

REDUCING AGENTS FROM BROWN COAL FOR THE PRODUCTION OF SPONGE IRON

Prof. Dr. W. WENZEL

Dr. M. MERAIB

Institut für Eisenhüttenkunde,
RWTH Aachen, Abt.

Eisenhüttenmannische Verfahrenstechnik

Direktor Dipl.-Ing. P. SPEICH

Dr. F. H. FRANKE

Rheinische Braunkohlenwerke AG, Köln.

Sponge iron technology in comparison to the conventional procedures.

Conventional procedures.

The production of steel from iron ores takes place in three stages:

- a) separation of the oxygen from iron.
- b) melting of the iron and the gangue and
- c) separation of the accompanying elements with possible additions of alloying components or adjustment of the final analysis.

With the conventional procedure, blast furnace-converter, the first two stages are carried out by the blast furnace. The separation of the accompanying elements from the pig-iron is carried out by the converter.

In the blast-furnace, only those fuels, such as coke, or more recently in the trial stage, hot briquettes, that fulfill the quality requirements in respect of strength and low ash, volatiles, and sulfur content are used.

The replacement of reducing coke by oil, natural gas, coal dust or reducing gas injection is limited by the fact that the coke serves as a support for the burden and ensures an uniform passage for the gases through the burden column.

In the electric furnace scrap melting process, which is a competitor to the blast furnace-converter process, only small changes in the chemical analysis are effected by additions. A rise in the content of such element as copper is regarded as detrimental to the steel quality, which, however, can be compensated for by additions of new iron in the form of pig-iron or sponge-iron.

Direct reduction process

In the so-called direct reduction processes (rotary kiln, fluidized bed, retort, and shaft furnace processes) the separation of the oxygen from the iron takes place in the reduction chamber itself, where the reduction of the iron ores proceeds in the solid phase. The subsequent treatment of the sponge iron produced to steel is carried out mainly by the electric furnace. Another possible application is a cooling scrap in the converter.

The application of direct reduction processes enables iron metallurgy to dispense with the use of costly, high-grade coking coal.

Possible reducing agents and energy carriers in addition to the solid fuels such as bituminous coal and brown coal are given by gaseous fuels (coal gasification, methane reforming) and liquid fuels (oil-conversion).

The direct reduction processes (Krupp sponge-iron-, SL/RN-, and Kawasaki-steel-processes) are suitable for the application of iron rich fine grain ores, concentrates or pellets.

The application of solid reducing agents with increased volatiles content and high reactivity, as in the case of brown coal, is of advantage for reduction in the rotary kiln. The use of brown coal is a well-established technique.

In the gas reduction processes (HIB, Hyl, Armco, Midrex, Purofer) the required reducing gas is, at present, produced by the reforming of natural gas, (the deposits of natural gas at the present rate of consumption, will be probably exhausted by the year 2000). By comparison,

coal in Germany is considered to be available for a long time yet and is, at least as far as brown coal is concerned, stable in price. This means that for the production of reducing gas in Germany brown coal can be utilized for some considerable time.

Summarizing, in the short term, rotary kiln reducing processes may be operated with brown coal; in the medium and long term gas-reduction in the shaft furnace may be effected by gas from brown coal.

The sponge-iron serves as iron charge in the electric furnace whose product is comparable with the liquid steel from the blast furnace converter route.

A comparison of the various process-routes for the production of liquid steel.

A cost analysis of liquid steel production by the blast-furnace-LD-route the electric-furnace-scrap-melting-route, and the sponge-iron-electric-furnace-route has been done.

Table 1 Cost of liquid steel based on location Duisburg, Germany

Alternative routes	blast furnace-converter		scrap-electric furnace		sponge iron-electric furnace	
	DM/t	%	DM/t	%	DM/t	%
material costs	111,33	42,5	184,64 *)	78,0	125,92 *)	49,0
fuel costs	71,58	27,3	-	-	39,76	15,5
energy costs	11,79	4,5	19,96	8,4	25,58	9,9
labour costs	8,50	3,2	4,57	1,9	9,61	3,7
imponderables	8,22	3,2	4,18	1,8	6,94	2,7
investment-costs	50,83	19,3	23,37	9,9	49,38	19,2
total costs	262,05	100 %	236,72	100 %	257,19	100 %

*) cost of electrodes included in material costs

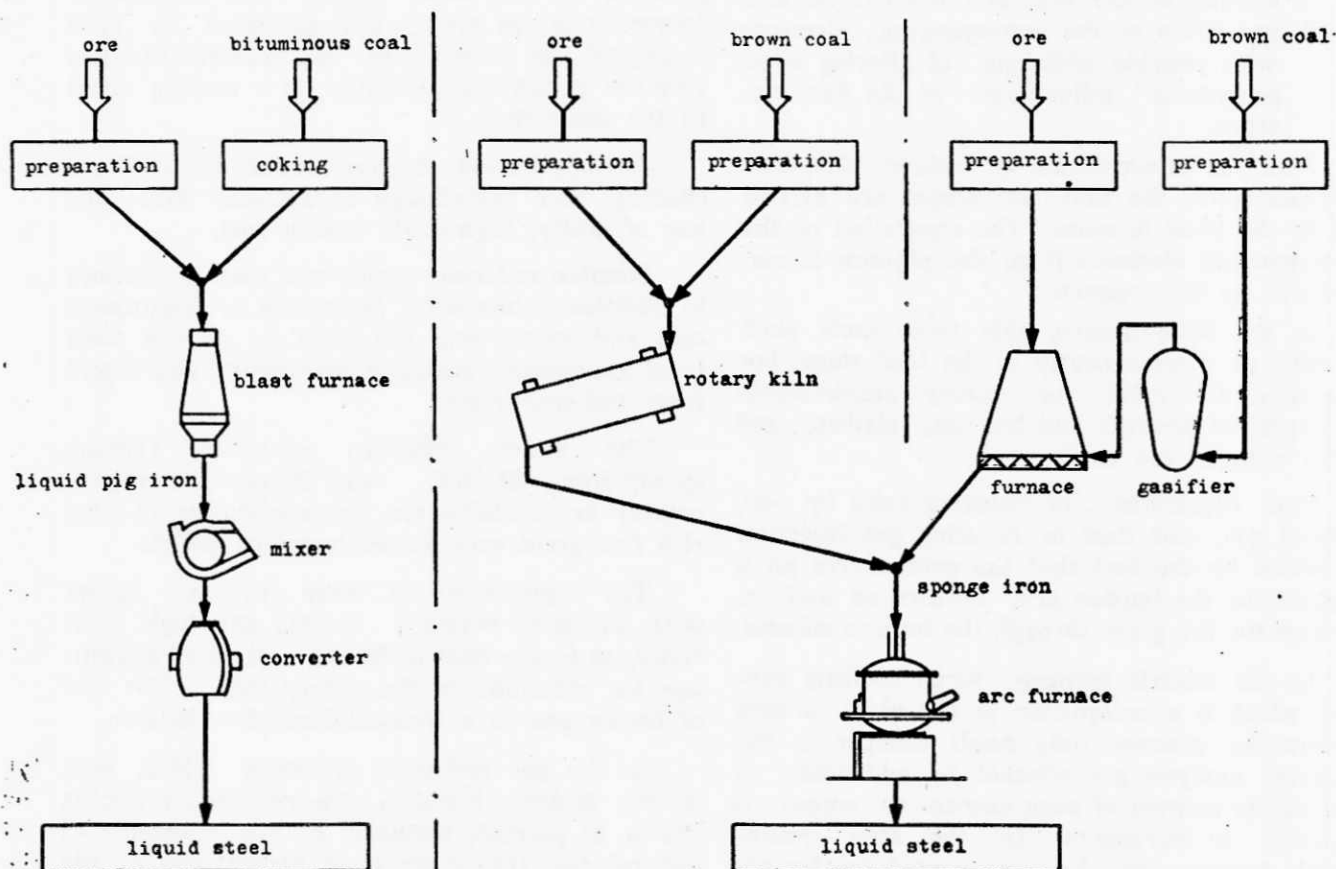


Fig. 1 comparison: Production of liquid steel by the blast furnace-converter-or direct-reduction-arc furnace process

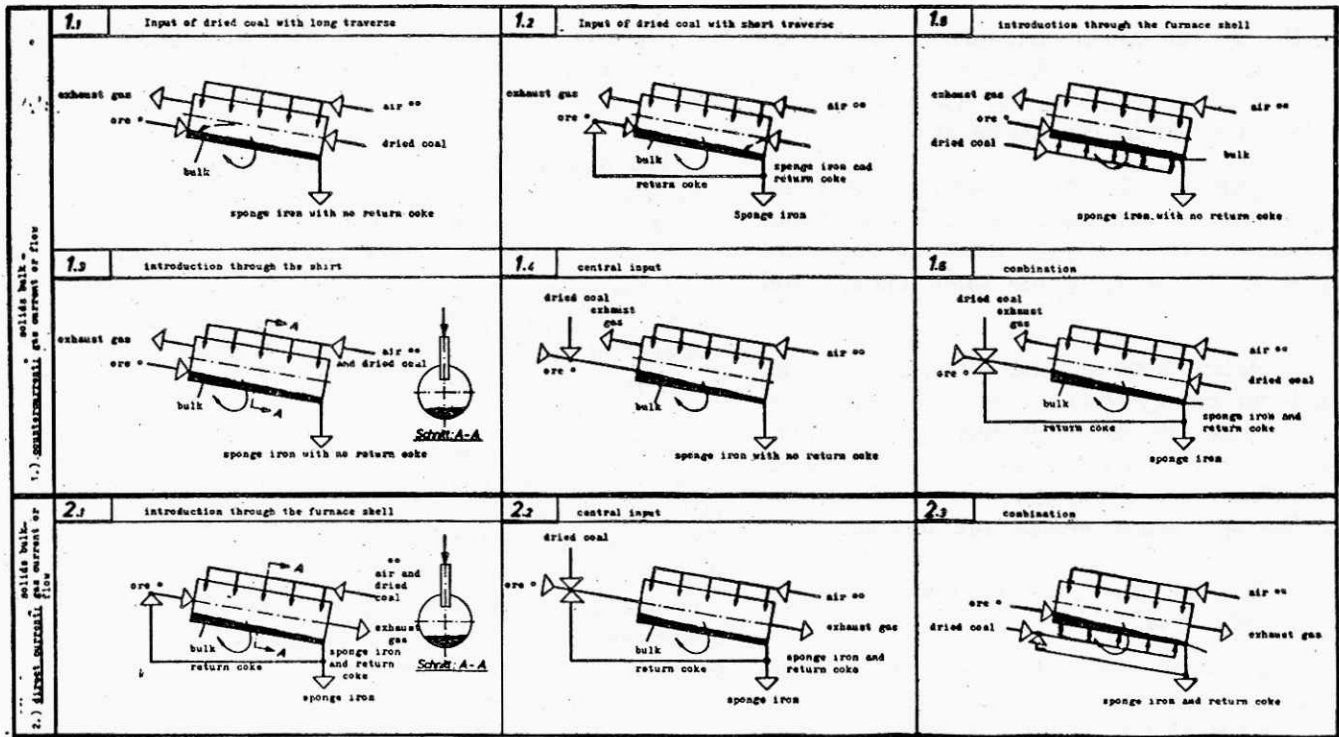


Fig. 2 Different variants for direct reduction in a rotary kiln with input of dried brown coal
 * ore (possibly preheated); ** air (possibly with oxygen enriched)

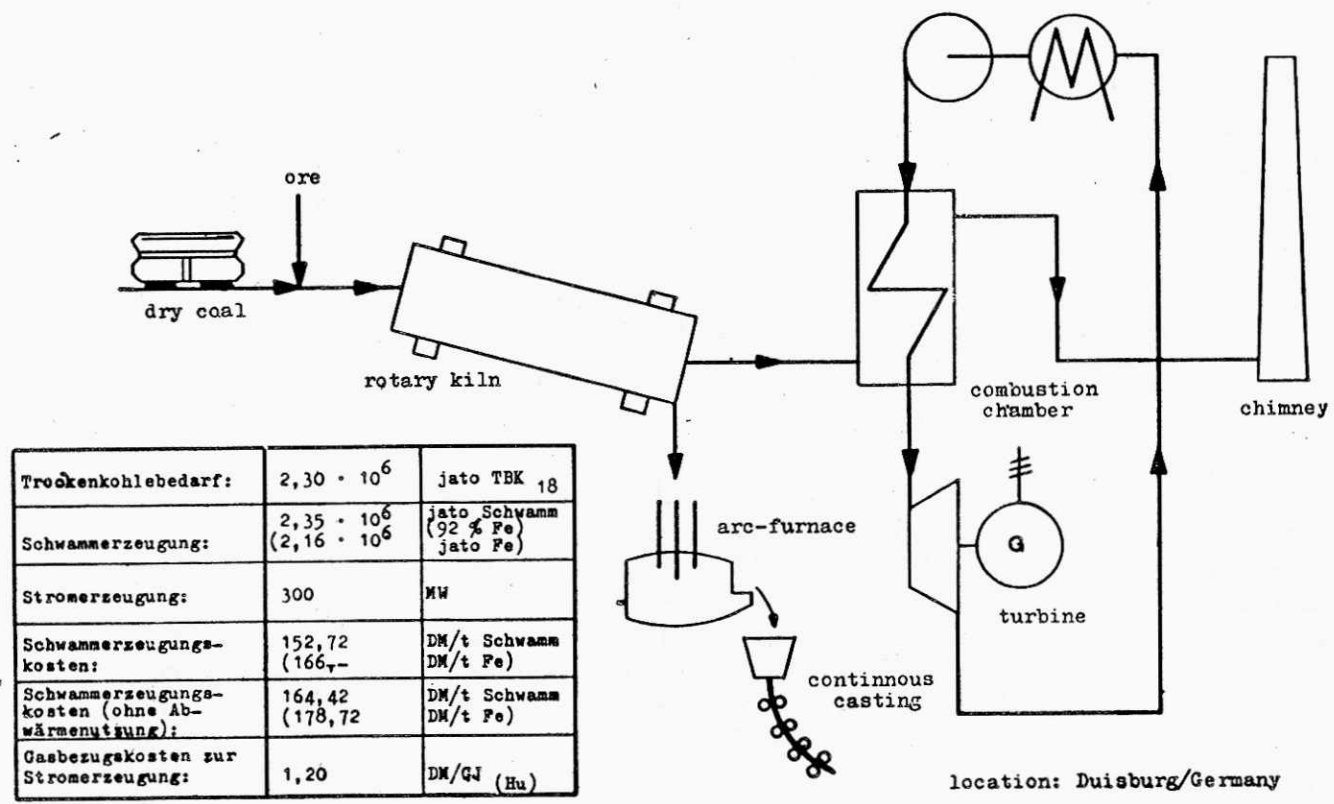


Fig. 3 Steel production by reducing ore in an rotary kiln-power production

In the sponge-iron-electric-furnace-route an average was taken of the sponge-iron production costs in the Krupp, SL/RN, and the Midrex processes and inserted in the above table.

A comparison of the blast-furnace-LD-route with the direct reduction-electric-furnace-route shows that the sum of the material, fuel, and energy-costs is practically the same (74.3% and 74.4%).

The relation between the material-costs and the fuel-and energy-costs determines the most economic basis in the long-term trend (with slowly rising fuel-and energy-costs in the sponge-iron-electric-furnace route the trend will be towards the sponge-iron-electric-furnace-route).

Rotary kiln processes using brown coal from the Rhine-area

A large scale rotary kiln process plant using New Zealand brown coal is operating with a rated capacity of 140×10^3 t yearly production of sponge iron. The dimensions of the plant are 4 m \varnothing and 75 m length.

Investigations on the reactivity of New Zealand brown coal towards carbon-dioxide (Boudouard-reaction) and on the chemical composition of coke from Rhine-and New Zealand-brown coal have shown these to be very similar.

Comprehensive tests by the firms Krupp, Lurgi and Polysius in their pilot-plants in Essen, Frankfurt and Neubeckum in Germany have confirmed that (Brown coal dried to a briquetting

water content of about 15 to 18% is very suitable for the production of sponge-iron in the rotary kiln).
ETA

The application of non-coking brown coal with increased volatiles offers many advantages in the reduction in the rotary kiln.

Brown coal with a softening point of the basic ash of above 1200K does not require the addition of lime as desulfurising agent. Also there is no residue coke.

Due to the high reactivity of brown coal, high performance rates in the rotary kiln may be attained as the rate-determining step of the whole process is the conversion of the carbon-dioxide formed by the ore-reduction with the solid carbon of the fuel. The endothermic Boudouard-reaction in the gas regeneration fixes the performance of the rotary kiln in respect of both the required reaction heat and the material changes.

Table 2 shows the reduction results obtained with Rhine brown coal.

The reduction in the rotary kiln with brown coal also yields an usable gas, which can be burnt in a waste heat boiler to deliver steam or power.

The utilization of the waste gases is closely linked with the economics. To operate a 300 MW_{el}-unit for the utilisation of waste gases a yearly production of sponge iron

Table 2: Chemical analysis of sponge iron from a SL/RN-rotary kiln⁵

Chem. analysis (weight %)

Fe-carrier	Ore		Sponge iron			
	Fe	S	Fe	S	C	metallization
Hamersley-lump ore (6-20 mm)	66,2	0,018	92,9	0,020	0,12	96,8
Hamersley-lump ore (6-40 mm)	67,2	0,007	93,5	0,015	0,12	98,5
Kiruna-pellets	64,1	0,010	86,8	0,012	0,30	97,2
Itabira-lump ore	68,1	0,005	95,2	0,010	0,18	97,0
Itabira-pellets	65,8	0,005	90,5	0,008	0,30	99,2
Grangesberg-pellets	70,8	0,028	95,3	0,017	0,12	96,6
Eveleth-pellets	67,2	0,013	90,8	0,016	0,32	99,0
Wabush-concentrate	65,7	0,030	91,0	0,020	0,13	95,7
dust of rolling mills	70,4	0,010	89,0	0,020	0,37	97,4
LDAC-converter dust-pellets	60,7	0,200	84,0	0,200	1,03	98,6

of about 2.35×10^6 t yearly (2.16×10^6 t yearly of iron with 92% iron in the sponge) is necessary.

At the present state of rotary kiln technology at least 4 rotary kilns would be necessary to operate the above size of electric unit. Such a combination related to the location Duisburg would in any case be attractive in the light of increasing shortages of natural gas for power generation. This cost exercise (fig. 3) is based on coal costs (or partially dried coal with 18% water content) of 2.48 DM/GJ (10.38 DM/Gcal) and lump ore costs of 82 DM/t of iron. The waste gas produced at about 1123 K with about 0.55 G J/t sponge iron (approx. 2.3 Gcal/t sponge iron) is rated at 1.2 DM/GJ (5.0 DM/Gcal).

Gas reduction processes using coal

The conventional processes for the production of reducing gases from coal have not been able up to now to satisfy the requirements of a reducing gas with respect to the degree of oxidation.

The previous gasification processes are characterized partly by discontinuous operation, the production of tar, dust, sulfur, water, steam, carbon-dioxide and methane, and partly by low gasification temperatures and insufficient gasification performance.

The need for gas purification and gas heating in these processes limits their application in the production of reducing gases. A high-temperature-gasification HTW process will now be considered that very largely fulfills the aims of higher gasification performance and higher reduction potential. This process is called the high-temperature-Winkler-process (HTW-process).

In this process the reducing gas can be produced with the correct analysis directly "in one heat" i.e. without the need for any gas-conversion, gas-washing, or gas-desulfurization.

The exit temperature of the reducing gas from the gasifier is, at least in the case of reactive brown coal, of the order of the gas temperature required at the entry into the reduction shaft.

In addition to brown coal, practically all types of coal can be utilized in the high temperature Winkler-generator, which can be designed for large scale units (special measures may be necessary to overcome difficulties with coking coals).

For reasons of economy air can be used as gasification medium: oxygen as gasification medium can be dispensed with.

Working principle of the HTW-process*

In the normal Winkler-process the low reac-

tion temperature of about 1200 K. prevents fusion of ash. This relatively low temperature must be observed, since the ash from normally available coals tends to cake together and cause build up above this temperature.

This has up to now prevented the application of fluidized bed principles to large scale conditions at higher temperatures.

The new approach of the HTW-process consists in the addition of such substances to the coal to be gasified that form a product of higher melting point with the coal ash. The addition of lime stone is particularly advantageous.

Even with gasification temperatures in the range of 1800 K there was no molten slag formed, only a granular sinter, which could be removed without difficulty.

Due to its high reactivity, the addition of brown coal yields at gasification temperatures of about 1400 K already a reducing gas rich in $\text{CO} + \text{H}_2$ and poor in $\text{CO}_2 + \text{H}_2\text{O}$.

Experimental investigations of the high temperature Winkler-process

The investigations in the Institut für Eisenhüttenkunde der RWTH Aachen were conducted in the experimental apparatus illustrated in fig. 4. In an experimental apparatus consisting of an externally heat-insulated cylindrical tube rammed up in alumina clay (100 mm \varnothing , 1500 mm high). The coal was introduced into the reaction tube by means of a screw feed and the ash, which becomes granular after treatment with lime stone, was removed by means of a discharge conveyor. The gasification medium, in the form of indirect preheated air from a combustion chamber was blown in through a porous stone diaphragm as inlet. The coal was gasified concurrently with the air in the fluidized bed, the exit velocity in the fluidized bed being only slightly above the lower fluidizing velocity.

* The HTW-process is a further development of the Winkler-generator, named after the discoverer and developed as the first large scale 'Fluidized Bed Process' at the German BASF in 1922.

Using Rhine brown coal, 2 gas generators with an output per gasifier of up to $17 \times 10^3 \text{ Nm}^3/\text{h}$ have been operating with success at temperatures of up to 1173 K according to the Winkler-principle for the production of synthetic gas in UK Wesseling, a subsidiary company of the Rheinische Braunkohlenwerke.

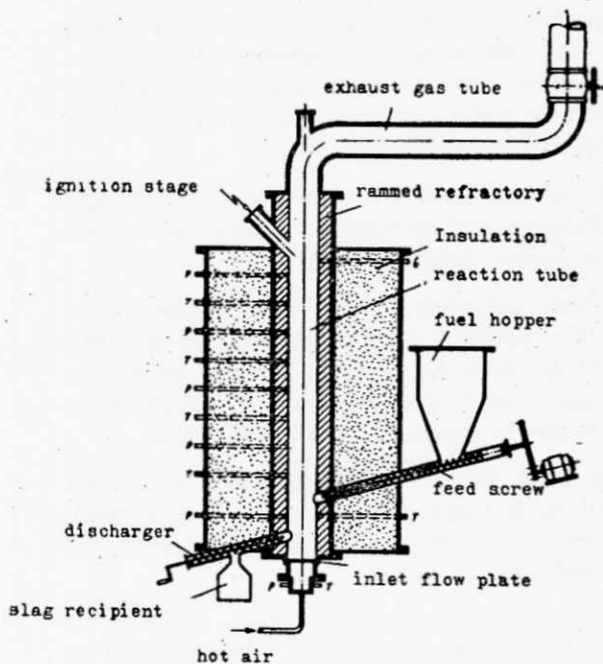


Fig. 4 Laboratory apparatus for the gasification of low carbonized brown coal and dried brown coal

The influence of the mean gasifying temperature on the composition of the product gas is shown in fig. 5 for low temperature carbonized brown coal (carbonization temperature 573 K, grain size 0-10 mm). The relation between the output and the mean reaction temperature in fig. 6 indicates the important rise in output in the high temperature procedure to be obtained by lime stone addition to the coal, as opposed to the conventional Winkler-process.*

Possible alternatives for the linking of gasification units with iron ore reduction furnaces have been indicated.

The high temperature Winkler-generator can be linked with the normal counter current shaft furnaces or with the so-called cross-flow-counter-current shaft furnaces. In this case, a part of the top gas can be drawn off from the magnetite

* In the technical scale predried brown coal with a water content of 8% will be used.

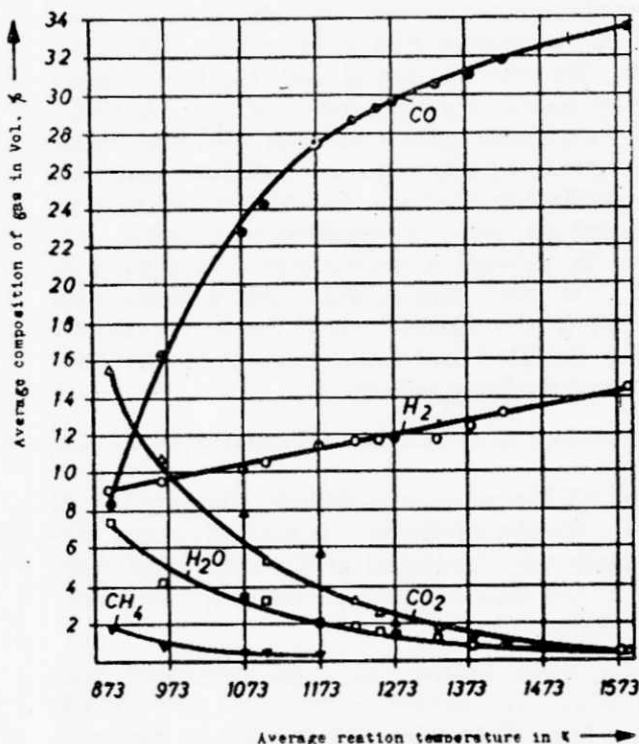


Fig. 5 Composition of dried gas from gasification of low carbonized Rheinisch Brown Coal with air attained by the RTW-Process at different reaction temperatures (temperature of low carbonization 573 K)

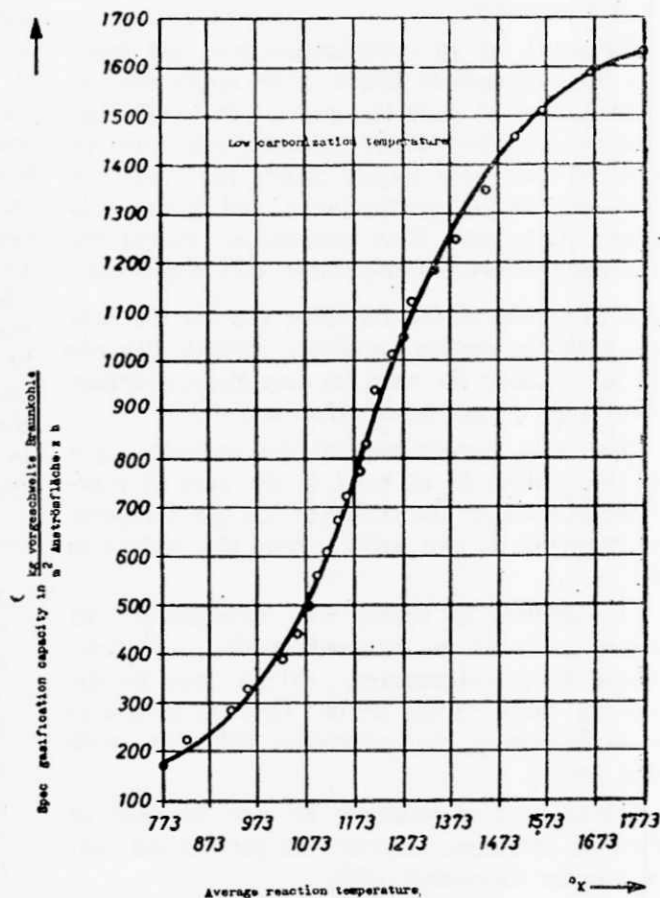


Fig. 6 Gasification capacity by reaction of low carbonized coal with air attained by the RTW-Process at different reaction temperatures

stage and introduced into a $\text{CO}_2/\text{H}_2\text{O}$ -washer. The top gas, largely free of oxidizing components can serve either as auxiliary gasifying medium in the gasifier (for further regeneration) or it can be mixed with the generated reducing gas predried or low temperature carbonized brown coal can be used. The predried or low temperature carbonized coal, with additions of agents such as lime stone is converted in the high temperature Winkler-gasifier by the gasifying medium, possibly containing additions of partially regenerated top gas. In the case of autothermal gasification the heat required for the endothermic gasifying reaction is provided by the exothermic conversion of the fuel. With the high temperature Winkler-generator a process is aimed at, in which the advantages of gasifying at high temperatures are combined with the advantages of gasifying in the fluidized bed.

The artificial enrichment of the coal ash with lime stone gives, on gasifying at high temperatures, a high melting granular slag, which does not cause obstruction or bridging in the gas generator.

Due to the coarse grain of the slag it is to be expected that this slag will separate from the fluidized coal and settle on the bottom of the generator. On a small scale this effect has already been established. A special advantage of the high temperature Winkler-generator is given in the presence of lime stone the metallurgical desulfurization of the gas occurs in the generator itself. The hydrogen formed by the addition of volatiles and of water vapour reacts with the sulfur, so that the sulfur in the generating gas occurs chiefly as H_2S , with air or oxygen as gasifying medium.

The metallurgical desulfurization depends on the coal being mixed with a sufficient quantity of lime stone (approximately 100 g lime stone per kg. dry fuel) to convert the hydrogen sulfide directly after its formation into CaS with the CaO present. Furthermore the lime forms with the ash a granular slag whose chemical composition approximates closely to that of normal cement (cement however may only be considered a possible by-product, as the gas quality determines the operating procedure of the generator).

Alternatives for the link-system between high temperature Winkler-generator and direct reduction plant.

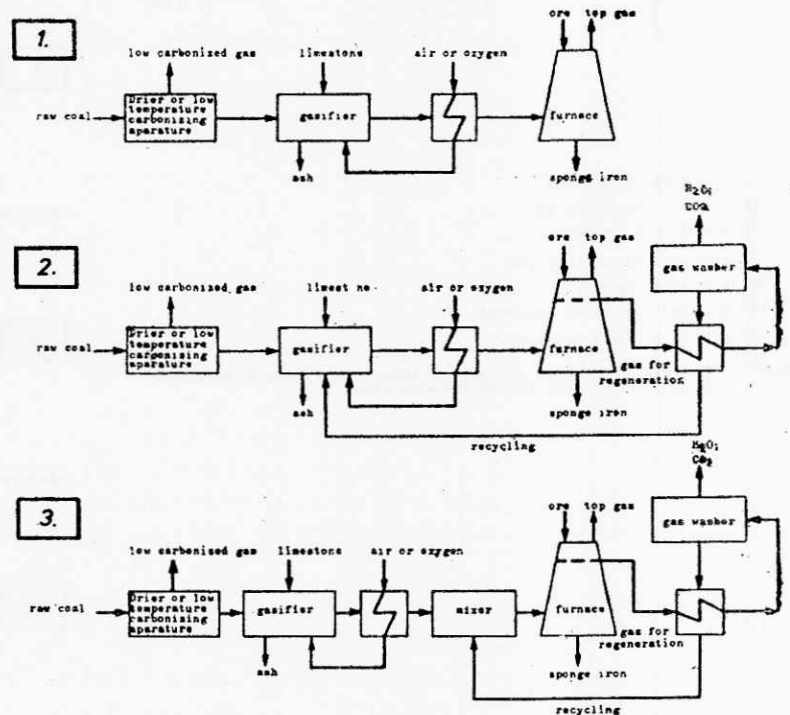
For the systematic project planning of a sponge iron plant with reducing gas based on solid carbon carriers the alternatives 1, 2, and 3 in fig. 9 were programmed.

With the aid of computer analysis the influence of the following parameters was investigated.

- additives (CaCO_3 , CaO)
- gasifying media (oxygen enriched air, air)
- gasifying pressure
- shaft pressure
- need for a CO_2 -wash.

A comparison of the possible gas utilization with the shaft furnace reduction process has shown that (The optimum effect is obtained with a continuous counter current process with removal of part of the gas flow from the magnetite stage. for regeneration).

The programmed counter-current shaft furnace follows the operating line shown in variation 2 of fig.



Control and adjustment alternatives			
1.	One-stage process		
2.	Multi-stage process with gas recycling		
3.	Multi-stage process with mixer		
1.1	2.1	3.1	Drier, air
1.2	2.2	3.2	Drier, oxygen
1.3	2.3	3.3	Low-temperature carboniser, air
1.4	2.4	3.4	Low-temperature carboniser, oxygen
1.5	2.5	3.5	Drier, enriched with oxygen
1.6	2.6	3.6	Low-temperature carboniser, enriched with air
1.7	2.7	3.7	Pressure in the gasifier and in the furnace
1.8	2.8	3.8	Preheating of gasification medium
-	2.9	3.9	H_2O and CO_2 Cleaning

Programmed possibilities for the combination of the HFM-Gasifier and the production of sponge iron

Fig. 9

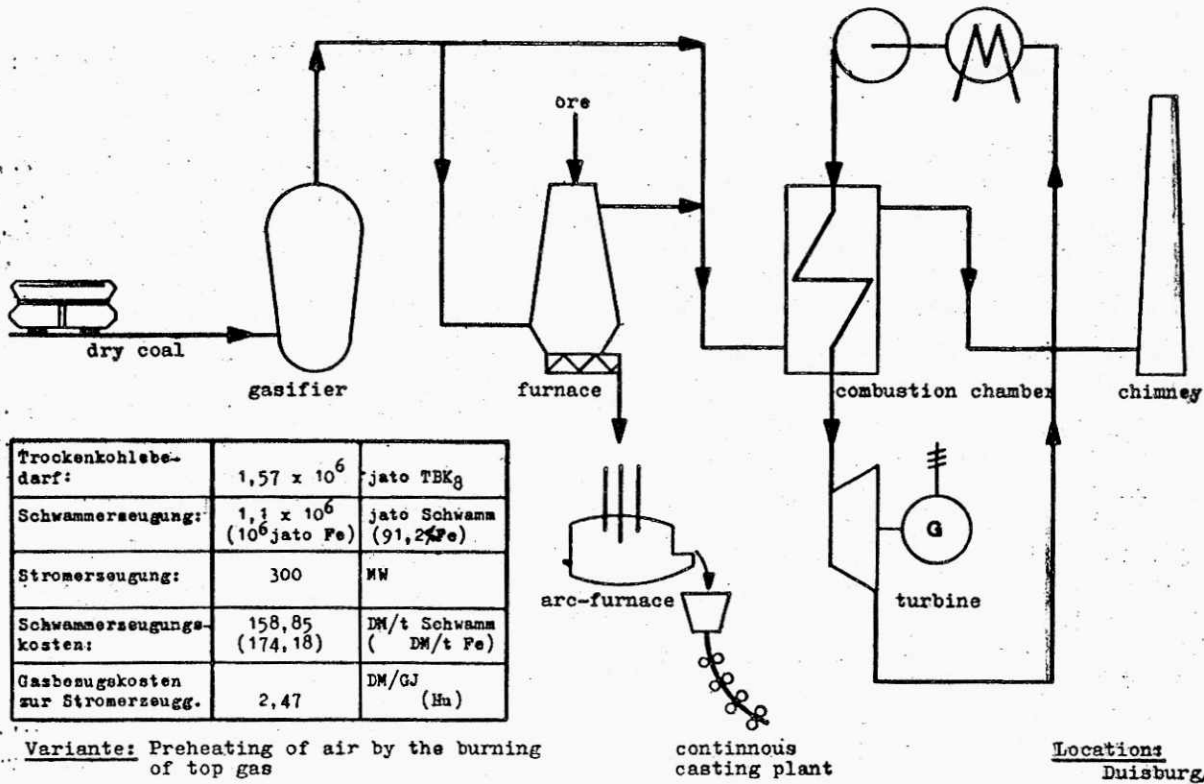


Fig.:10 Steel production by reducing iron ore with gas from brown coal

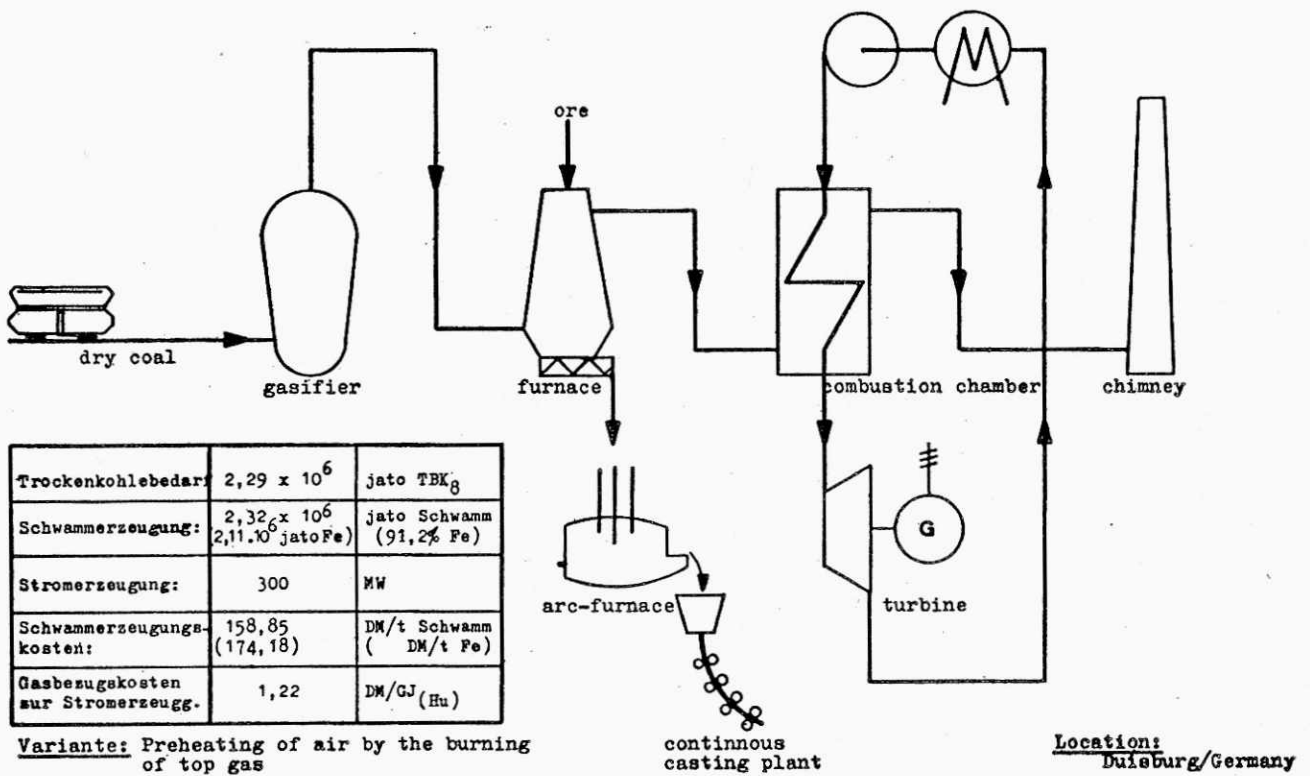


Fig.:11 Steel production by reducing iron ore with gas from brown coal

The necessary quantity of reducing gas is fed continuously into the counter-current shaft furnace.

A portion of the top gas is drawn off from the magnetite stage for regeneration, i.e. the product of gas quantity and reduction potential of the successive stage is lowered. The top gas drawn off from the magnetite stage is introduced into a CO₂-wash with water condensation.

Initial economic considerations of possible link alternatives with the high temperature Winkler process

Based on the location Duisburg in Germany various alternatives for the coupling of the high temperature Winkler-process with a direct reduction plant have been worked out. For example, in the single stage reduction shaft of conventional shaft design (without extraction of gas from the magnetite stage) and based in a yearly production of sponge iron of 10⁶ t of iron, top gas is fed into the power generator, which for the operation of a 300 MW-unit takes further

fresh gas from the high temperature Winkler-process. The relevant data are compiled in fig. 10.

With a yearly production of 2.11 x 10⁶ t of iron, the power generation of 300 MW is completely met by top gas, as can be seen in fig. 11. If a multi-stage shaft furnace procedure is adopted, with removal of part of the gas flow from the magnetite stage, the relationships obtained after gas purification of the drawn off portion of gas may be set up as in fig. 12.

The calculations are based on the coal costs at Duisburg with a price of dried coal (8% water content) of 2.42 DM/GJ and gas purification costs of 7 Dpf/Nm³ washed out CO₂.

The low costs obtained for sponge iron with gas purification result chiefly from the low coal inputs.

Summary

The economics of steel production from sponge iron depend in the long term on the evo-

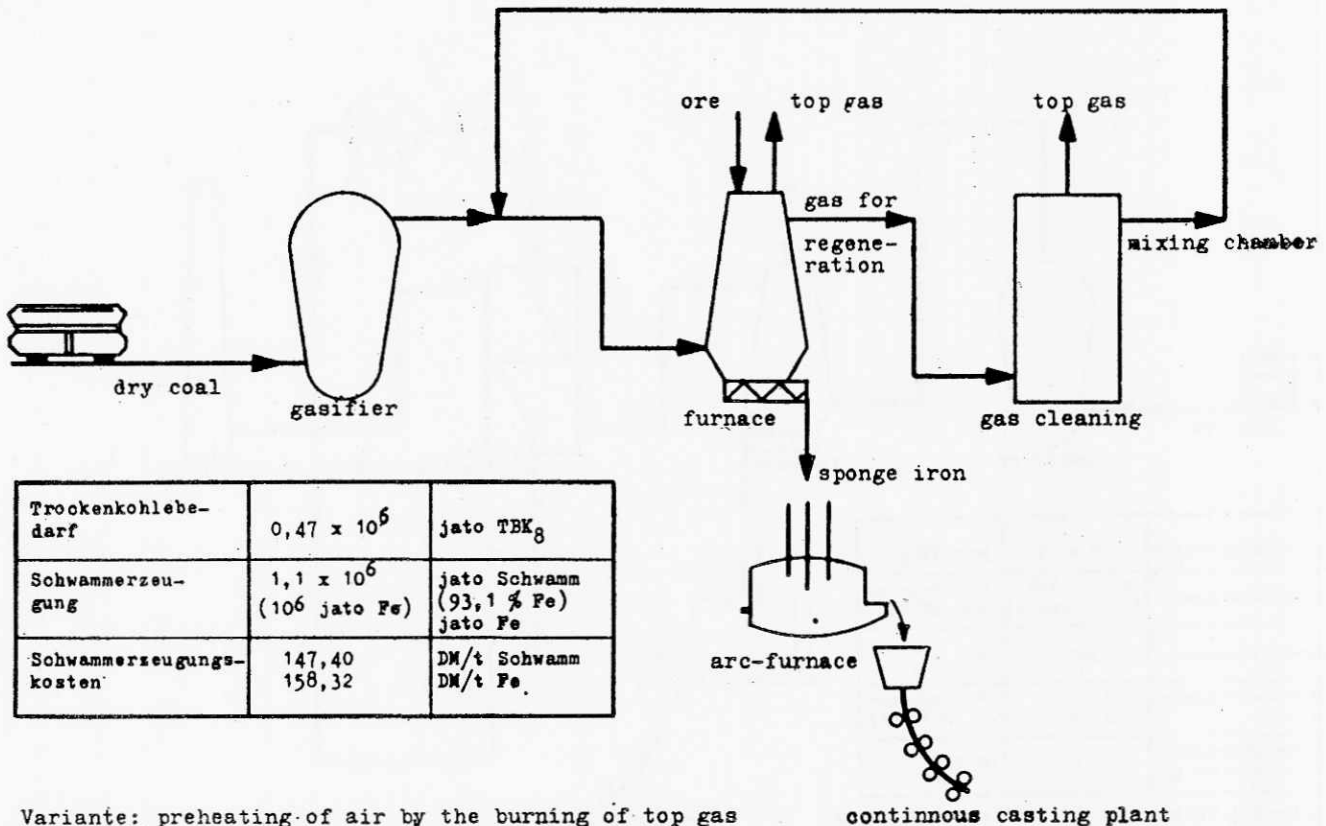


Fig. 12 Steel production by reducing iron ore with gas from brown coal

lution of the ratio material costs to fuel and energy costs. On the assumption of low fuel and energy costs, rotary kiln technology has been illustrated within the concept of the sponge iron-electric-furnace-route using brown coal particularly in the light of impending shortages of natural gas in Germany for power production. The utilization of the exhaust gases produced has a very deciding influence on the economics of rotary kiln technology with brown coal.

In addition the production of reducing gases for reduction shaft furnace on the basis of coal has been treated.

The production of reducing gases from the so-called high temperature Winkler-process, capable of realisation in the medium term, has been discussed.

A desulfurized reducing gas with a degree of equal or less than 5% can be produced directly from the gasifier and fed into the reduction unit, such as a shaft furnace.

Data assembled from test results have been introduced into the cost analysis of possible link systems between for gas generator, reduction plant, power generator or gas generator.

The funds for this research were provided by Landesamt für Forschung des Landes Nordrhein-Westfalen, Düsseldorf and the firm Rheinische Braunkohlenwerke AG, Kiln.

The evaluation of the test data and the link alternatives was carried out in the computer centre of the RWTH Aachen.

Literature

1. Femmer, U.
Dissertation, RWTH Aachen, Jan. 1973.
2. Franke, F.H. u. U. Femmer
Nichtöffentliche Studie:
Technische und wirtschaftliche Basis der Erzeugung von Eisenschwamm, Köln, Juli 1972.
3. Wenzel, W., F.H. Franke und M. Meraikib.
Braunkohle, Heft 4, S. 119/28 (Juli 1971).
4. Meyer, G. und R. Wetzel
Techn. Mitt. Krupp, Werksberichte Bd. 30, Heft 1, S. 19/28 (1972)
5. Janke, W. u. G. Reuter.
Braunkohle, Heft 4, S. 110/13 (1971).
6. Meraikib, M.
Dissertation, RWTH Aachen (1967).
7. Dreyer, B.
Dissertation, RWTH Aachen in Vorbereitung.
8. Meraikib, M. und F.H. Franke
Chemie-Ing: Technik 42, Nr. 12., S. 834/36 (1970).
9. Franke, F. H.
Habilitation, RWTH Aachen in Vorbereitung
10. Flesch, W. u. G. Velling
Erdöl, Kohle, Erdgas, Petrochemie 15, S. 710/13 (1962).