425°, so that a mixture of hydrogen and carbon dioxide results from the conversion. The carbon dioxide is then removed in any desired manner, for instance, by putting the gas under pressure of 5–30 atmospheres and washing with water in a known manner at temperatures of 10–20° C. or by means of carbonic acid absorbing liquids. The putting under pressure may be also carried out previous to the conversion. After the washing a gas is obtained which consists almost entirely of hydrogen. The reduction is now undertaken with this gas whereby the following advantages are obtained.

About a third to two fifths of the gases are used up during the reduction. The gas mixture which leaves the furnace consists therefore, for example, of about 60% hydrogen and 40% steam. Now it is possible to work up the residual gas in a very simple manner, it is only necessary to cool them, whereby the water vapour is condensed and pure hydrogen remains behind. The gases thus purified, after the oxidized 40% of the hydrogen has been replaced by fresh hydrogen, are then again supplied to the reduction drum.

Naturally, by this process, it is necessary to wash out the same quantity of carbon dioxide as in the process first described. However in the altered process the relations are considerably more favourable. If the process is started with a definite quantity of carbon monoxide–hydrogen mixture, for example one cubic metre, then three cubic metres must be led through the reduction drum if one third of these gases is always used for the reduction. After the first process the carbon dioxide must be washed out from 3 cubic metres of gas, while according to the altered process, it is only necessary to wash out from one cubic metre of gas. Since now the quantities of water required for washing are in the first place proportional to the quantities of gas and the quantities of water to be used are likewise proportional to the mentioned output of gas i. e. the amount of gas passing through the device in one unit of time, say one minute, so the energy required for washing out the carbon dioxide in the first process is about three times as large as it would be with the altered process. This shows clearly the great advantage of this method of carrying out the process.

The reduction drum for carrying out the process, especially the process with the alternate addition of oxygen, consists of an iron drum (*a*, Fig. 2) with a refractory lining (*q*), which is also heat-insulating. On the surface of the drum there are provided several pipes (*r*) which lead the oxygen to the nozzles (*s*). Several nozzles are connected with each pipe on the spots where heat is wanted due to the reduction i. e. where oxygen must be introduced. The number of the nozzles of course depends on the size of the drum, for ordinary sizes a number of 8, 30 or 50 nozzles will come into consideration. The nozzles discharge into the interior of the drum not radially, but wholly or partly in axial direction in such a way, that a somewhat longer path of the flames is obtained. Thereby it is avoided that the flames touch the opposite wall.

By means of a control head (*v*) it is possible to discharge oxygen only if the nozzles lie above the middle plane. Therefore the nozzles indicated in the drawings by + are working, whilst the nozzles not marked are not supplied with oxygen. The regulation of the oxygen is carried out by means of the control-head (*v*). Each of the pipes (*r*) discharges into this control-head. The rotary part of the control-head (cut *x–x*, Fig. 5) is subdivided in a number of chambers—10 corresponding to the number of pipes. Each chamber shows a trapeze-shaped opening in the front flange, through which the connection with the stationary part (*u*) of the control head is formed. Oxygen is introduced at (*t*) into the part (*u*). The chamber (*u*), according to the cut *y–y*, (Fig. 6) does not show any subdivision into cells, but the upper part of the final flange is missing in such a way that the oxygen can enter only into these cells of the rotating part of the control-head which are just on top.

I claim:—

1. Process for the production of iron powder from fine iron ores in a rotary reduction drum comprising the step of bringing into contact the iron ores with reducing gases comprising carbon monoxide, introducing pure oxygen into the drum, the heat for heating the ores to 750° C. to 900° C. being produced by internal combustion of a part of the reducing gases by means of said pure oxygen, the further step of removing the combustion products from the discharged reducing gases by cooling the latter to 10° C. to 20° C. and washing the reducing gases leaving the drum by means of carbon dioxide absorbing liquids, and reconveying the purified gases together with fresh gases to the drum.

2. Process for the production of iron powder from fine iron ores in a rotary reduction drum comprising the step of bringing into contact the iron ores with reducing gases comprising carbon monoxide and being practically free from hydrocarbons, introducing pure oxygen into the drum, the heat for heating the ores to 750° C. to 900° C. being produced by internal combustion of a part of the reducing gases by means of said pure oxygen, the further step of removing the combustion products from the discharged reducing gases by cooling the latter to 10° C. to 20° C. and washing the reducing gases leaving the drum by means of water and reconveying the purified gases together with fresh gases to the drum.

3. Process for the production of iron powder from fine iron ores in a rotary reduction drum comprising the step of bringing into contact the iron ores with reducing gases comprising carbon monoxide and being practically free from hydrocarbons, introducing pure oxygen into the drum, the heat for heating the ores to approximately 815° C. being produced by internal combustion of a part of the reducing gases by means of said pure oxygen, the further step of removing the combustion products from the discharged reducing gases by cooling the latter to approximately 15° C. and washing the reducing gases leaving the drum by means of water under a pressure of 5–30 atm. and reconveying the purified gases together with fresh gases to the drum.

4. Process for the production of iron powder from fine iron ores in a rotary reduction drum comprising the step of bringing into contact the iron ores with reducing gases comprising carbon monoxide and being practically free from hydrocarbons, introducing pure oxygen into the drum, the heat for heating the ores to approximately

815° C. being produced by internal combustion of a part of the reducing gases by means of said pure oxygen, the further step of removing the combustion products from the discharged reducing gases by cooling the latter to approximately 15° C. and washing the reducing gases leaving the drum with water after they have been compressed to a pressure of 5-30 atm. heating the purified gas by the waste gases of the drum, expanding the mentioned purified gases and reconveying them to the drum.

5. Process for the production of iron powder from fine iron ores in a rotary reduction drum comprising the step of bringing into contact the iron ores with reducing gases comprising carbon monoxide, introducing pure oxygen into the drum, the heat for heating the ores to 750° C. to 900° C. being produced by internal combustion of a part of the reducing gases by means of said pure oxygen, which is added at many points alternately in such a way that only an indirect heating of the iron takes place, the further step of removing the combustion products from the discharged reducing gases by cooling the latter to 10° C. to 20° C. and washing the reducing gases leaving the drum by means of carbon dioxide absorbing liquids and reconveying the purified gases together with fresh gases to the drum.

6. Process for the production of iron powder from fine iron ores in a rotary reduction drum comprising the step of bringing into contact the iron ores with reducing gases comprising carbon monoxide and being practically free from hydrocarbons, introducing pure oxygen into the drum, the heat for heating the ores to 750° C. to 900° C. being produced by internal combustion of a part of the reducing gases by means of said pure oxygen, which is added at many points alternately in such a way that only an indirect heating of the iron takes place, the further step of removing the combustion products from the discharged reducing gases by cooling the latter to 10° C. to 20° C. and washing the reducing gases leaving the drum by means of carbon dioxide absorbing liquids and reconveying the purified gases together with fresh gases to the drum.

7. Process for the production of iron powder from fine iron ores in a rotary reduction drum comprising the step of bringing into contact the iron ores with reducing gases comprising carbon monoxide and being practically free from hydrocarbons, introducing pure oxygen into the drum, the heat for heating the ores to approximately

815° C. being produced by internal combustion of a part of the reducing gases by means of said pure oxygen, which is added at many points alternately in such a way that only an indirect heating of the iron takes place, the further step of removing the combustion products from the discharged reducing gases by cooling the latter to approximately 15° C. and washing under a pressure of 5-30 atm. the reducing gases leaving the drum by means of water and reconveying the purified gases together with fresh gases to the drum.

8. Process for the production of iron powder from fine iron ores in a rotary reduction drum comprising the step of bringing into contact the iron ores with reducing gases comprising carbon monoxide and being practically free from hydrocarbons, introducing pure oxygen into the drum, the heat for heating the ores to approximately 815° C. being produced by internal combustion of a part of the reducing gases by means of said pure oxygen, which is added at many points alternately in such a way that only an indirect heating of the iron takes place, the further step of removing the combustion products from the discharged reducing gases by cooling the latter to approximately 15° C. and washing the reducing gases leaving the drum with water after they have been compressed, heating the purified gas by the waste gases of the drum, expanding the mentioned purified gases and reconveying them to the drum.

9. Process for the production of iron powder from fine iron ores in a rotary reduction drum comprising the step of bringing into contact the iron ores with reducing gases comprising carbon monoxide and being practically free from hydrocarbons, introducing pure oxygen into the drum, the heat for heating the ores to approximately 815° C. being produced by internal combustion of a part of the reducing gases by means of said pure oxygen, which is added at many points alternately in such a way that only an indirect heating of the iron takes place, the stream of oxygen being introduced into the gas room in a substantially axial direction, the further step of removing the combustion products from the discharged reducing gases by cooling the latter to approximately 15° C. and washing the reducing gases leaving the drum with water after they have been compressed to 5-30 atm., heating the purified gas by the waste gases of the drum, expanding the mentioned purified gases and reconveying them to the drum.

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