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Inventor(s)

JÄGER; Nils et al.

## METHOD FOR THE DIRECT REDUCTION OF IRON ORE

### Abstract

The invention relates to a process for direct reduction of iron ore to sponge iron, wherein the iron ore passes through a reduction zone for reducing the iron ore to sponge iron. The reduction zone is subdivided into a pre-reduction zone supplied with a first reduction gas and into an end-reduction zone supplied with a second reduction gas. The first reduction gas has a different gas composition compared to the second reduction gas. A first reduction gas has a hydrogen proportion at least 5% by volume higher compared to the second reduction gas.

**Inventors:** JÄGER; Nils (Mülheim, DE), SCHUBERT; Daniel (Duisburg, DE)

**Applicant:** ThyssenKrupp Steel Europe AG (Duisburg, DE)

**Family ID:** 1000008605237

**Assignee:** ThyssenKrupp Steel Europe AG (Duisburg, DE)

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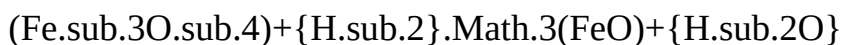
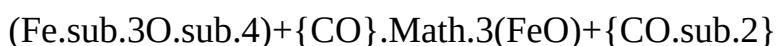
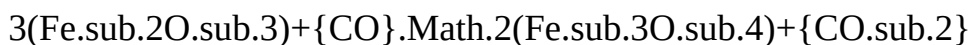
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## Background/Summary

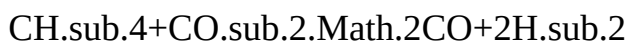
[0001] The invention relates to a process for direct reduction of iron ore to sponge iron.

[0002] The direct reduction process comprises carrying out a solid-state reaction in which oxygen is removed from the iron ore. It typically comprises employing coal and/or natural gas or hydrocarbon-containing compounds and/or compounds of carbon and oxygen as reduction gas. The trend in recent times is that hydrogen is increasingly also being proposed as reduction gas. The reaction is carried out below the melting point of the iron ore so that the outer structure of the ore remains unchanged. Since the removal of oxygen leads to a weight reduction of about  $\frac{1}{4}$  to  $\frac{1}{3}$ , a honeycomb microstructure of the reaction product (solid porous iron with many air-filled interspaces) results. Therefore, the direct reduced iron is often also referred to as sponge iron. The direct reduction process classically employs a shaft furnace as the reactor with a reduction zone through which the iron ore passes counter to the reduction gas. In a special variant of the process the reduction zone is arranged above a cooling zone in the shaft furnace, wherein a cooling gas is passed through the cooling zone. The iron ore then passes through the shaft furnace in the vertical direction from top to bottom. Such shaft furnaces allow good passage of cooling gas and reduction gas through the iron ore on account of the underlying chimney effect. In particular the reduction gas is passed through the reduction zone counter to a direction of motion of the iron ore. Accordingly, the cooling gas likewise flows through the cooling zone counter to a direction of motion of the produced sponge iron. Both the cooling zone and the reduction zone are thus run in countercurrent to achieve efficient reaction between the gases and the solids.

[0003] The reduction gas employed may in particular be CO or H<sub>2</sub> or a mixed gas comprising CO and H<sub>2</sub>. The reduction reactions are as follows (“( )” indicates solids: “{ }” indicates gaseous substances):



[0004] The reduction gas is typically produced from fossil hydrocarbons (e.g. natural gas and/or coal gas). The reactions for methane (the main constituents of natural gas but also biogas) as starting gas are elucidated below by way of example. Other hydrocarbons are likewise possible as starting gas. The reduction gas is produced from methane, CO<sub>2</sub> and steam in a gas reformer (MID-REX® process).



[0005] This results in a gas circuit in which the consumed methane is replaced by new methane mixed with the purified process gas from the shaft furnace upstream of the gas reformer. The process gas from the shaft furnace contains CO<sub>2</sub> and steam as products of the reduction

reaction. The reduction gas H.sub.2 and CO are produced from methane, CO.sub.2 and steam by means of a catalytic reaction in the gas reformer. This reduction gas mixture is supplied to the shaft furnace where it reduces the iron ore according to the above reaction equations. CO.sub.2, steam and sponge iron are formed as the substantial reaction products. CO.sub.2 and steam and unconsumed reduction gas are mixed with methane and returned to the gas reformer.

[0006] The production of sponge iron comprises essentially two fundamental steps. A first step comprises performing the reduction of the iron ore to sponge iron in a reduction zone with a suitable hot reduction gas. A reduction gas typically comprises essentially compounds or mixtures of carbon and hydrogen (for example CH<sub>4</sub>), compounds or mixtures of carbon and oxygen (for example CO) and/or hydrogen (H.sub.2) at temperatures in the range from 700° C. to 1100° C. In a second step the produced sponge iron is cooled to temperatures typically below 100° C. using a cooling gas in a cooling zone. Corresponding processes are known from practice.

[0007] DD 153 701 A5 further discloses a congeneric process which describes introducing different gas streams into the reduction zone of a shaft furnace for iron ore reduction at different planes. The process discloses a simple and cost-effective utilization of a sulfur-containing gas from a gas source in the direct reduction process.

[0008] It is an object of the present invention to develop these processes in such a way that less carbon dioxide is produced.

[0009] This object is achieved by a process for direct reduction of iron ore to sponge iron, wherein the iron ore passes through a reduction zone for reducing the iron ore to sponge iron, wherein the reduction zone is subdivided into a pre-reduction zone supplied with a first reduction gas and into an end-reduction zone supplied with a second reduction gas, wherein the first reduction gas has a different gas composition compared to the second reduction gas, wherein a first reduction gas having a hydrogen proportion at least 5% by volume higher compared to the second reduction gas is used.

[0010] The first reduction gas having a hydrogen proportion at least 5% by volume higher compared to the second reduction gas allows the substantial reduction work to expel oxygen from the iron ore in the pre-reduction zone to be carried out more effectively than in the prior art. The higher hydrogen proportion in the pre-reduction zone, which is simultaneously also the (last) reduction stage before emission of the discharged process gas, makes it possible to simultaneously also provide a reaction opportunity in addition to the reduction work so that the discharged process gas comprises substantially lower proportions of carbon dioxide, thus reducing the emission of carbon dioxide.

[0011] Providing two reduction regions in the reduction zone allows two isobaric regions having low flow rates to be established within the reduction zone, thus making it possible to increase the reaction duration of each of the pre-reduction and the end-reduction and reduce exchange of the respective reduction gases with one another. It is thus advantageously also possible to produce less discharged process gas which, based on volume, can correspondingly be reformed/recycled more economically in order, if required and thus optionally, to be reintroduced into the end-reduction zone of the reduction zone as second reduction gas in the circuit at least in admixture with fresh gas.

[0012] In one embodiment of the process the first reduction gas has a hydrogen proportion of at least 55% by volume. The more hydrogen is introduced into the pre-reduction zone the more effectively the reduction work may be carried out. The first reduction gas has a hydrogen proportion of in particular at least 65% by volume, preferably at least 75% by volume, preferably at least 85% by volume. The further proportions of the first reduction gas may contain at least one compound or mixture of carbon and oxygen and/or steam and unavoidable impurities, for example sulfur compounds and/or nitrogen.

[0013] It is particularly preferable when the first reduction gas consists of hydrogen to make it possible to carry out the greatest possible and optimal reduction work. The use of hydrogen would

result in the carbon content of the pre-reduced iron ore generally being particularly low since no side reaction with carbon-containing compounds which would deposit carbon in the pre-reduced iron ore can occur in the pre-reduction zone, so that a carbon content in the pre-reduced iron ore of less than 0.25% by weight would be expected after the pre-reduction zone.

[0014] In one embodiment of the process the first reduction gas is heated to a temperature between 500 and 1200° C. Before introduction into the pre-reduction zone of the reduction zone the first reduction gas is heated to the required temperature in a gas heater to bring about the pre-reduction of the iron ore. In case of introduction of (essentially 100%) hydrogen it is possible to carry out an introduction especially without further addition of, and thus afterburning with, oxygen, i.e. this ensures complete utilization of the hydrogen for the reduction of the iron ore, thus allowing the process to be operated more economically. Very high hydrogen proportions need not be heated to such high process temperatures since the reduction of the iron ore may be carried out at low temperatures, cf. Baur-Glässner diagram.

[0015] In one embodiment of the process a second reduction gas having a higher proportion of at least one compound or mixture of carbon and hydrogen and/or at least one compound or mixture of carbon and oxygen compared to the first reduction gas is used. The higher proportion of the at least one compound or mixture of carbon and hydrogen and/or at least one compound or mixture of carbon and oxygen in the second reduction gas which is introduced into the end-reduction zone introduces heat into the process through corresponding reaction and effects further reduction and at least partial carburization of the pre-reduced iron ore from the pre-reduction zone. The second reduction gas thus comprises at least one compound or mixture of carbon and hydrogen and/or at least one compound or mixture of carbon and oxygen with a proportion of at least 55% by volume, in particular at least 60% by volume, by preference at least 65% by volume, preferably at least 70% by volume. The further proportions of the second reduction gas may contain oxygen as an oxidant for temperature elevation, hydrogen and/or steam and unavoidable impurities, such as for example sulfur compounds and/or nitrogen.

[0016] It is particularly preferable when the second reduction gas comprises substantially hydrocarbon-containing compounds or mixtures and thus more compounds or mixtures comprising carbon and hydrogen than compounds or mixtures comprising carbon and oxygen.

[0017] The second reduction gas may also comprise a mixture of a make-up gas supplied from a source and a reformed gas produced from the discharged process gas and admixed with the make-up gas.

[0018] The carbon-containing, in particular hydrocarbon-containing, compound or mixture of the second reduction gas can efficiently bring about a carburizing of the pre-reduced iron ore in the end-reduction zone. The carbon from the at least one compound or mixture of carbon and hydrogen and/or carbon and oxygen in the second reduction gas can result in deposition of carbon on the pre-reduced iron ore upon passage through the pre-reduced iron ore in the end-reduction zone. The deposited carbon diffuses into the interior of the iron and then combines with the iron of the pre-reduced iron ore to form cementite. This makes it possible to increase the carbon content of the pre-reduced iron ore. The carbon content of the end-reduced iron ore/sponge iron after passage through the end-reduction zone may be in the range from 0.5% by weight to 3.5% by weight.

[0019] In one embodiment of the process the second reduction gas is heated to a temperature between 700 and 1300° C. Before introduction into the end-reduction zone of the reduction zone the second reduction gas is heated to the required temperature in a gas heater to bring about the end-reduction of the iron ore.

[0020] If hot employment of the sponge iron coming from the reduction zone at a temperature between 500 and 800° C. is not possible, the sponge iron, in one embodiment of the process, passes through a cooling zone. The process thus provides that the iron ore sequentially passes through a reduction zone for reducing the iron ore to sponge iron and a cooling zone for cooling the sponge iron. In the cooling zone a cooling gas is passed through the sponge iron. The cooling gas is used to

cool the sponge iron to a temperature suitable for further transport, for example below 100° C., and can also bring about a (further) “carburizing” of the sponge iron depending on the composition of the cooling gas, especially when carbon-containing compounds are used, preferably carbon dioxide (CO.sub.2), which can preferably be removed from the discharged process gas from the reduction zone and not sent for CCS or CCU for example. In the case of carbon dioxide and hydrogen for example the so-called Bosch reaction takes place in the cooling zone

CO.sub.2+2H.sub.2.fwdarw.C+2H.sub.2O

[0021] Carbon dioxide can be consumed during the “carburizing” of the sponge iron under the conditions prevailing therein. In the cooling zone and as a result of the cooling gas which comprises at least one carbon-containing compound the carbon content of the sponge iron after the cooling and/or after the cooling zone may be adjusted to more than 0.5% by weight, in particular more than 1.0% by weight, preferably more than 2.0% by weight. Furthermore, the carbon content of the sponge iron after the cooling zone may be adjusted to less than 4.5% by weight, in particular less than 4.0% by weight, preferably less than 3.5% by weight, which has the advantage that the sponge iron may be sent to the known further processing processes without any need to adapt the further processing processes. In particular, the sponge iron may be subjected to further processing for example in the Linz-Donawitz converter (also referred to as a “Basic Oxygen Furnace”). In addition, the melting point of the sponge iron may be reduced by increasing the carbon content. This also makes it possible to reduce the energy demand for melting in the electric arc furnace. [0022] Thus in one embodiment of the process the reduction zone comprising the pre-reduction zone and the end-reduction zone may be arranged above the cooling zone in a shaft furnace. The iron ore then passes through the shaft furnace in the vertical direction from top to bottom. Such shaft furnaces allow good passage of first and second reduction gas through the iron ore and subsequently of cooling gas through the end-reduced iron ore/sponge iron on account of the underlying chimney effect. In particular the first and second reduction gas flow through the pre-reduction and end-reduction zone counter to a direction of motion of the iron ore. Accordingly, the cooling gas likewise flows through the cooling zone counter to a direction of motion of the produced sponge iron. Both the reduction zone and the cooling zone are thus run in countercurrent to achieve efficient reaction between the gases and the solids.

[0023] In an alternative variant of the process the reduction zone comprising a pre-reduction zone and an end-reduction zone comprises at least one or more fluidized bed reactors in each case and/or the cooling zone comprises one or more fluidized bed reactors. In a fluidized bed reactor a small-particle-size solids bed is fluidized via the gas which is continuously introduced from below via a gas distributor. This likewise enables efficient reaction between the gases and the solids.

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## Description

[0024] The invention is more particularly elucidated with reference to the following exemplary embodiments in conjunction with the figures. The sole FIG. 1 shows an example of a process according to the invention with reference to a schematic representation of a shaft furnace.

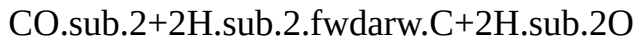
[0025] FIG. 1 elucidates the invention using the example of a shaft furnace (10). Iron ore (io) is introduced at the upper end of the shaft furnace (10). The produced sponge iron (si) is with-drawn at the lower end of the shaft furnace (10). The shaft furnace (10) has a reduction zone (11) comprising a pre-reduction zone (12) and an end-reduction zone (13) and optionally a cooling zone (14) arranged in it. The reduction zone (11) is arranged above the optional cooling zone (14). The cooling zone (14) is not mandatory if hot employment of the hot sponge iron directly exiting the reduction zone (11) is possible and/or the second reduction gas (23) introduced into the end-reduction zone (12) comprises at least one carbon-containing compound or mixture which not only

further reduces the pre-reduced iron ore by reaction in the end-reduction zone (13) of the reduction zone (11) but can also simultaneously achieve sufficient “carburization” to allow supply to the subsequent processes with the required carbon content. The first reduction gas (22) as well as the second reduction gas (23) are passed through the iron ore in the reduction zone (11) in countercurrent and thus counter to a direction of motion of the iron ore. The second reduction gas (23) is passed through a gas heater (33) and heated to a temperature of up to 1300° C. before introduction. The second reduction gas (23) comprises a make-up gas (NG) from a source comprising at least one compound or mixture of carbon and hydrogen and/or at least one compound or mixture of carbon and oxygen, wherein it is preferable to employ natural gas having a very high proportion of hydrocarbon-containing compounds or mixtures, methane (CH<sub>4</sub>). The make-up gas (NG) may be mixed with a reformed gas (RG) which is worked up from the process gas (40) discharged from the reduction zone (11) of the shaft furnace (10). The discharged process gas (40) may be composed of unused reduction gas from any gaseous reaction products. The discharged process gas (40) may comprise hydrogen (H<sub>2</sub>), at least one compound or mixture of carbon and oxygen (CO, CO<sub>2</sub>) and/or at least one hydrogen-containing compound (H<sub>2</sub>O) and unavoidable impurities. The discharged process gas (40) may be supplied to a first process step in which at least one compound or mixture of the process gas and/or at least portions of the unavoidable impurities are separated and/or removed, for example in a unit for process gas purification and dedusting in which at least a portion of the unavoidable impurities are removed from the discharged process gas (40). In a further process step the process gas may be passed through a unit, for example through a condenser, and correspondingly cooled so that the steam (H<sub>2</sub>O) present in the process gas is condensed and thus removed from the process gas. The condensing and discharging of the condensate “dehumidifies” the process gas. A portion of the “dehumidified” process gas or the entirety of the “dehumidified” process gas, shown as a dashed line, may be used as gas (sub)stream a) for firing the gas heater (32, 33). Should insufficient “dehumidified” process gas be available an appropriate fuel gas for firing the gas heater (32, 33) is provided in whole or in part. If a portion of the “dehumidified” process gas or the entirety of the “dehumidified” process gas is not provided for firing the gas heater (32, 33) it is possible to separate carbon dioxide (CO<sub>2</sub>) from the “dehumidified” process gas in a further process step, for example in a scrubber. The separated carbon dioxide may be employed as cooling gas (24) or a portion of the cooling gas (24) in an optional cooling zone (14). However, the process gas freed of carbon dioxide may alternatively also be used, in whole or in part, shown as a dashed line, as gas (sub)stream b) for firing the gas heater (32, 33). Should insufficient gas (sub)stream b) be available an appropriate fuel gas for firing the gas heater (32, 33) is provided in whole or in part. In addition or as an alternative the process gas/reformed gas (RG) freed of carbon dioxide may also be returned to the direct reduction in a further process step by mixing it with the make-up gas (NG), in particular before the mixture is heated to a temperature between 700 and 1300° C. in the gas heater (33). It is possible, optionally and thus indicated by a dashed line, to additionally admix the hot reduction gas with oxygen (O<sub>2</sub>) to increase the reactivity of the second reduction gas (23) in the end-reduction zone (13) of the reduction zone (11) and thus the heat input.

[0026] The first reduction gas (22) introduced into the pre-reduction zone (12) of the reduction zone (11) compared to the second reduction gas (23) has a hydrogen proportion at least 5% by volume higher compared to the second reduction gas (23) and especially has a hydrogen proportion of at least 55% by volume. It is particularly preferable when the first reduction gas (22) consists of hydrogen (H<sub>2</sub>). The first reduction gas (22) may be heated to a temperature between 500 and 1200° C. in a gas heater (32) before introduction into the pre-reduction zone (12).

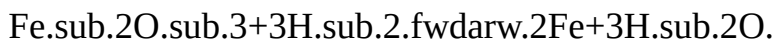
[0027] After exiting the reduction zone (11)/the end-reduction zone (13) the sponge iron enters the optional cooling zone (14). The sponge iron has a temperature in the range from 500 to 800° C. In the cooling zone (14) cooling gas (24) is also passed through the sponge iron counter to the direction of motion of the sponge iron. Unconsumed cooling gas, together with any gaseous

reaction products, is discharged again as process gas (25). A certain proportion of the cooling gas (24) may enter the end-reduction zone (13). A certain proportion of the second reduction gas (23) may likewise enter the cooling zone (14). Mixtures of cooling gas (24) and reduction gas (23) can therefore occur at the transition between the end-reduction zone (13) and the cooling zone (14). The cooling gas (24) especially comprises a carbon-containing compound or mixture, preferably carbon dioxide (CO.sub.2) or methane. Hydrogen (H.sub.2) may, if required, be ad-mixed with the cooling gas (24), as a result of which the cooling gas (24) undergoes the Bosch reaction in the presence of the hot sponge iron as catalyst in the cooling zone (14). Hydrogen (H.sub.2) and carbon dioxide (CO.sub.2) in the cooling gas thus react according to the reaction

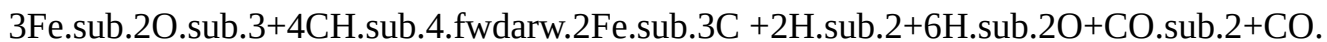


to afford steam (H.sub.2O) and carbon (C), wherein the carbon is deposited on the sponge iron serving as catalyst. The steam with other gaseous reaction products is discharged as process gas (25) from the cooling zone (14) of the shaft furnace (10). The deposited carbon then diffuses into the interior of the sponge iron and forms cementite (Fe.sub.3C). This effect increases the carbon content of the sponge iron (si) to 2.0% by weight to 4.5% by weight. The sponge iron (si) carburized and cooled in this way may be withdrawn in the lower region of the shaft furnace (10) and sent for further processing in the known manner of steel production.

[0028] The particularly preferred process mode for direct reduction of iron ore (io) to sponge iron (si) provides for the use of hydrogen (H.sub.2) as the first reduction gas (22) which after heating to a temperature between 500 and 1200° C. is introduced into the pre-reduction zone (12) of the reduction zone (11) in a shaft furnace (10). When using hydrogen (H.sub.2) as the first reduction gas (22) the reaction of the iron ore to pre-reduced iron ore in the pre-reduction zone (12) is substantially based on



[0029] As the second reduction gas (23) of the particularly preferred process mode provides for natural gas as the make-up gas (NG) which, after heating to operating temperature between 700 and 1300° C., is mixed with oxygen (O.sub.2) if required and introduced into the end-reduction zone (13) of the reduction zone (11) of the shaft furnace (10). When using a make-up gas composed of natural gas (NG) without supply of additional oxygen the reaction is the reaction of the pre-reduced iron ore to sponge iron in the end-reduction zone (13) is substantially based on



[0030] A cooling gas (24) composed of carbon dioxide (CO.sub.2) and hydrogen (H.sub.2) may be introduced into the cooling zone (14) and the sponge iron (si) cooled to a temperature below 100° C.

[0031] As shown in FIG. 1 after its “dehumidification” the process gas (40) discharged from the shaft furnace (10) above the reduction zone (11) is supplied in its entirety as fuel gas/as a portion thereof to the gas heater (33), as indicated by a dashed line, and is not supplied to the make-up gas (NG) and mixed therewith.

[0032] The particularly preferred configuration allows variable optimal adjustment of a direct reduction process with hydrogen (22) and natural gas (23) in variable mixing ratios of 0% to 100% in terms of CO.sub.2 emission, efficiency and the availability of the reduction gases.

[0033] Alternatively and not shown here the invention may also be performed in a cascade of fluidized bed reactors. At least one fluidized bed reactor in each case forms a pre-reduction and end-reduction zone of a reduction zone and depending on the circumstances and if hot employment is not possible at least one further fluidized bed reactor may be used in the cascade as a cooling zone. The iron ore would thus successively pass through the first and the second fluidized bed reactor of the reduction zone and optionally a third fluidized bed reactor of the cooling zone while

undergoing stepwise conversion into sponge iron. If required, the last fluidized bed reactor can effect cooling of the sponge iron using cooling gas. The principle substantially corresponds to that of a shaft furnace but distributed over a plurality of fluidized bed reactors instead of a shaft. The number of fluidized bed reactors can be interconnected as required.

## Claims

1. A process for direct reduction of iron ore to sponge iron, wherein the iron ore passes through a reduction zone for reducing the iron ore to sponge iron, wherein the reduction zone is subdivided into a pre-reduction zone supplied with a first reduction gas and into an end-reduction zone supplied with a second reduction gas, wherein the first reduction gas has a different gas composition compared to the second reduction gas, wherein a first reduction gas having a hydrogen proportion at least 5% by volume higher compared to the second reduction gas is used.
  2. The process as claimed in claim 1, wherein the first reduction gas has a hydrogen proportion of at least 55% by volume.
  3. The process as claimed in claim 2, wherein the first reduction gas consists of hydrogen.
  4. The process as claimed in claim 3, wherein the first reduction gas is heated to a temperature between 500 and 1200° C.
  5. The process as claimed in claim 4, further comprising using a second reduction gas having at least one of (i) a higher proportion of at least one compound or mixture of carbon and hydrogen and (ii) at least one compound or mixture of carbon and oxygen compared to the first reduction gas.
  6. The process as claimed in claim 5, wherein the second reduction gas comprises at least one of (i) at least one compound or mixture of carbon and hydrogen and (ii) at least one compound or mixture of carbon and oxygen in a proportion of at least 55% by volume.
  7. The process as claimed in claim 6, wherein the second reduction gas comprises at least one of (i) at least one compound or mixture of carbon and hydrogen and (ii) at least one compound or mixture of carbon and oxygen in a proportion of at least 70% by volume.
  8. The process as claimed in claim 7, wherein as a result of the carbon-containing compound or mixture the second reduction gas brings about a carburizing of the pre-reduced iron ore in the end-reduction zone.
  9. The process as claimed in claim 8, wherein the carbon content of the sponge iron after passage through the end-reduction zone is in the range from 0.5% by weight to 3.5% by weight.
  10. The process as claimed in claim 9, wherein the second reduction gas is heated to a temperature between 700 and 1300° C.
  11. The process as claimed in claim 10, wherein the sponge iron passes through a cooling zone arranged downstream of the reduction zone which is supplied with a cooling gas.
  12. The process as claimed in claim 11, wherein the reduction zone above the cooling zone is arranged in a shaft furnace and the iron ore passes through the shaft furnace in a vertical direction.
  13. The process as claimed in claim 11, wherein the reduction zone comprises a pre-reduction zone and an end-reduction zone comprising at least one fluidized bed reactor in each case.
  14. The process of claim 13 wherein the cooling zone comprises one or more fluidized bed reactors.
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