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Hydrogen Plasma Smelting Reduction: An Option for Steelmaking In The Future

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Abstract: The steel industry is a major source of global-CO2 Emission. Larger reductions of green house gases are the challenge to develop new processes, like Hydrogen Plasma Smelting Reduction (HPSR). A technology assessment shows the potential for a cheaper and most environment friendly steel-making in the future, when extensive development work has been done.

Carbothermic reduction of oxide minerals is one of the major routes to obtain the corresponding metals. This process produces a lot of CO2, which is responsible for greenhouse effect. Alternatively, hydrogen plasma containing hydrogen in atomic, ionic, and excited states can reduce almost every metal oxide even at lower temperatures. Besides this advantage, plasma processing also offers kinetic advantages. Further, hydrogen-water cycle does not pose any environmental problems. However, reduction of metal oxides in hydrogen plasma is not so straightforward—there are issues relating to introduction of material into the plasma zone, residence time, reverse reaction, and scale-up that must be resolved—yet, it holds the key to future environmental challenges particularly with respect to CO2 emission. This presentation provides an overview of reduction of oxide minerals by hydrogen plasma. The influences of various reaction conditions particularly with respect to reduction of oxides are discussed and some aspects of both thermal and non-thermal cold plasma linking oxidative as well as dissociative reduction are presented.

Keywords: Hydrogen plasma smelting reduction, C02 emission, direct steel making, hydrogen plasma, iron ore.

I. INTRODUCTION

Climate change has become a dominating problem around the globe, and as a result has been a priority in the agendas of many industrial countries in the last decade. This global problem requires a global solution and policies to encourage improved energy efficiency and reduced CO2 emissions in all fields. In that sense, the Kyoto protocol proved its potential due to the commitment of the participating countries to reduce CO2 and five other greenhouse gases, or engage in emissions trading if they maintain or increase emissions of these gases. However, such policies of imposing extra taxes and charges on the Kyoto-countries merely results in a switch of production to other parts of the world. This is likely to increase, rather than reduce, global greenhouse gas concentrations. A competitive global industry, such as the steel industry requires new and imaginative approaches for the future aspects.

Most CO2 gases generated by the steel industry is produced from the reduction of iron ore by carbon in the blast furnace, i.e. 75 % of the total CO2 emissions of the blast furnace-oxygen converter route (BF-BOF). However, it should be mentioned that the maturity and efficiency of the BF-BOF, as a conventional technology, means that in the most advanced facilities, the iron-reduction process operates close to thermodynamic limits. Therefore, by using conventional technologies, making substantial further reductions in CO2 emissions will be almost impossible. Awareness of the need of breakthrough technologies for CO2 mitigation recently arose on an international level.

II. OBJECTIVES

Research and development for new technology solutions to radically reduce the level of CO2 emissions into the atmosphere for each ton of steel produced has been recently undertaken by an EU project called ULCOS. ULCOS (Ultra Low CO2 Steelmaking) is a "concept development" program that explored, fairly extensively, a wide array of steelmaking routes with a high potential for CO2 mitigation. The technologies called for breakthrough innovation within the steel industry, as they will need extensive research & development to mature into commercial processes. The target is to develop new solutions, before the end of the 2010's, with 50 % less specific CO2 emissions compared to the modern blast furnace. These solutions should be in a position to be rolled out into production plants 15 to 20 years from now.

Many technologies have passed a first pre-screening. The technologies were mainly classified to the following sub-projects: (1) new carbon-based reduction concepts, making use of the shaft furnace but also (2) new smelting reduction processes with less common reactors; (3) natural gas based pre-reduction reactors (4) hydrogen-based reduction using hydrogen from CO2 lean technologies; (5)



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direct production of steel by electrolysis, and (6) the use of biomass, which circulates carbon rapidly in the atmosphere. (7) CO2 capture and storage will be included in the design. The priorities of the new processes were innovation in design, optimization of energy and the use of carbonlean or renewable resources.

A smelting reduction process is generally a two stage process, followed by a refining stage to produce liquid steel. These two stages include a first pre-reduction/pre-heating stage, followed by a final stage, which takes place in a bath of liquid iron and slag, figure 1. In the case of carbon based processes, oxygen is also injected with the reductant. This second stage is the so called bath smelter, electric arc or hydrogen plasma reactor. Using fines ore, rather than pellets, lumps and sinter is a main advantage of the process, where raw materials pre-treatment and coking (in the case of carbon usage as a reductant) are ruled out.

III. BASICS

All iron-based materials are originally produced from iron ore. The manufacturing of steel from scrap, a recycling process, will therefore not be considered and compared here. The Fe-O and Fe3-C phase diagrams are the basis of iron and steel making. In Fig-1, the areas corresponding to the raw materials, semi-finished products and finished products are indicated on these equilibrium diagrams, along with schematic manufacturing routes.

The longest and most time-consuming route is the production of liquid steel blast via blast furnace and BOF, which is a 4-step process in total. Although this is the route with the highest CO2 and other emissions, it is the route optimised for energy efficiency. Around 95% of all steel made from ore is produced in this way. The COREX process can be deemed to be a variant of this route



Fig. 1 Manufacturing routes for steel

5% of all steel is made from sponge iron (DRI) via various direct reduction processes and all the electric arc furnace. This is a 3-step route, including pellets, or a 2-step route when ore fines are used directly (FINMET). The main reducing agent and energy carrier are reformed methane and electrical energy respectively. A significant reduction in energy consumption would be possible if DRI could successfully be charged hot and continuously into the EAF.

A third route in future should be the production of liquid iron (LI) in 1-step, continuously from ore fines (sinter feed). The LI could then undergo secondary metallurgy, as in the other routes, to produce the required grades. In our opinion, Hydrogen Plasma Smelting Reduction (HPSR) is the most suitable process. When hydrogen and electrical energy are generated without using fossil fuels, then CO2 emissions are very low.





Fig 2. Ellingham-Richardson Diagram

Fig 3. Baur-Glaessner Diagram at higher temperatures

A. Thermodynamics At Plasma Temperature

The reducing potential of hydrogen is given in comparison to other redox equilibrium reactions in the Ellingham-Richardson diagram (Figure-2.) The enthalpies of the reactions $2H_2 + O_2 \rightarrow 2H_2O$ and $2Fe + O_2 \rightarrow 2FeO$ are approximately equal over the temperature range shown, meaning that when only thermodynamics are considered, molecular hydrogen does not have sufficient potential to reduce liquid iron oxide.

The situation is completely different for atomic and ionized hydrogen. Both of these states of hydrogen can be achieved at very high temperatures, such as those present immediately in the vicinity of an electric or plasma arc. Hydrogen then reduces all other oxide systems. The temperature ranges at which iron oxides are stable in equilibrium with H_2 or H_2O and CO or CO₂ mixtures can be taken from Gaur-Glaessner diagram (Figure-3.) Here, curves are given only upto 1300°C temperature in the figure.



Fig. 4 Transport of active particles to the reacting surface in absence of external electric field.





In plasma metallurgy, hydrogen can be chosen as a reducing gas for ore reduction purposes. Hydrogen under ambient temperature exists only in its molecular form, H2, where the covalent bond between the two atoms is secured by a shared electron pair. At higher temperatures, H2 starts to dissociate, as shown in figure 3, where the amount of H exceeds that of H2 at approximately 3500°C. Increasing the temperature furthermore leads to the ionisation of H into H+. H+ exceeds H at a temperature of 15500°C.

A brief review of the diagram has been described in this work, where H and H+ equilibrium calculations were corrected using FactSage program, with respect to the published calculations done by Dembovsky. FeO-CO equilibrium line was also included. The importance of the Ellingham-Richardson diagram, figure 3, in any set of compounds, such as oxides, is the possibility to readily determine the thermodynamics of all possible reactions. Important reactions and equilibriums can therefore be separated from those which are not important.

Furthermore, the diagram can be used to quantitatively estimate how changes in temperature, pressure and composition affect the relevant chemical equilibrium. The standard free energies of formation of various metal oxides, ΔG° , are represented in figure 4 as a function of temperature.

For temperatures at which the H2/H2O, CO/CO2, C/CO equilibrium lines lie below the oxide lines, the corresponding reducing agent can reduce the oxides to their sub-oxides or elements. Furthermore, the ratios of H2O/H2 and CO2/CO in equilibrium with any of the metal-metal oxide systems are easily determined using the right hand side scales shown in the diagram.

At higher temperatures, up to 2000°C, in the case of plasma application, where the existence of H2, H and H+ is possible, a potential reduction of extremely stable oxides, from the thermodynamical point of view, is obtained. In other words, the use of plasma shifts the ΔG° values further towards the negative domain. This is evident from the shown free energies of formation of H2O by means of H and H+ with respect to H2.

$2H2(g) + O2(g) \leftrightarrow 2H2O(g)$	$\Delta G298 = -474.38 [kJ.mol-1]$
$4H(g) + O2(g) \leftrightarrow 2H2O(g)$	$\Delta G298 = -1287.55 \ [kJ.mol-1]$
$4H(g) + O2(g) + 4e \rightarrow 2H2O(g)$	$\Delta G298 = -6512.24 \text{ [kJ.mol-1]}$

B. Kinetics Of Molten Iron Oxide Reduction

For a chemical reaction to occur, the reacting particles must first collide, i.e. come into physical contact with one another with enough kinetic energy to climb the activation energy barrier. As the temperature of the system increases, the number of molecules that carryenough energy, to react when they collide, also increases. However, mutual contact is not all that is needed. Many reactions are restricted to rates far lower than the limit imposed by the frequency of collisions. By varying the concentrations of some of the reagents, we can not only influence the reaction rate, but also identify the slowest event in the mechanism, that restricts the overall reaction rate.

Mechanisms of the reduction in heterogeneous gas liquid system for iron oxide reduction by hydrogen plasma are governed by the following steps:

- 1) Mass transfer of hydrogen through a gas film from the bulk phase to the reaction
- 2) interface between plasma gas and molten iron oxide or FeO bearing slag.
- 3) Mass transfer of oxygen through a liquid film from molten iron oxide or molten FeO
- 4) bearing slag bulk to the reaction interface.
- 5) Adsorption of the molecular or atomic hydrogen at the reaction interface.
- 6) Adsorption with dissociation of FeO at the reaction interface.
- 7) Chemical reaction at the reaction interface.
- 8) Desorption of H2O from the reaction interface.
- 9) Mass transfer of H2O through a gas film from the reaction interface to the bulk phase.





Fig 6. Schematic diagram of activation energy for reduction of metal oxides with different hydrogen species.

As shown in the figure, reduction of metal oxide to metal by molecular hydrogen may proceed at high temperatures. However, high activation energy actually prevents the reduction from occurring. Hydrogen can be excited to active states (atomic or ionic) and the energy of excited species remains high. The activation energy (E2 and E3) of reactions with active hydrogen species are lower than the one (E1) with hot molecular hydrogen. When the active species are of much higher energy, the activation energy (EA) may be zero or negative. Due to the presence of plasma, the activation energy decreased from 46 to 5.36 kJ/mol probably due to vibrationally excited hydrogen molecules. They also reported that vibrationally excited hydrogen molecules stimulate the chemical processes through their surface dissociation and diffusion of hydrogen atoms into the crystal structure, thereby increasing the rate of reduction. Apart from affecting the gas phase chemistry, the active species also affect the plasma surface interactions. The surface chemical processes (e.g., reduction of metal oxides by hydrogen plasma) are strongly dependant on fluxes and energies of incoming active species to the surface. The vibrational-translational relaxation of the active species is much faster on metal oxide surface than that in gas phase. This relaxation of the active species causes overheating of the surface. This can stimulate the surface oxide reduction processes further while keeping the gas temperature low.

IV. EXPERIMENTS AND RESULTS

The reduction kinetics – the speed at which a reaction moves towards equilibrium and the rate of reaction cannot be gleaned by considering the equilibrium. The kinetics was measured in comprehensive laboratory tests. Initial blow-through and top-blow-tests with pure hydrogen in iron oxide melts did not lead to success. No significant reduction occurred within realistic, practical timeframes and the degree of utilization of hydrogen lay well under the expected 40% and scattered greatly depending on the test conditions. The cause was recognised to be the short stay of hydrogen in the oxide melt and the fact that the reducing potential of hydrogen lies too low for liquid iron oxide. This led to the construction of a plasma smelting reduction furnace on a laboratory scale. Figure below shows the current configuration of the furnace.





Approx. 100g ore fines or iron oxide are melted in an extra-low carbon steel crucible in an Ar atmosphere and then reduced using a hydrogen-argon gas mixture. The energy consumption and the gas flow are measured continuously and the composition of the exhaust gas is continuously monitored using a mass spectrometer. The water-cooled furnace chamber is pressure sealed and the crucible is cooled via its walls. The current and the reducing gas are fed via an adjustable height, hollow electrode made from graphite or tungsten. The electrical circuit is completed at the anode at the bottom of the crucible. This results in plasma being formed. The melt can be observed and the temperature measured optically through a window. The continuous analysis of exhaust gases documents the progression of the reduction and the utilisation of the gas i.e. how much of the hydrogen added to the system is converted to water vapour. When the reduction is finished, the furnace is cooled with argon, the crucible containing the solidified metal is weighed and the metal and slag are analysed. The plasma arc results in a large temperature difference within the melt, as illustrated by the 3-zone model in Figure

The temperature of the melt in the arc attachment zone (AAZ) is over 3000°C, and the atomised and partially ionised plasma gas is hotter still. Oxides and reduced metals are vaporised and condense in colder zones. The temperature gradient at the water-cooled wall and base of the crucible is so large that a solid layer forms at the walls and protects the crucible. On the other hand, the liquid metal around the anode slowly melts the base of the crucible away. The temperature difference and the stirring action of the plasma arc cause a current in the AAZ, where the rate of reaction is very fast. The reduced heavier metal atoms collect at the base of the crucible.



Fig. 9 Gas-oxide surface reaction model



The structure of a molten ore consists of cations, anions and anion-complexes in a particular short-range order, whereby the simple Fe+2 and O2- ions account for approx. 95%. Figure 6 illustrates this schematically for an advanced reduction stage with flow lines and the surface area where the molten oxide comes into contact with the gas. The H2/H2O + O2- \leftrightarrow H2O + 2e- reaction occurs here and especially in the AAZ at a high rate. Water vapour escapes and promotes the reduction. The electrons neutralize Fe2- to Fe, which then falls to the base of the crucible due to gravity. The same procedure occurs for the divalent Mn and Cr cations. Silicon and Aluminium oxide from the acidic gangue in the ore form anionic complexes which agglomerate. This makes them more difficult to reduce than the simple oxygen ions and they are therefore pushed out to the colder outlying areas of the melt and become more inactive. At the end of the reduction the acidic, highly viscose slag can be found mainly at the crucible edge. Fe, Mn and SiO also vapourise in the AAZ.

In the laboratory tests the ore is melted in Ar atmosphere. Weight is lost in the form of water vapour and perhaps CO2 and by the loss of oxygen as Fe2O3 and Fe3O4 are converted to FeOx.



Fig. 10 Loss of weight during heating Fig. 11 Example for test results in Argon atmosphere

As an example, figure-10 shows the weight loss of two ores when heated to a melting point using a thermo balance. Approx 3% by weight is lost as oxygen when Haematite ores (sinter feed) are melted.

Hydrogen reduction begins at this liquid oxide phase in the laboratory tests. Figure-8 shows the progression of the reduction reaction with time, which is similar in principle for all high grade ores. Numerous tests with different ores have shown this to be true. Even the gangue has no significant influence in the usual quantities of up to 5%.

At the start, Fe3+ is primarily reduced. This reaction proceeds at the highest rate and with the highest degree of gas utilization. The Fe2+ reduction phase is characterized by a constant rate of reaction until the melt becomes rich in O2- and the transport in AAZ controls the reaction rate at a degree of reduction of about 90%. In this final phase, manganese, phosphorous and to a lesser degree, silicon is increasingly reduced. The degree of utilization of the gas decreases to about 40 to 50%, that means 50 to 60% H2 in an H2/H20 mixture and agrees with the equilibrium calculations. During the final phase, the degree of reduction is driven to 97 to 99% in order to keep the iron losses below 3%. Towards the end of the reduction the plasma arc forces the molten oxides, whuch are becoming increasingly viscous, outwards and reacts direct with the iron melt. The hydrogen content in the exhaust gas increases sharply. Specific tests showed that the de oxidation of the iron to less than 100 ppm O could be achieved during this phase. The remaining slag has a glass-like structure and is primarily made up of silicon, aluminium and manganese oxides. The filter dusts contain mainly iron oxide, manganese oxide and silicon dioxide as well as lower levels of metallic iron formed by vaporizing and successive partial reoxidation.

H2-Ar mixtures containing 15 to 60% H2 were tested in laboratory experiment. An increase in the hydrogen content of the mixture leads to an increase in the reduction speed, but a decrease in the degree of utilization of the hydrogen. The rate controlling reaction is that at the surface interface between the plasma and the oxide melt.

The liquid iron is a mixture of the reduced iron form oxides and molten crucible material. This is why the tramp element of samples were allowed to solidify in the crucible and later analysed. Figure-11 shows a concentration gradient from the crucible wall towards the centre of the melt. Liquid iron is always cleaner than the crucible material. The C-content, for example, is always lower in the



sample than would be expected from calculations, even though a graphite electrode was used. This means that carbon is being oxidized by the oxide melt or takes part in the reduction process. The CO and CO2 contents of the exhaust gas confirm this.

Sulphur, which can come from the ore, the crucible and/or the graphite electrode, is also mostly removed from the melt via the gas phase. Manganese, phosphorous and silicon are reduced at the end of the process. Depending on the degree of reduction(<99%) and their activities, manganese can be reduced by up to 70% and phosphorous by up to 60% in the sample. Silicon is not significantly reduced but rather vaporizes and can be found as a white SiO precipitate in the furnace and the exhaust gas.

A comparison of Carajas ore and Sishen Ore, which was reduced further, showed higher Si and P contents in the sample where the P content before reduction was about the same range and the end slags were similar. Since the degree of reduction is calculated by analyzing the exhaust gas, the reoxidation od reduced iron can cause the degree of reduction to be low by 1 to 1.5% so that the curve never reaches 100%.

V. HPSR TECHNOLOGY ASSESSEMENT

The mass and energy balance, the way in which the reactions proceed and the metallurgical aspects of HPSR Technology can be adequately described and evaluated based on the results of laboratory tests.

The concept behind an HPSR plant for the continuous production of 75 tons liquid steel per hour is shown in figure 9. This equates to 1.2 million tons per year per plant. Ore fines with a particle diameter of between 1-10 mm are pre-heated in the hot exhaust gas of the furnace and pre-reduced to wustite. This pre-reduction reaction is not of significant importance in comparison to that in the direct reduction processes, since the rate of reduction is much faster and the degree of utilization of the hydrogen gas much higher in the subsequent liquid-state reaction.



Fig. 12 Concept of HPSR-process

The pre-heated fine ore is transported continuously into the HPSR reactor with any small amount of lime or dolomite added to achieve a suitable slag viscosity. Six obliquely positioned hollow electrodes, via which the reducing gas and the direct current are brought into the melt, into a high-energy concentration. A continuously 82 MW electricity supply and H2 gas supply of 100 Nm3/h are required. Hot exhaust gas from the reactor is mixed with cold gas to achieve the temperature required to pre-heat the ore. The gas is subsequently cleaned and dried, and additional H2 and Ar are added to make up the required concentration before the gas is recycled back into the process. The degree of reduction is set at least 97%. This allows the iron losses to be kept under 3% and the reduction of phosphor to be kept under 50%. Slag(max 6t/h) is forced to the wall of the reactor, where it acts as a protective layer, and is tapped off in a controlled manner. The degree of utilisation can be controlled by the holding time.

The liquid iron is continuously tapped off by the siphon and undergoes degassing to remove hydrogen therefore it is deoxidised and alloyed to the required grade.

A full scale version of this concept can only be realised following several years of research work on a pilot plant. The following problems must be addressed:

- continuous pre-heating of the ore fines and transport into the HPSR reactor.

- design of the reactor and control of the high-energy concentration.



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- control of the necessary large volumes of gas.

- refractory lining and cooling of the reactor.

In the assumption that these problems can be suitably solved and that an economic comparison of current and possible manufacturing routes for steel was carried out.

In figure 13, the current costs per tonne of hot strip produced by this process have been assigned a value of 100%. The comparison shows that HPSR is the best choice, with a cost reduction of 21% and the FINMELT is a further development of FINMET process in which the hot DRI is continuously charged into the EAF in a similar way to HPSR. A variation in the costs of different factors of \pm -40% based on the current costs of those factors, was also stimulated in the calculations and the effect of this variation on the individual cost groups and total costs are investigated.

The costs for the 18 most important production factors, including factors such as throughput and credits, were incorporated into a calculated model. The comparison of the two best of 14 production processes is shown with a cost-optimised integrated steel mill with a yearly capacity of 3.2 million tonnes.

An increase in the cost of electrical energy- the largest influencing factor for HPSR and required to produce H2 and for melting – of 40%, results in the production cost of hot strip increasing by 8% per tonne. Even then the costs lie approximately 13% lower than the current optimised costs of an integrated steel works.

The other cost groups are significantly lower due to the one-step process, continuous nature of the process. This is similar for the two-step FINMELT process.



Steel-Making technologies.

Fig. 14 Technology assessment for different processes.

Figure 14 shows a comprehensive evaluation of the potential of each of the three production routes. A decision on a large development and financial investment cannot be made based on production costs alone. In addition to the quantifiable or partly quantifiable values there are a number of factors which can only be judged subjectively, relative to each other: the smaller the value or the smaller the area on each of the six axes, the more advantageous the process.

The "investment price" (axis-1) includes the costs and capital required as well as the revenue from product sales. It indicates the subjective value of a process for each production unit. Since an integrated steelworks is only cost effective as a long process chain of large units, the necessary costs of plant push the "investment price" above that of the simpler and smaller FINMELT and HPSR production lines.

Future possibilities and development potential, relative values, which cannot be calculated but only given subjective, are shown under the heading "vision" on axis-2. The development potential(quality and price of products, energy consumption, flexibility, new processes, infrastructure,etc) of the integrated steelworks, already extensively technically and economically optimised, is low. FINMELT has significant potential for the future in combination with pre-reduction of ore fines. The one-step HPSR process has the highest potential for development on all points, in particular the quality of the new product.

Environmental considerations (axis-3) also indicate HPSR as a first choice, as long as the energy used does not come from fossil fuel sources. CO2, SO2, Nox emissions are reduced to a minimum. Wastewater contamination is unlikely.



Throughput times and flexibility are considered on axis-4. In a well-organized steelworks, it takes approximately 40 hours to get raw materials to hot strip, and the process has associated cooling losses and internal storage. The estimated throughput time for FINMELT is 6 hours and that for HPSR is 3 hours. This results in a high degree of flexibility of customer service and in the quantities produced, and a low level of interim storage for products in progress and consequently lower capital lockup.

The research and development needs and the investment risk are highest for HPSR process (axis-5) and lowest for a conventional steelworks. FINMELT is less risky due to the fact that the direct reduction of ore fines is already proven.

Infrastructure criteria (axis-6) are particularly important when a new steelworks is built. Factors such as the distance to the customer, consistent quality and delivery times, flexibility, simple logistics and the co-operation in development between those who make the steel and those who use it will become increasingly important in the future. An HPSR plant, which is cost effective fulfils these requirements ideally. The smaller production units allow specialised production for particular customers. The flexibility to react to the customer requests in quality and quantity with short transport distances and no need for warehousing is an unbeatable advantage. In comparison to an integrated steel works the fixed costs such as investment, infrastructure and personnel costs are lower due to smaller plant size. In addition to the lower costs required in running the plant at full capacity, lower fixed costs are advantageous if the plant must be run under nominal capacity. Smaller units also benefit production synergy and are easier to manage.

In the evaluation of all criteria, the small area for HPSR shows that it has the greatest potential for the future but also high development costs. FINMELT is limited to use in certain locations due to the need for natural gas. The necessary development work on continuous melting with hot charging of DRI would also be advantageous for the development of HPSR.

VI. CONCLUSION

A new steel making technology based on iron ore with hydrogen as reducing agent and electric energy has been shown. The basic scientific work and technological solutions have been carried out. The results of experiments have led to the development of a concept for HPSR plant on industrial scale with the capacity to continuously produce an iron melt free of carbon and sulphur in a single stage using iron ore fines. A technology assessment has shown that HPSR, if it were available today could result in steel production up to 20% cheaper than by conventional steel making routes but with higher product quality, flexibility and in an absolutely environment friendly way.

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