ASPECTS OF IRONMAKING RESEARCH AT IEHK[1]

Keywords of keynote lecture: Aachen University; Dioxin and Sintering, briquettes and dust injection into shaft furnaces, sticking and plating in DR and SR

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After a short revue of iron and steelmaking history, including the Aachen University and the Institute of Ferrous Metallurgy a comparison of the conventional and alternative steel production routes with four examples are given with the procedure and results of discussions: 1) The dioxin problematic with fundamentals and models for iron ore sintering are introduced, the high technical efforts are discussed. The transfer to scrap and sponge iron melting in EAF are shown. New different filter systems are reported; 2) A shaft furnace was developed to use as charging material carbon containing briquettes with dust and residuals to produce pig iron. The different reduction and melting behaviours of normal pellets in comparison to carbon containing briquettes with models were developed; furthermore the injection of material, which is difficult to briquetize, is added; 3) The knowledge of injection of coals, charcoal, anthracite, gas containing coals in blast-, cupola furnaces and imperial smelters were considered. The injection of fine ore, additives, iron bearing dust, residuals and plastics was researched including the raceway depth and gas composition. The aim was to lower the coke amount and increase the productivity, 4) Problems of DRI Production – swelling and disintegration are well known; to produce prereduced material sticking was researched. The LRI (Low reduced iron) –research is added and the problems of plating; solid reduction agents are discussed.

Aspectos dos estudos na área de redução e fusão no IEHK

Universdade de Aachen: Doxinas e sinterização de minério de ferro; utilização de briquetes e injeção de pós em fornos de cuba; colagem e encrustações na redução direta e na fusão-redução.

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Após uma breve revisão sobre a história da produção do ferro e aço no IEHK da Universidade de Aachen, será feita uma comparação entre as rotas tradicionais e alternativas de produção de aço. Quatro exemplos de pesquisa serão abordados e seus resultados discutidos: 1) A problemática das dioxinas e modelos de sinterização do minério de ferro. Uma abordagem para a rota forno elétrico à arco (EAF) será considerada. 2) O processo oxycup para produção de ferro-gusa, utilizando como carga briquetes auto-redutores contendo pó e resíduos. Uma comparação entre o comportamento de redução e fusão de pelotas e briquetes auto-redutores untilizando modelos. A injeção de materiais difíceis de briquetar será avaliada; 3) Considerando o conhecimento adquirido sobre a injeção de carvões minerais, vegetais, antracitos e gases em alto-forno, forno cubilo e no processo imperial smelters. Além disso com objetivo de diminuir o consumo de coque e aumentar a produtividade, a injeção de finos de minério, aditivos, pós contendo ferro, resíduos e plásticos, foi investigada incluindo aspectos relacionados à profundidade de raceway e composição dos gases. 4) Problemas relacionados à produção do ferro esponja – inchamento, colagem e desintegração será apresentado. Assim como um estudo sobre LRI (baixo grau de metalização) e problemas relacionados à incrustrações.

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Introduction: The first “man made iron” was produced 1500 BC by the Hittites in Anatolia, today a part of Turkey, fig. 1 and 2. It was not invented by the Egypts for their iron-dagger was a forged meteroit. This knowledge went to the West, fig. 3, e.g. to Greece and they thought that only Gods could make such a wonderful metal, fig.4.

Fig. 1 Tuthalija in Hattusa

Fig. 2 Hittitic prisoners in Egypt, 1280 B.C.

Fig. 3 Hittitis and Way of Iron

Fig. 4 Ironmaking by Gods, Vase 600 B.C.

This news arrived via Spain and Portugal e.g. to Brazil and North America.

In Germany still today we can find such furnaces. Only molten slag left the furnace and steel is formed and agglomerated to “Luppe” in the furnace and can be forged, fig. 5. If this iron was molten, it was first unwanted „pig iron“. Later on a lot of such furnaces were built e.g. since 1308 in Dassel, fig. 6.

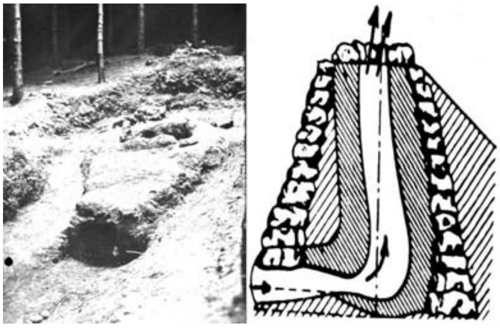


Fig. 5 Sauerland, Germany

Fig. 6 Eisenhütte Dassel

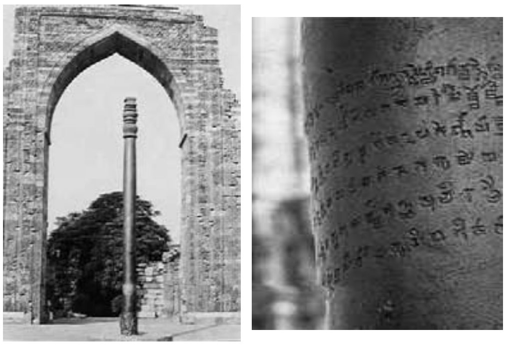


Fig. 7 Indian Pillar, Kutub, 400 A.C.

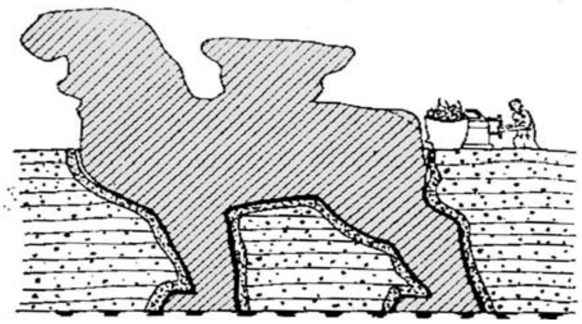


Fig. 8 Cast-iron Lion 954 B.C

These skills of the new metal also went to the East – via India, fig. 7, and on the silk route to China and big animals out of cast iron were produced, fig. 8.

In Aachen in 1865 one of the best equipped steel-works with Thomas-Converter was built up "Rothe Erde" sometimes with more than 5000 workers.

It started by a new situation; no steel should be imported to Germany, but clever merchants found out that the import of cheap pig iron was allowed! Therefore pig iron was transported from many countries: Belgium, England and Luxembourg just behind the border to Aachen and was changed into steel. In Aachen steel-works there was never a blast furnace, no coke-oven and no sinter-strand, fig. 9-10. In 1926 when the company made bankruptcy, the coworkers became unemployed.

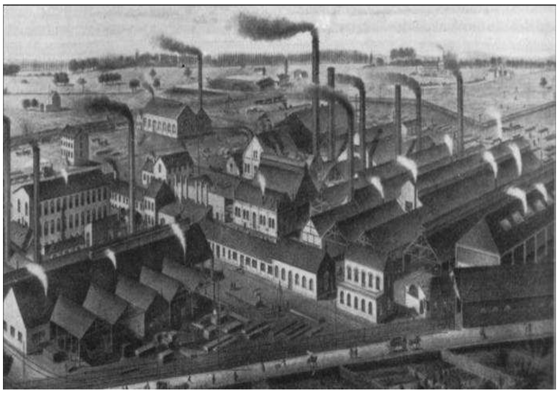


Fig. 9 Steelworks Rothe Erde 1884

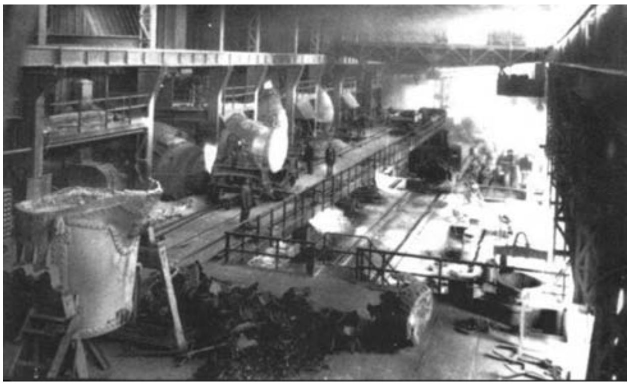


Fig. 10 Thomas-Converter in Rothe Erde

In the prosperous time our University was founded. After chemistry, mining and electro-technics our Institute of Metallurgy started in 1910, fig. 11. The renewed building after the war looks like fig. 12.

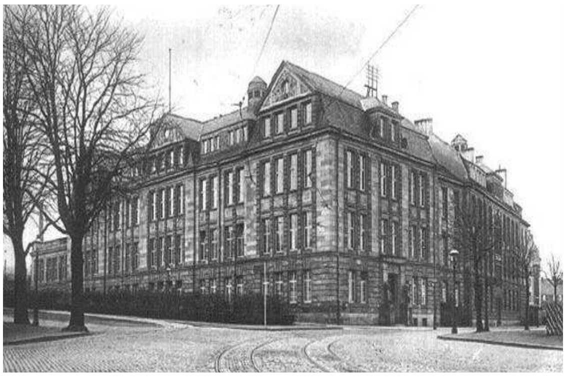


Fig. 11 Institute of Ferrous Metallurgy 1930


Fig. 12 Institute of Ferrous Metallurgy, today

In fig. 13 appears our laboratory. Fig. 14 shows some well known faces of 1983, fig. 15 of 2005 and fig. 16 of 2008 /1.2/.


Fig. 13 Laboratory Hall of IEHK


Fig. 14 Researchers of 1993

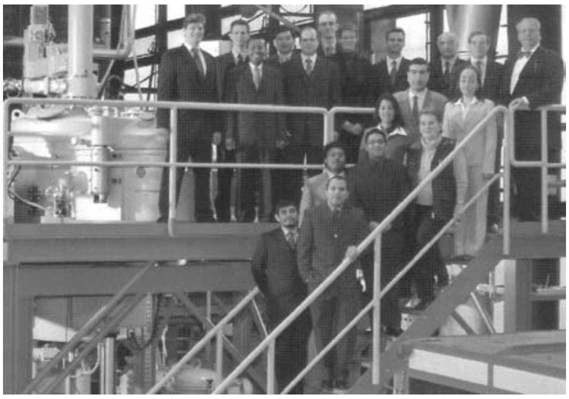


Fig. 15 Team 2005 upon Vacuum Facilities

Fig. 16 Main Building of RWTH Aachen University'08

Fig. 17 gives the four routs of steel production and in the following text results and discussions of our team with the help of many friends are presented.

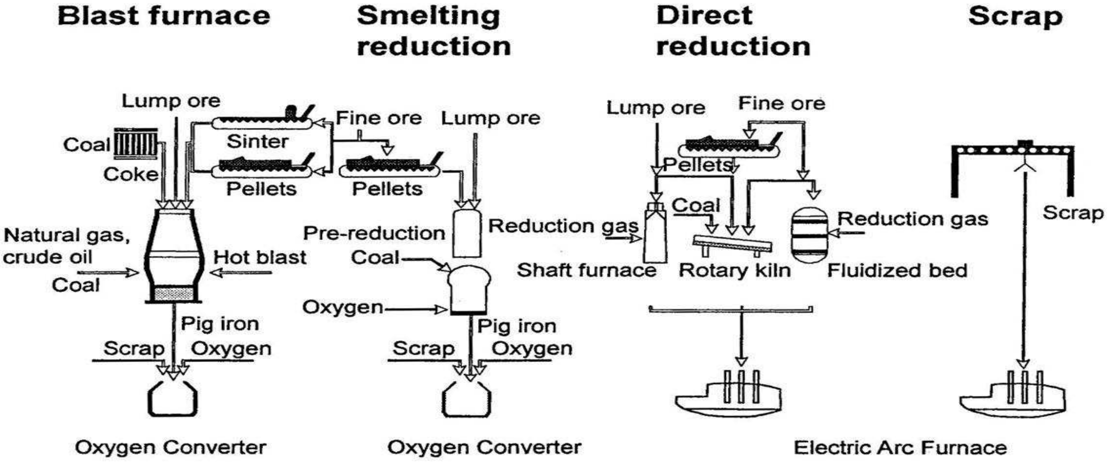


Fig 17 Routes of Steel Production

1. Dioxin problematic: The basic structure of dibenzodioxins and –furans are based on two benzene rings. These can be connected by one oxygen bridge (furans) or two oxygen bridges (dioxins); fig 18. Compounds with different chlorination degrees characterized by the number and the position are congeners (210), with their toxicological properties (TEF) fig. 19; the so called "Seveso- poison" is the most toxical. /6-8/

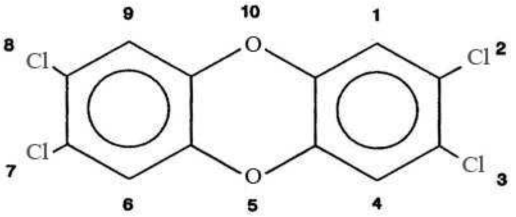


Fig. 18 Structural formula of 2,3,7,8-Tetrachlorodibenzodioxin

Dioxin congeneres	TEF	Furan congeneres	TEF
2,3,7,8-TCDD	1	2,3,7,8-TCDF	0,1
1,2,3,7,8-PeCDD	0,5	1,2,3,7,8-PeCDF	0,05
1,2,3,4,7,8-HxCDD	0,1	2,3,4,7,8-PeCDF	0,5
1,2,3,6,7,8-HxCDD	0,1	1,2,3,4,7,8-HxCDF	0,1
1,2,3,7,8,9-HxCDD	0,1	1,2,3,6,7,8-HxCDF	0,1
1,2,3,4,6,7,8-HpCDD	0,01	1,2,3,7,8,9-HxCDF	0,1
OCDD	0,001	2,3,4,6,7,8-HxCDD	0,1
		1,2,3,4,6,7,8-HpCDF	0,01
		OCDF	0,001
Total: 75		Total: 135	

Fig. 19 Congeners

The standard dioxin formation takes place at 400 to 800°C. Above 800°C the pyrolysis (thermal decomposition) and the reaction with oxygen start. At the cool down process dioxins can be formed by the de novo reaction, fig 20.

The sintering process is described as a non-stationary bed-reactor being flown through by the parallel-flow-process. According to fig. 21 "sinter peak" the lower layer of the sinter bed (G) is heated to 100°C by the hot process gas leaving the layers. This zone is followed by the drying zone (F). After the drying of the sinter mix dehydration (E) is taking place from 300 to 800℃, the expulsion of carbon dioxide and reduction follows (D). From 900℃ starts the ignition of the fuel. The burning of the fuel heats up the sinter bed to 1250-1350℃. The range between the ignition of the fuel and the maximum temperature is called flame- and sintering zone (C). The maximum temperature depends on the composition of the sinter mix and the content of coke breeze. After the burning- and sintering zone the sinter is reoxydated (B) and cooled by the gas stream (A). /9, 10/

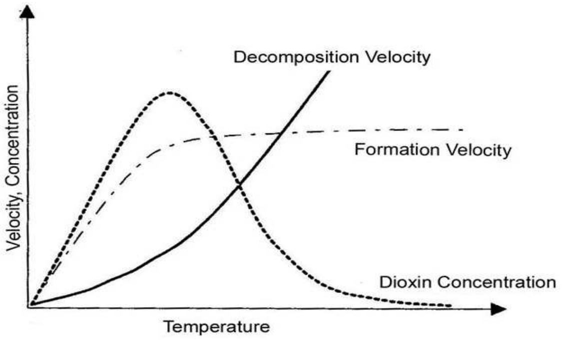


Fig. 20 Dioxin concentration

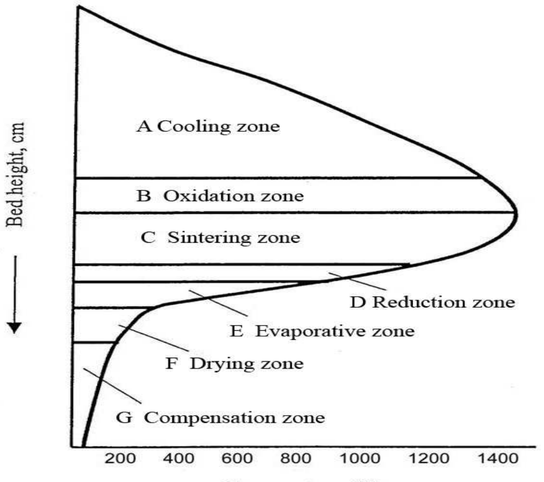


Fig. 21 Thermal & chem. processes in the sinter bed

The figure 22 shows that SO_2 has a maximum of SO_x at the middle of the sintermaschine length; it is explained that the SO_x of the drying and burning zone is accumulated there and then decreases /9/.

In fig. 23 a continuous increase of dioxins from windbox 3 to 11 is shown and the main emission is measured in windbox 11, /7, 8/.

In Fig. 24 a model is presented, the sinter-peak-curves of the sinter strand are given at four different points of time: The **Sinterpeak I** is starting at the ignition point; neither Zone A nor Zone B exists. Production of PCCD/F is only possible below the ignition point. In Zone D it is possible that the temperature-range of 800-1100℃ is undershot because of the low initial temperatures and therewith the production. Therefore only minor PCDD/F emission is expected.

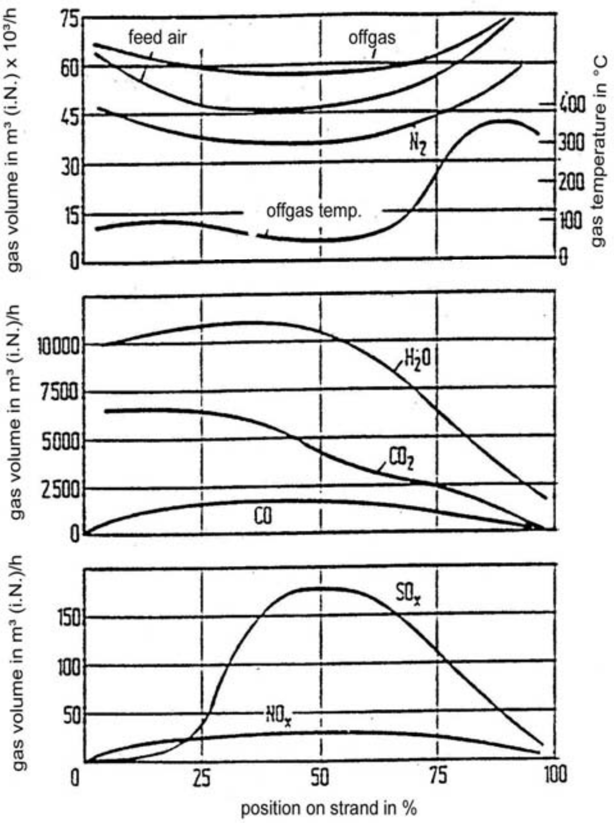


Fig. 22 Temperature and gas composition

Sinterpeak II is in the middle of the sinter process. As a result dioxin production is possible in Zone A; however pyrolysis starts in Zone B, which is continued in the next sinter zone. The main production zone for PCDD/F is similar to Sinterpeak I in the zone E. Only the chance of absorption is lower, so slightly higher emissions are expected. In the third selected area the sinter process is mostly finished. The compensation zone (Zone G), the

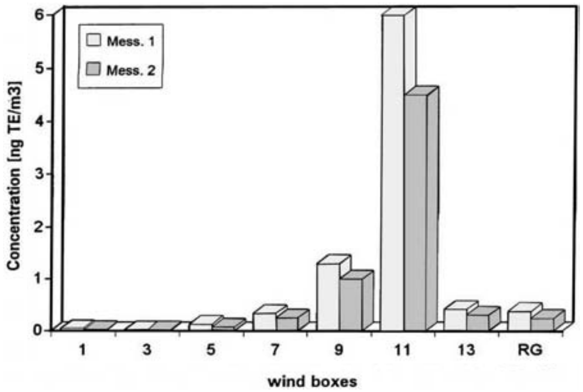


Fig. 23 PCDD/F- concentration over the length of a sinter strand

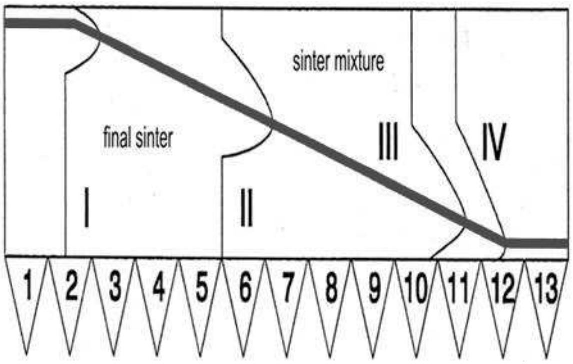


Fig. 24 Sinter-peak-curves of the sinter strand

drying zone (Zone F) as well as parts of Zone E discontinues. The dioxin synthesized in Zone E gets into the waste gas.

Sinterpeak III: In a 3-7 cm thick layer ahead of the fire front an enrichment of evaporation products occurs. Together with the simultaneous enriched PCDD/F, they are released there. Moreover the in Zone E absorbed PCDD/F is driven out; there is a desorption because of the higher temperatures.

In **Sinterpeak IV** the temperature in the sinter bed is characterized by achieving the maximum temperature of the waste gas and the end of the sinter process. The Zone C expires in this zone whereby the possibility of destroying the dioxins produced in Zone A is significantly reduced. The consequence is that in Sinterpeak IV a higher dioxin load occurs. Additional PCDD/F production is benefited by the higher gas-temperature, which is with 350℃ very attractive for the production.

After passing the **Sinterpeak IV** the sinter only cooled down. The dioxins produced in Zone A are carried out by the waste gas. The reaction potential is minimized compared with Zone E, because the larger amount of free carbon and the lower waste gas temperature. Therefore a significantly reduced PCDD/F emission in the sinter cooling zone can be expected.

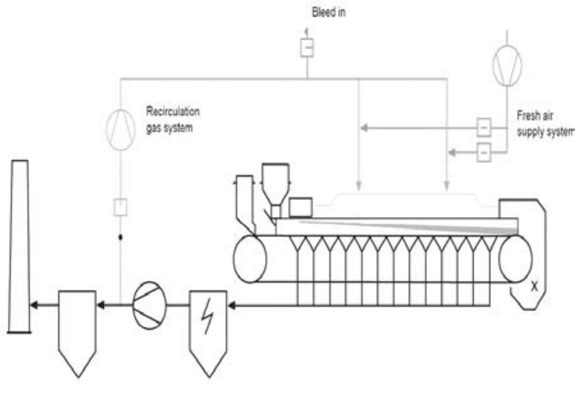


Fig. 25 Optimized Sinter Proces (EOS)

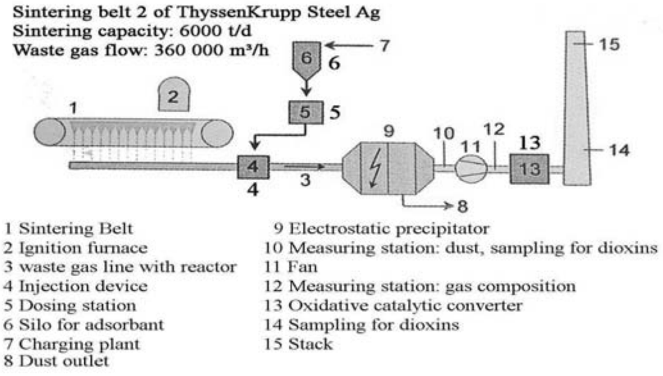


Fig. 26 Waste gas cleaning with adsorption stage and catalytic converter

The EOS takes advantage of the fact that only a part of the oxygen in the air is consumed for coke combustion; a part stream of the offgas is recycled via the hood, enriched with oxygen. This reduces the off-gas volume by about 40–50% and also brings 10% reduction of the coke breeze. 75% of the Dioxins could be avoided /11/, see fig. 25.

The Low-Emission-and-Energy-optimized-Sintering-Preocess (LEEP) was developed at HKM. Furthermore Cu was tested under sinter strand conditions /12/.

Tests were described and discussed to bring down the dioxin emission of sinter strands < 0,1ngTEQ/m². A demonstration was shown at the sinter strand at TKS fig. 26; the dates of the sinter strand were given and the qualities of the lignite coke breeze /13/.

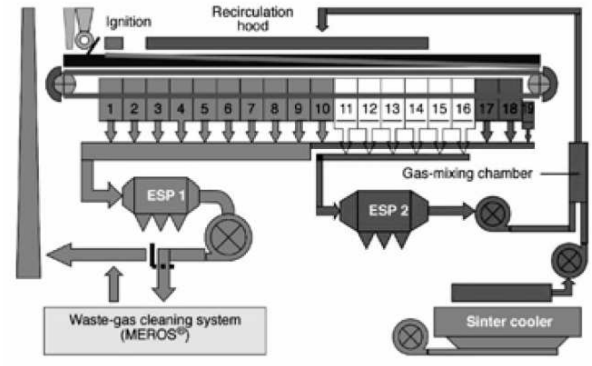
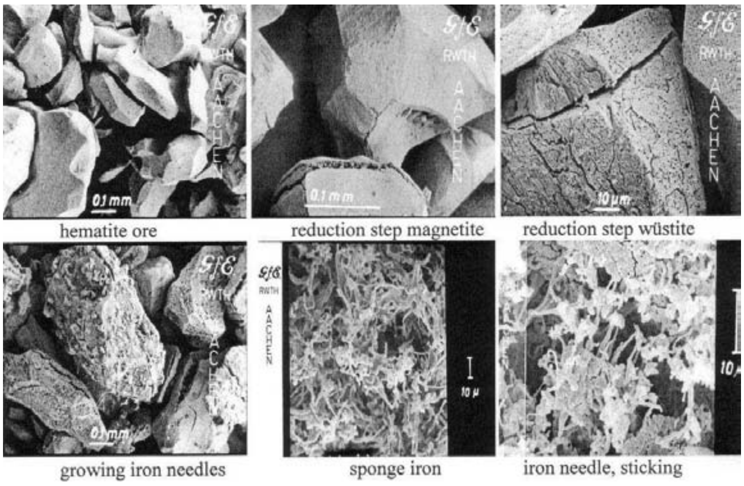


Fig. 27 Eposint waste gas recycling and MEROS

The "Environmentally Process Optimized Sintering" (Eposint) was developed with the knowledge of the Airfine process. In this process the gas is selectively recycled from the windboxes, see fig. 27. Low SO2 and dioxin concentrations were reached. With the addition of MEROS an efficient dry-type gascleaning process was developed /14/.

More different ways were presented, beside fixed bed systems with active coke, an interesting way was presented by Nüsser: a mixture of a fixed and moving bed.

2. Carbon containing briquettes – discussed and explained was the stepwise reduction of Fe2O3 to Fe, see fig. 28, e.g. with the anisotropic reduction speed (hexagonal to kubic cristals) from Fe2O3 to Fe3O4, with crack-formations and increase of volume or from FeO to Fe by formation of iron needles e.g. with catastrophic swelling or to dense iron according to the reduction temperature and gascomposition, fig. 29.



hematite ore
reduction step magnetite
reduction step wüstite
growing iron needles
sponge iron
iron needle, sticking

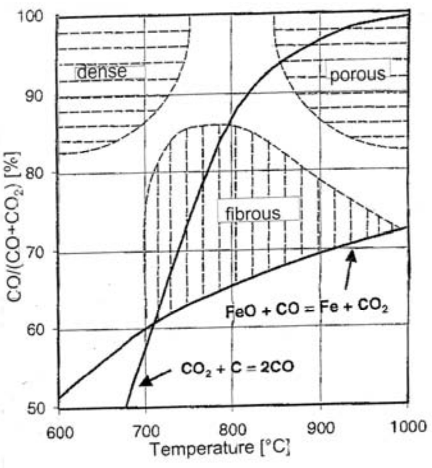


Fig. 28 Iron ore reduction (SEM) /15-18/

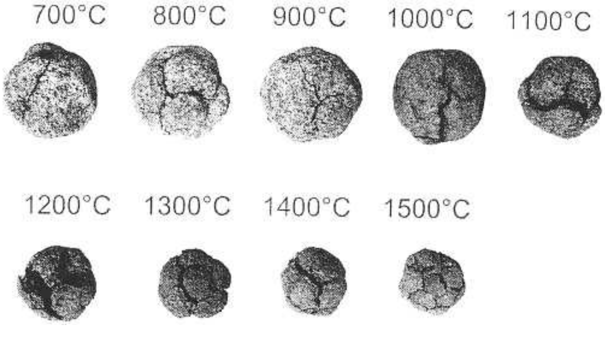
Fig. 29 Areas of iron precipitation /3/

The problems of disintegration, swelling, sticking and accration-formation were studied e.g. in fluidized beds /15, 16/ or rotary kilns /17, 18/ by mixing of different ores or changing the solid reduction medium – lignite or anthracite – or in shaft furnaces for direct-, smelting reduction and in blast furnaces.

This knowledge was taken into consideration to produce and reduce carbon containing agglomerates. Another aim was to use iron containing dusts or slurries of the steel industry, furthermore material e.g. with high Zn-contents should be tested and also e.g. shreder-light-fraction from motorcar-recycling and (DSD), plastic wastes as possible solid reduction material.

Different types of furnaces were proved to use iron containing dust, e.g. for the OxiCup process several research campaigns were started. Iron ore pellets with embedded carbon showed first a small swelling but then an intensive shrinking, fig. 30 and 31 /19, 20/.

By adding anthracite into the green pellets the reduction started at 1000° C.



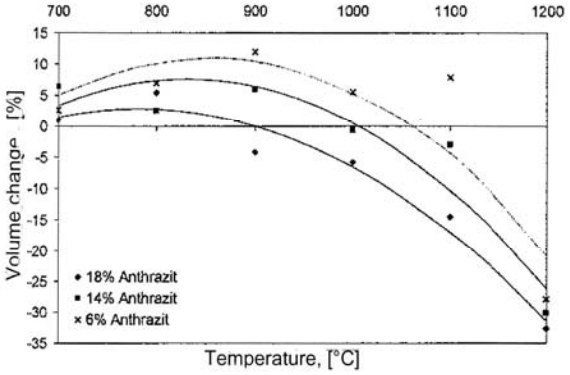
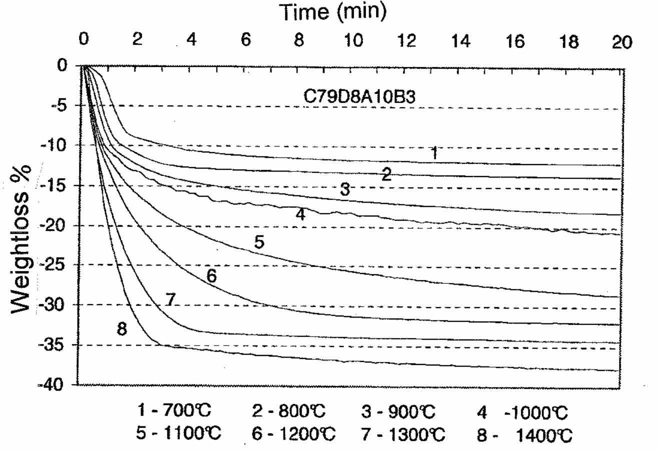


Fig. 30 Shrinking ot pellets with embedded Carbon /19/

Fig. 31 Effect of temperature and anthracite content on the volume change

In fig. 32 the small influence of DSD (plastic) appeared for this material was gasified already before the reduction started /8, 9/. Also briquettes showed a simular behaviour, fig 33 /20, 21/.



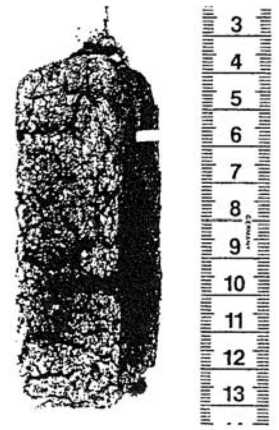


Fig. 32 Weightloss of CVRD-Pellets, 8% DSD+10%C

Fig. 33 Porous briquette after reduction

In blast furnaces „hollow icicles of iron“ were found after quenching /22/. A possible explanation of this phenomena was given by the reduction- and melting behaviour of iron ore pellets in blast furnaces. When the pellets are only reduced by gases first an iron shell is formed on the surface. By increasing the tepmerature the remaining FeO as Fayalit in the innerpellet is melting. At the connection points of the pellets no iron shell is formed; the molten FeO drops from one pellet into the next, fig. 34 /22/.

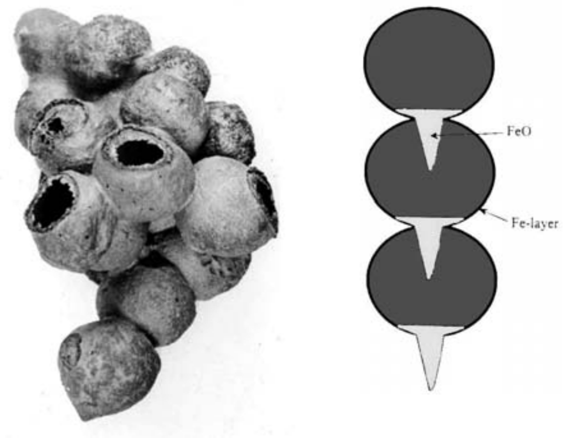


Fig. 34 Hollow Icicles

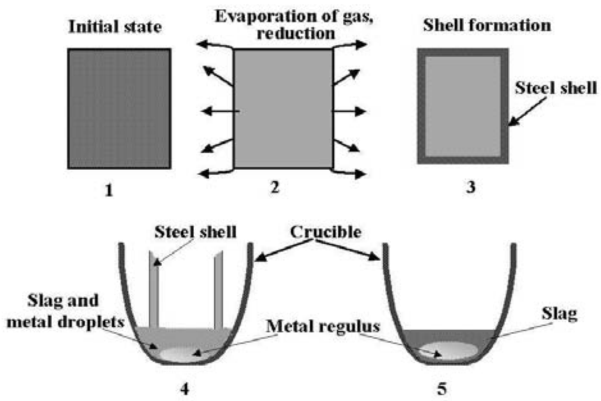


Fig. 35 Reduction and smelting mechanism of briquettes with embedded carbon

This happens with briquettes too, fig. 35, /23/. Parallel to briquettes of iron oxid and carbon /24/ a broadening this subject a research was started to utilize Mn ore and Fe ore-briquettes in a direct - and smelting reduction processes to get a prematerial for the production of high Mn steels /25/.

Fig. 36 Laubinger furnace for smelting reduction

3. Injection into shaft-furnaces - since 1980 literature about injection was evaluated and research projects started to use injected coal instead of coke /26-28/; a horizontal rig was developed and different injection methods compared. Results of lab-tests and blast furnaces were discussed and publications e.g. of research with Brazil were published with different coals /29/ and charcoal /30/. The horizontal rig was changed into a vertical version, see fig. 37. Furthermore a coke-bed-simulator was built up to study the behaviour of dust or particles /31, 32/, see fig. 38.

Fig. 37 Injection rig

Fig. 38 Coke-bed-simulator

Different coal mixtures were tested; fig. 39 shows that a small amount of lignite increases the ignition of the mixture intensively e.g. 20% lignite brings the behaviour of 100% lignite /27/. In further campaigns additives were added; fig. 40 shows the influence of BaCO3 and ZWS /33/.

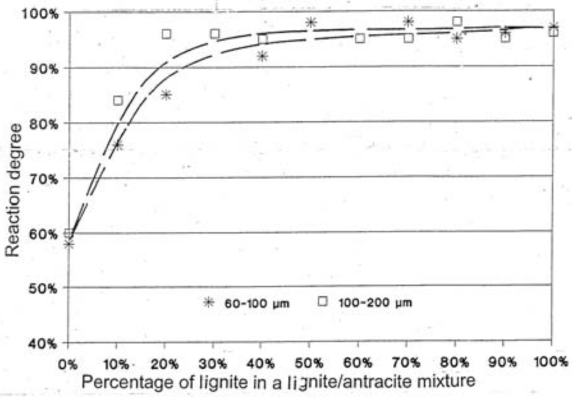


Fig. 39 Burning behaviour of lignite/anthracite mixture

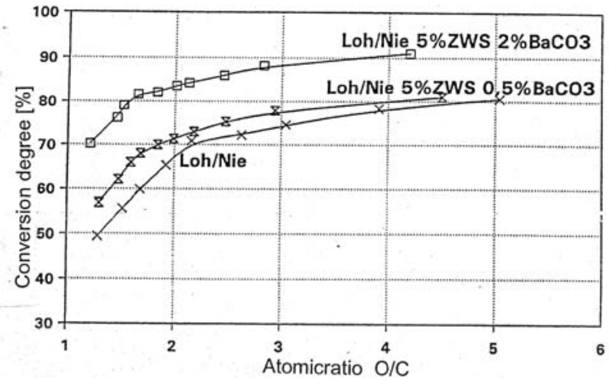


Fig. 40. Coal mixture with BaCO3 addition

As a connection to the last chapter research projects were added to inject e.g. BOF dust into the OxiCup, see fig. 41, /34, 35/. This dust contains minerals which need energy to melt; furthermore the oxidizing/reducing step of the iron part of the dust effects the situation in the injection rig.

Also the behaviour of injected material e.g. iron dust and swarf were tested with additional injection of CH4 and O2 to simulate a campaign for cupola-furnaces /31, 32/ - see fig. 42 – in the coke-bed-simulator. The melt of the injected material was dropping down on the coke pieces.

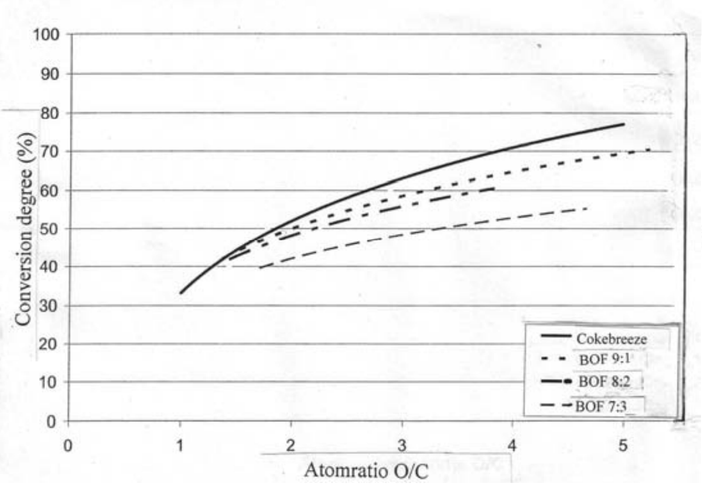


Fig. 41 Conversion behaviour of coke and BOF dust /34/

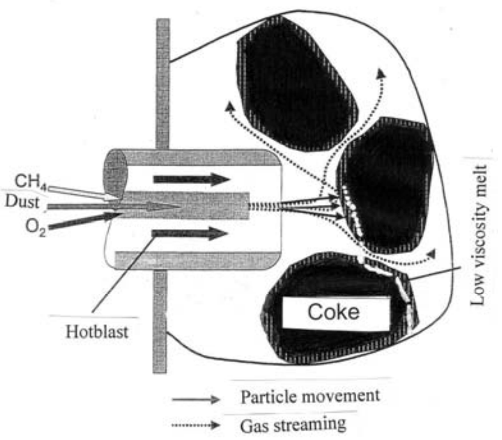


Fig. 42 Injection of Fe-dust /31/

Different models were built up e.g. with different material injections and different ways of recycling of top gas /1/ see fig. 43 and 44.

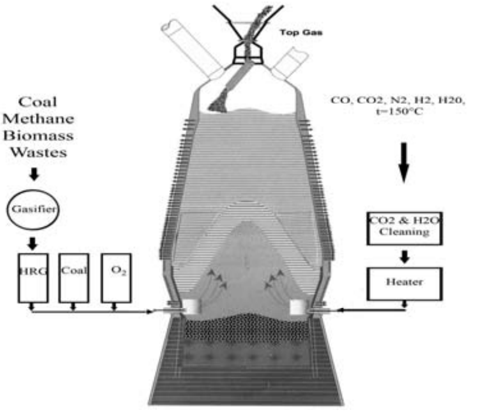


Fig. 43 Hot reduction gas injection

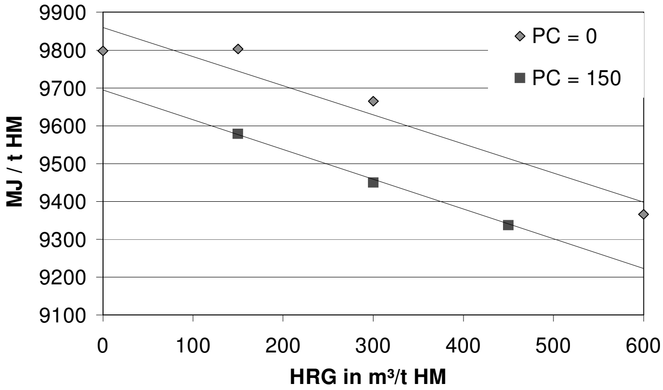


Fig. 44 Total energy loss

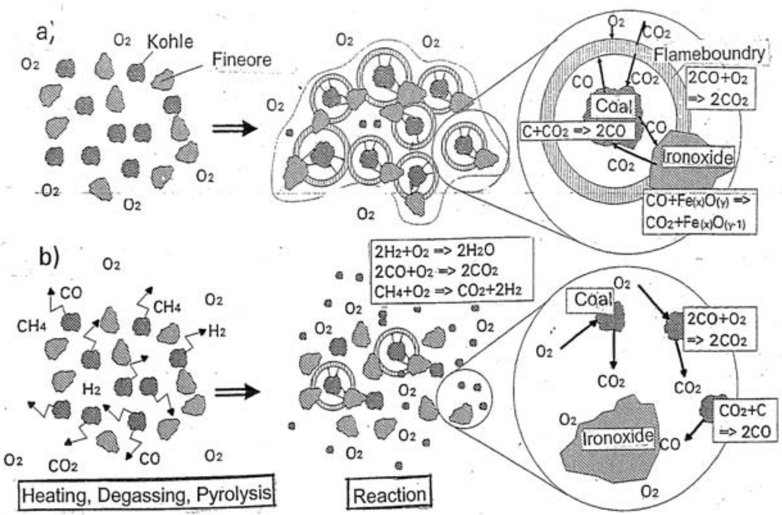


Fig. 45 Mechanism of combine coal/ore injection
a) gas rich coal b) low volatile coke

Very interesting were the results of the research with injecting of Brazilian fine ore /36, 37/. Three possible steps to inject fine ore and coal were tested and a way of taking out probes out of the raceway of a blast furnace was developed /36/. Using the knowledge that gas rich coals can explode by injecting into the raceway – zone and that the gas is immediately transformed into CO_2 a model was built up that explained the better reduction of iron ore in the raceway by injecting low volatile coal. In the flame front such coals form CO and the reduction starts, see fig. 45.

Further research about different lances /38, 39/, the raceway depth and the gas composition /40, 41/ were published; blast furnaces of injecting different materials /42, 43/ into cupola-furnaces and imperial smelters were tested.
Interesting will be the next campaigns in an enlarged coke-bed-simulator to get results from the tip of the injection lance to the dead man.

Plating

The phenomena of "plating" are discussed and evaluated /44/. The problems of accretions are known, e.g. by formation of rings of reduced material at the wall of rotary kilns, which influence the material flow, or in BF which influence the gas streaming and efficiency; furthermore the problem of sticking among the reduced materials forming cluster were published. Plating is one technical problem which occurs by direct reduction of iron ore, e.g. in fluidized bed-reactor /45/. Plating is defined as an accumulation of freshly reduced iron ore on the wall of reduction reactors. Also plating is understood as fouling of heat exchanger tubes. It mainly occurs at elevated temperatures by contact between metallic surfaces (tubes) with iron containing particles in dust carried with the reduction reactor off-gas /46/. Plating leads to disturb the material flow inside the reactors; sometimes it was explained by sticking of iron reduced material at the steel tubes /47/. The problem may occur at sharpedged connections and dead zones in heat exchanger. Plating phenomena differs from metal dusting. Two test apparatuses were applied to clarify the plating problems: the first one was a muffle furnace to simulate the static simulation during reduction, another was a Salvis furnace, fig 46, to get the influence of movement - this rotary furnace was already used in 1986 /48/. Effects on formation of plating layers including for instance chemical composition of iron ore, chemical property of different steel plates were investigated and discussed /44, 49/.

No plating was e.g. noticed by fine Ilmenite ore with steel plates, also an increasing content of SiO_2 in the ores encountered to decrease the plating trend, fig 47. High Chromium steel restrained the formation of plating, fig 48 /49/.

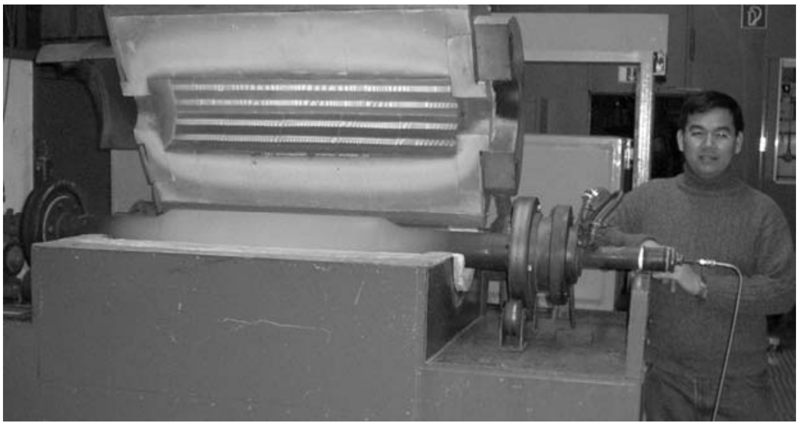


Fig. 46 Experimental rotary kiln furnace (Salvis furnace)

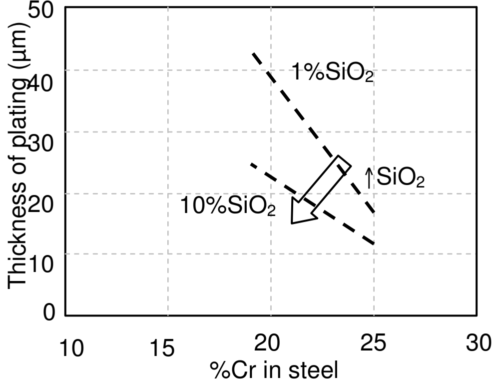


Fig. 47 Effect of chromium content of steel plate and SiO_2 content in iron ore

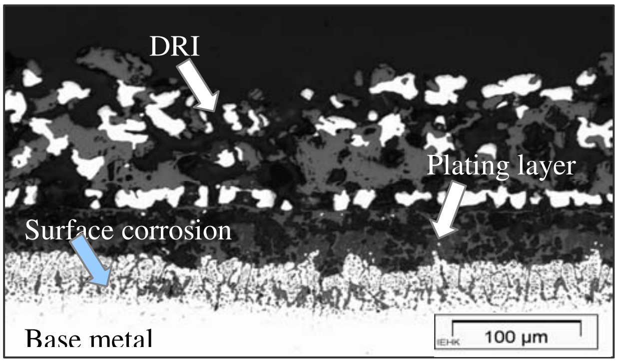


Fig. 48 Plating formation on steel plate

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