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INDUSTRY TO DEVELOPING COUNTRIES**

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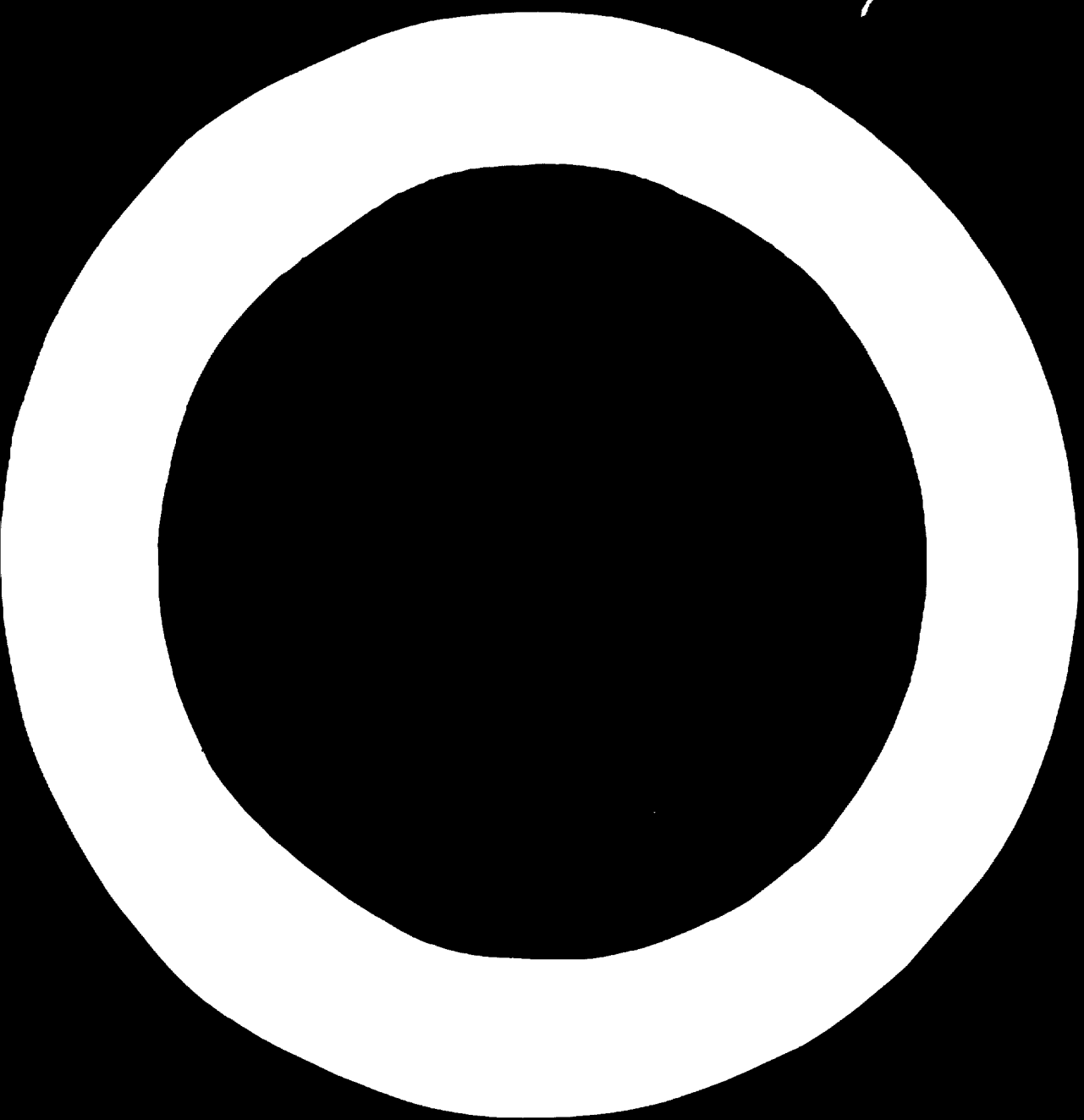
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**THE PUROFER DIRECT REDUCTION PROCESS**

**by**

**Ludwig Von Bogdandy**

**GE.63-15145**



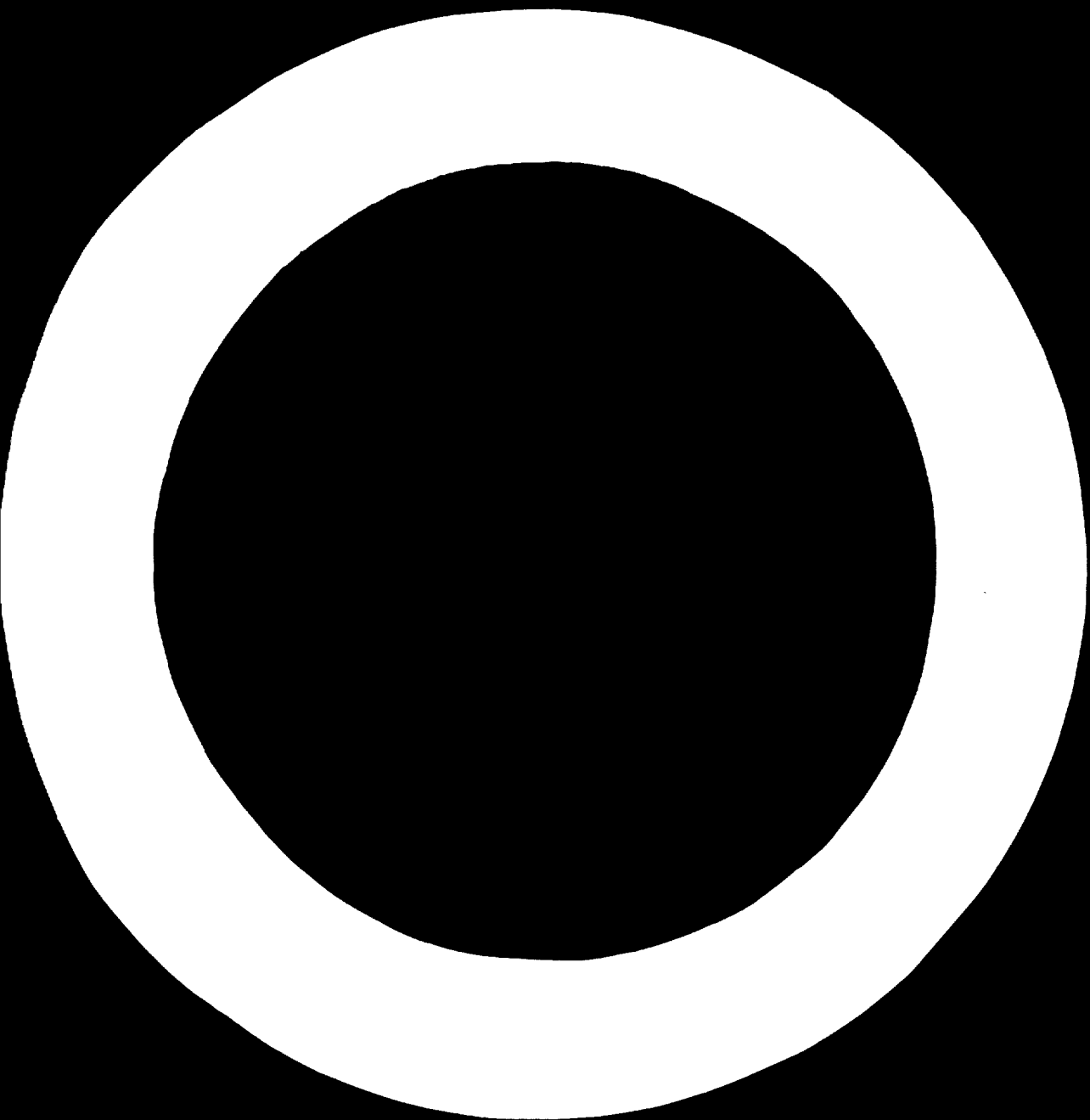
## THE PUROFER DIRECT REDUCTION PROCESS \*)

DR. LUDWIG VON BOGDANDY

### Table of Contents

1. Modification of the conventional blast furnace process to the application of 100 % natural gas
  - 1a. Aspects of modern blast furnace technique
  - 1b. Adaptation of the blast furnace to small quantities of natural gas
  - 1c. Basis of the development leading to the application of 100 % natural gas in iron production
2. Explanation of the principles governing the reduction of iron ores with hydrogen-rich gas mixtures
3. Principles underlying the transformation of methane into mixtures of hydrogen and carbon monoxide
4. Construction of the pilot plant
  - 4a. Construction of the gas reformer
  - 4b. Construction of the reduction furnace
5. Results of experimental section
  - 5a. Gas reforming
  - 5b. Reduction
6. Use of the sponge iron produced
7. Transformation to the industrial scale
8. Consideration of capital costs and operating costs

\*) This paper is based upon a publication of Dr. Ludwig von Bogdandy, "principles of the manufacture of sponge iron with natural gas in the shaft furnace", "STAHL UND EISEN" 82 (1962) Heft 13, S. 869/83, Verlag Stahleisen, Düsseldorf.



**1. Modification of the Conventional Blast Furnace Process to the Application of 100 p.c. Natural Gas**

The conversion of iron oxides into metallic iron is the basis for steel production, whatever steelmaking process may be used. The physical chemistry of the underlying reactions are now being understood as a result of extensive research work, which is underway in all major steel producing countries of the world.

The optimum conditions of reduction process with respect to the nature of the reducing gases, temperature and pressure in the reduction vessel and the optimum preparation of the ore can thus be predicted. Generally it seems that the shaft furnace with countercurrent flow of gases and solids gives the best conditions for the reaction if one considers chemical and thermal efficiency of the reducing gases and throughput of solid materials per unit volume of the vessel.

The shaft furnace for reduction with countercurrent flow of gases and solids has found worldwide application as the well known blast furnace for the production of liquid pig iron. Here the reducing gas is CO which is generated by the combustion of coke with compressed, heated atmospheric air. The conventional blast furnace is one of the most efficient units in applied chemistry, it can produce up to 2000 tons of iron per day and modern burden preparation has brought coke consumption down to 600 kg/t or less, figures which were not generally foreseen even a few years ago. Nevertheless, the coke-operated blast furnace seems no longer the right solution for all parts of the world.

It is well known that many countries are short of coking coal, but, on the other hand, possess natural gas in big quantities.

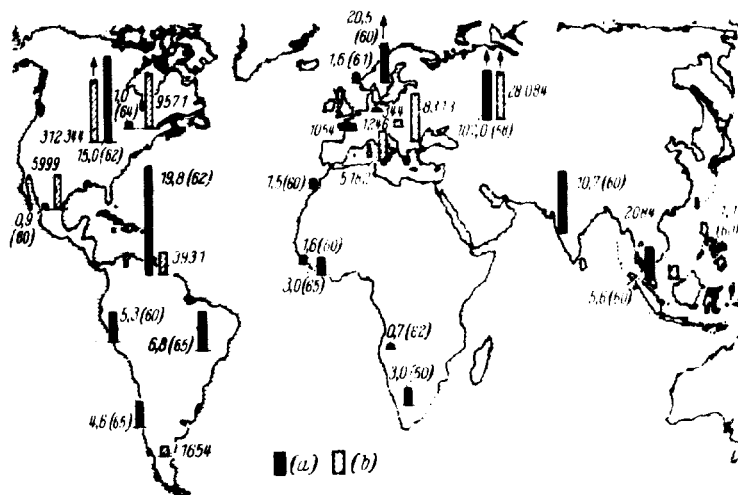
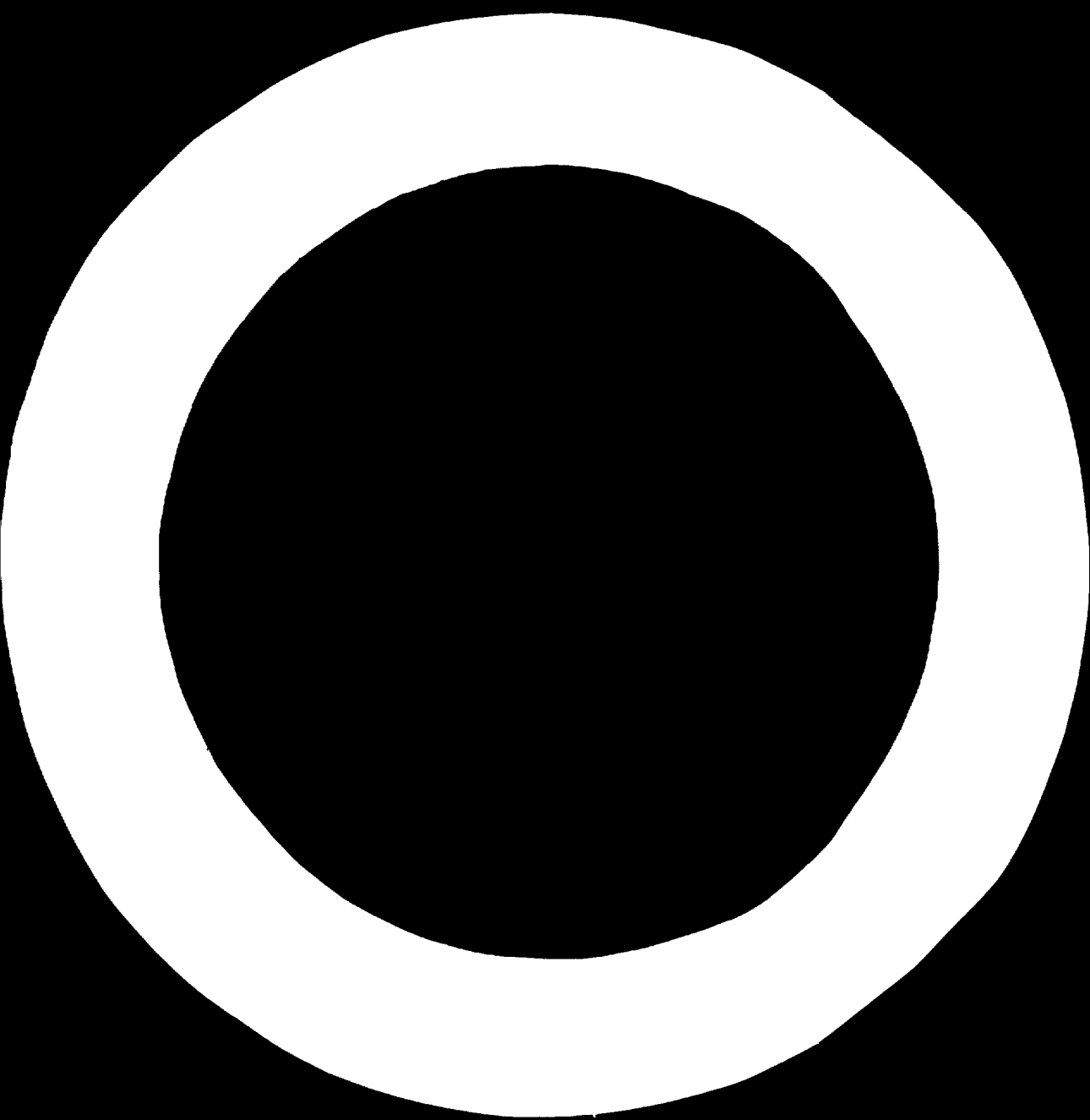


Bild 1. Rohstoffe für Direktreduktionsverfahren  
 Figure 1. Raw materials for direct reduction processes  
 a) Erzförderung mit über 58% Fe im Feuchtwert in Mill. t/Jahr einschließlich Pellets 1960. Zahlen in Klammern ... Fe. - Production of ore with more than 58% Fe in the wet, in million tons per annum, including pellets, in 1960. Figures in brackets ... Fe.  
 b) Gewinnung von Erdgas in Mill. Nm³/Jahr 1958. - Production of natural gas in million Nm³ per annum in 1958.



Moreover, it is in many places true that with increasing production and export of oil natural gas is simultaneously coming out of the earth in quantities that, at the moment cannot be transferred to industrial use. Thus, we have the contradictory situation, that some countries import coking coals or even coke for the production of pig iron, while their own reserves of natural gas are being burned off.

It seems, therefore, worthwhile to consider whether the blast furnace can be adapted to the use of 100 p.c. natural gas instead of metallurgical coke. For this transition there are some features of the blast furnace which must be taken into account.

### 1.a Aspects of modern blast furnace technique

The conventional blast furnace is simultaneously a gas producer, heat exchanger and reducer of iron ore. The functions may be characterized as follows:

The calculations are based on the results of one of our most modern blast furnaces, 8,5 m hearth diameter, 1450 m<sup>3</sup> working volume. This furnace uses a relatively rich fully prepared burden of 1900 kg/t pig iron, a hot blast temperature of 821°C; a dry coke consumption of 632 kg/t. The daily production is 1900 t pig iron.

The carbon of the coke is used for three purposes:

C in pig iron:	36 kg/t
C gasified by compressed air:	
C + 1/2 O <sub>2</sub> = CO	399 kg/t
C gasified in the "direct" reduction of ore: FeO + C = CO + Fe	122 kg/t
	<hr/>
	557 kg/t
	*****

The outgoing blast furnace gas is characterized as follows:

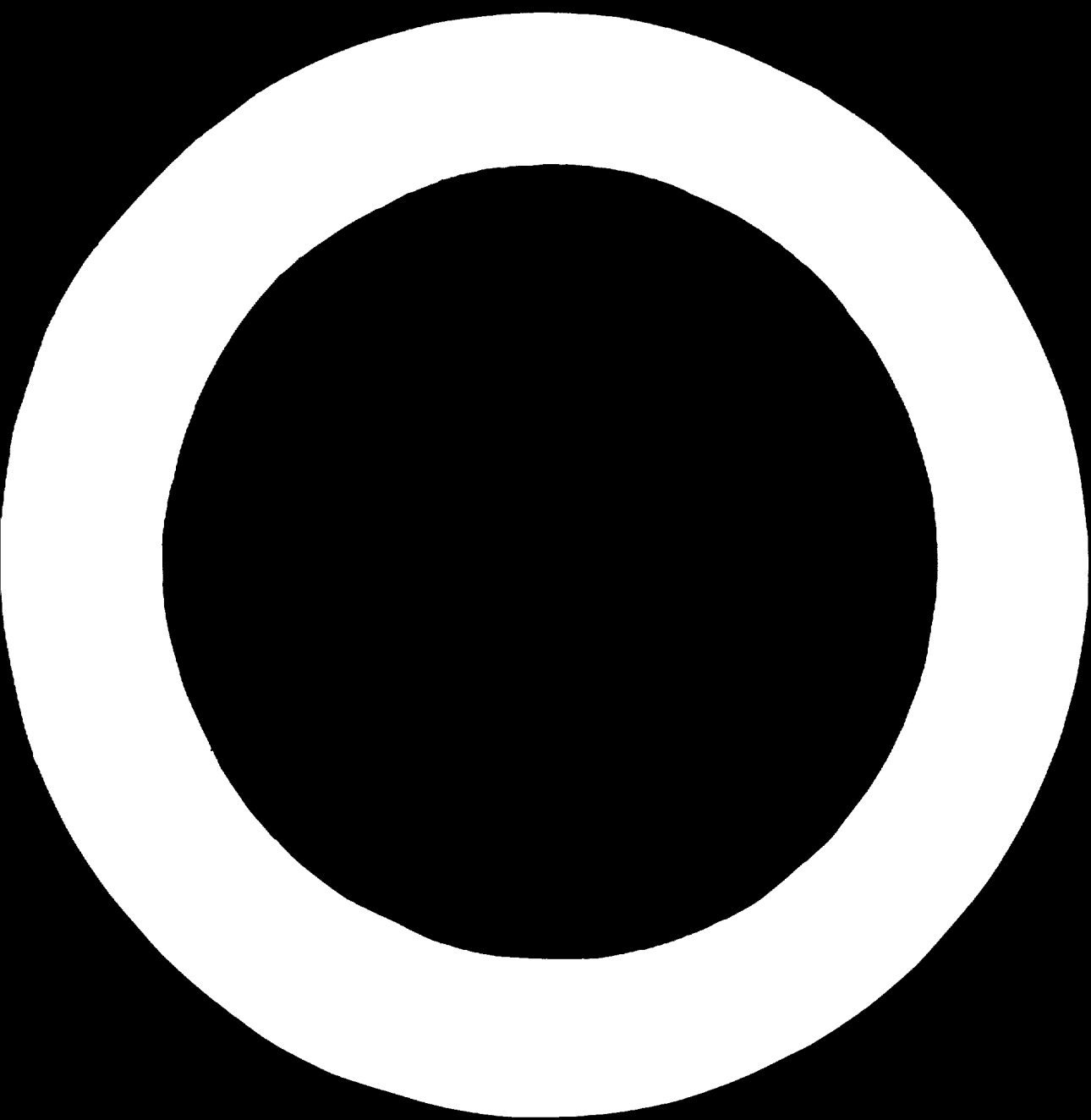
2364 Nm<sup>3</sup>/t, 27,6 p.c. CO, 13,7 % CO<sub>2</sub>, 229°C, H<sub>u</sub> = 879 kcal/Nm<sup>3</sup>.

The efficiency of the gas in the furnace is, therefore:

$$a) \text{ chemically} = \frac{\text{produced CO} - \text{outgoing CO}}{\text{produced CO}} = 33,0 \%$$

$$b) \text{ thermally} = \frac{H(\text{produced gas}) - H(\text{blast furnace gas})}{H(\text{produced gas})} = 87,0\%$$

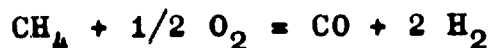




It is evident, that the thermal efficiency is very good the chemical efficiency however not very high. The unused CO cannot be circulated, it has to be used elsewhere. A further important characteristic of the blast furnace is its high productivity. One furnace with a working volume 1450 m<sup>3</sup> may produce 2000 t pig iron daily; that is 1.4 t/day x m<sup>3</sup>. The continuous manner of working leads to the possibility of a high degree of mechanization with respect to blast heating and burdening. The tapping - however - is a complicated operation, where human control is still necessary.

#### 1.b Adaptation of the blast furnace to small quantities of natural gas.

The simplest feasible way is to inject the natural gas into the blast tuyeres. In the hot zone near the mouth of the tuyeres the following reaction will take place:



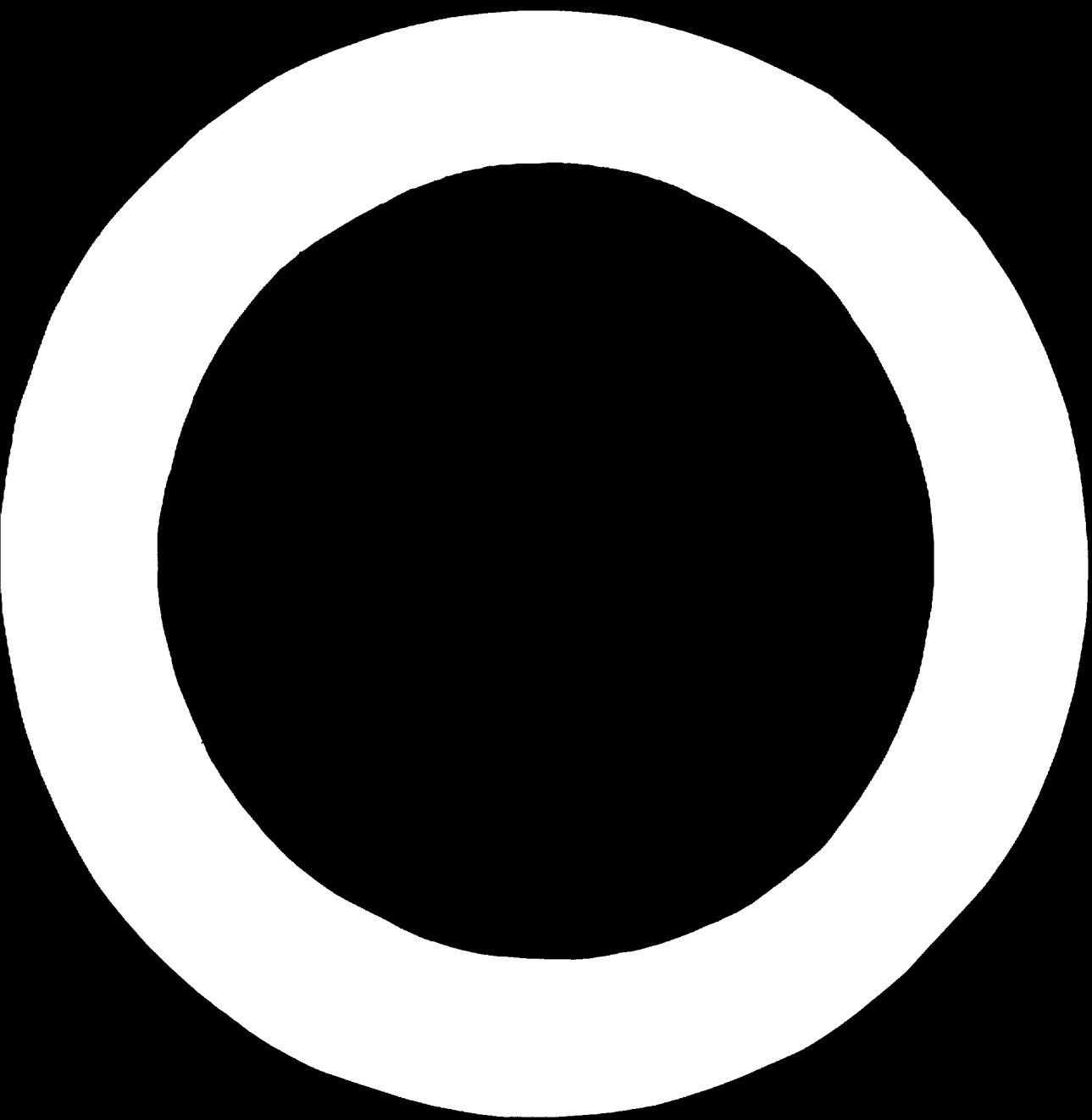
Now it is most important, that the temperature distribution in the lower part of the furnace will not be disturbed by this reaction. In other words: the average heat content of the tuyere gas cannot be lowered too much. The limit of natural gas addition will thus be approx. 100 Nm<sup>3</sup> CH<sub>4</sub>/t pig iron necessitating a substantial temperature increase in the hot blast. 100 Nm<sup>3</sup> CH<sub>4</sub>/t pig iron will replace approx. 200 kg coke/t pig iron that is 1/3 of the total consumption.

Summarizing it can be said, that the simple injection of natural gas into the tuyeres of conventional blast furnace can decrease the coke demand for the same production of pig iron by approx. 1/3, that is about 400 - 500 kg coke/t pig iron remain to be supplied. The transition to 100 % natural gas can only be achieved by further measures.

#### 1.c Basis of the development leading to the application of 100 % natural gas in iron production

The development work was directed to achieve the following points:

- I All heat and reducing gas required should be furnished by natural gas; no coke is allowed.
- II As many constructional characteristics as possible from conventional blast furnaces should be retained since they are based on great experience and minimize the risk of new installations.
- III The handling and reforming of natural gas should be as simple as possible taking the hot blast preparation as a model.



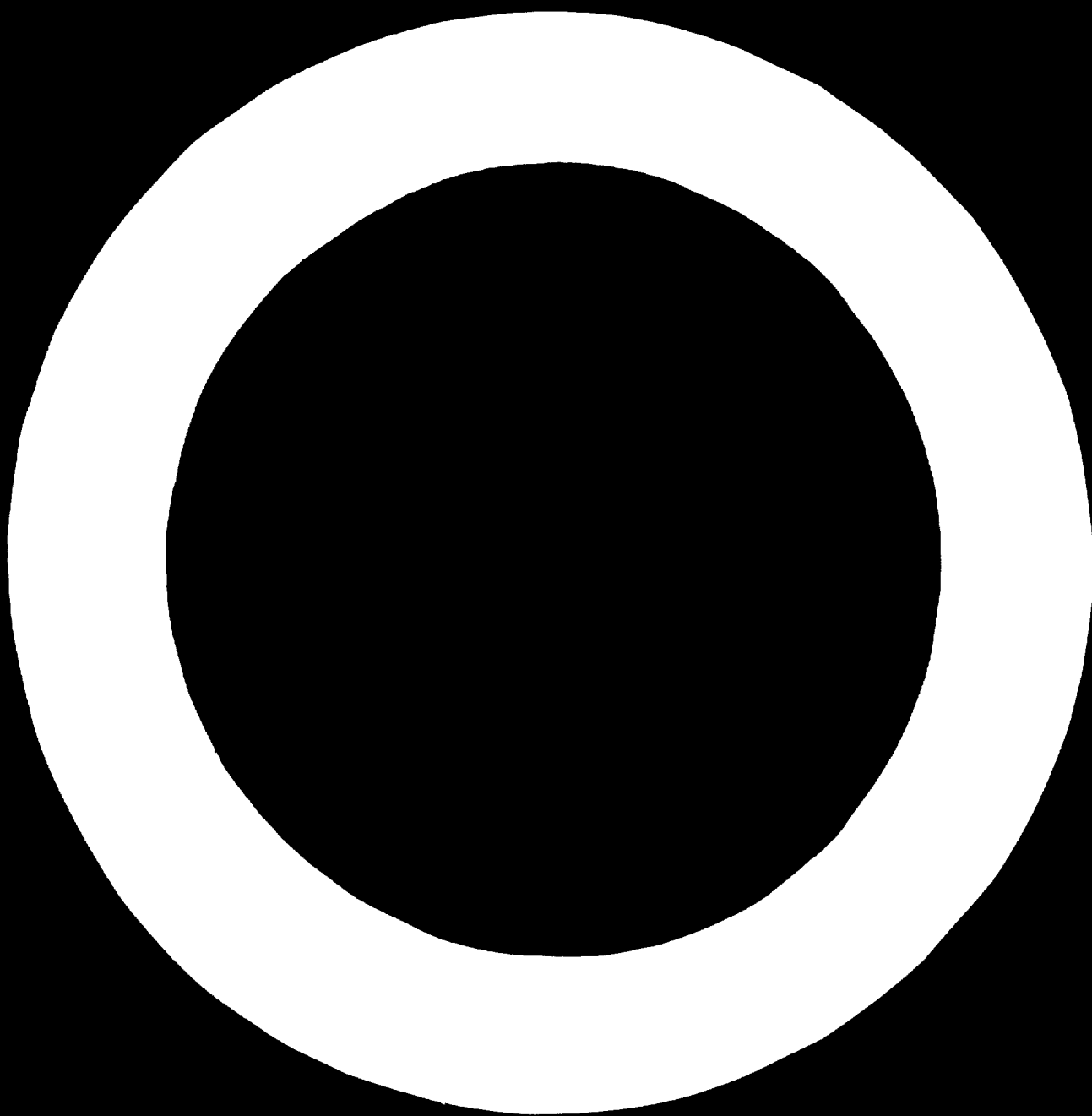
- IV The reducing gas should be used totally within the Reduction plant.
- V The throughput of iron ore and the production of metallic iron should be as high as possible, at least of the same order of magnitude as blast furnaces.
- VI The product should be sponge iron.  
The last demand results from the fact that most countries with high reserves of natural gas are short of scrap and, on the other hand, possess high grade iron ore with very low gangue contents.

2. Explanation of the principles governing the reduction of iron ores with hydrogen-rich gas mixtures

Methane, the main constituent of natural gas, is unsuitable for the reduction of iron ore. When reformed with oxygen carriers however a mixture of gases is formed which is rich in hydrogen and is a good reducing agent. The first step was to find out under what conditions, such as temperature, pressure, composition and flow rates the highest efficiency is obtained, i. e. good utilization and rapid reduction. The equilibrium diagram (figure 2) shows already that hydrogen-rich gases in equilibrium are utilized better at higher temperatures. To transform 1.43 metric tons of  $Fe_2O_3$  into 1 ton of iron 430 kg  $O_2$  must be removed; with complete transformation this corresponds to a hydrogen consumption of  $602 Nm^3/t Fe.$  -

As a consequence of the equilibrium however, larger proportions of hydrogen can react only at higher temperatures. Hence the equilibrium consumption - that is the theoretical lower limit of consumption possible - becomes less with rising temperature. (Fig. 3) With carbon monoxide the equilibrium curves have the opposite direction; here a lower temperature appears to be more favourable for equilibrium utilization. In fact temperatures less than  $700^\circ C$  cannot be used, as the decomposition of carbon monoxide commences below this temperature.

These equilibrium considerations have already shown the usefulness of high temperatures, while saying nothing about the reaction rates. For this reason the kinetics of reduction with hydrogen was examined in special experimental equipment (Fig. 4). The balance shown in the upper part of the diagram enables the reduction process to be observed continuously. Highly porous hematite pellets of various diameters were charged for the following experiments.



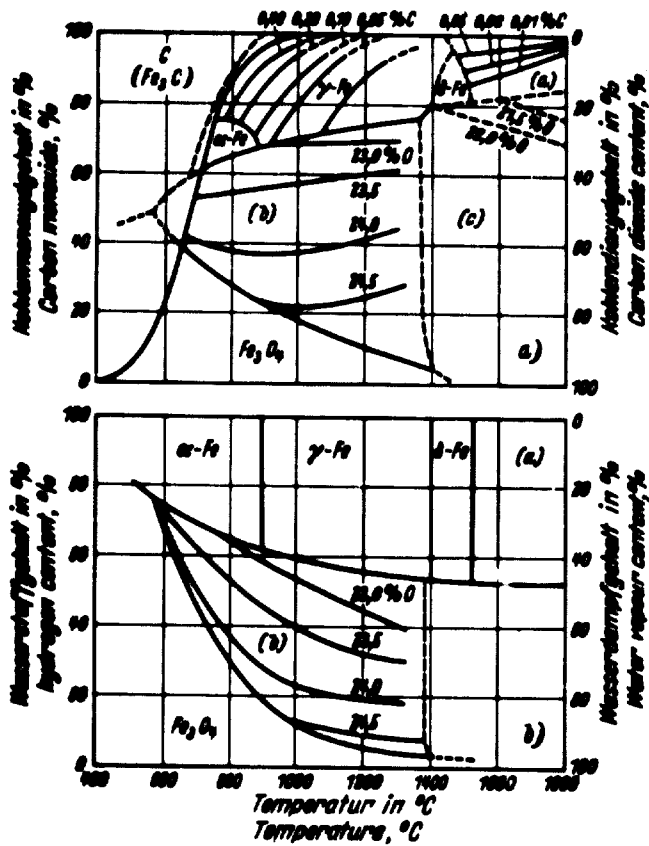


Figure 2

Bild 2. Reduktiongleichgewichte<sup>10)</sup>  
 Figure 2. Reduction equilibria<sup>10)</sup>  
 a) flüssiges Fe - liquid Fe b) Wüstit - wüstite  
 c) flüssiges Oxyd - liquid oxide

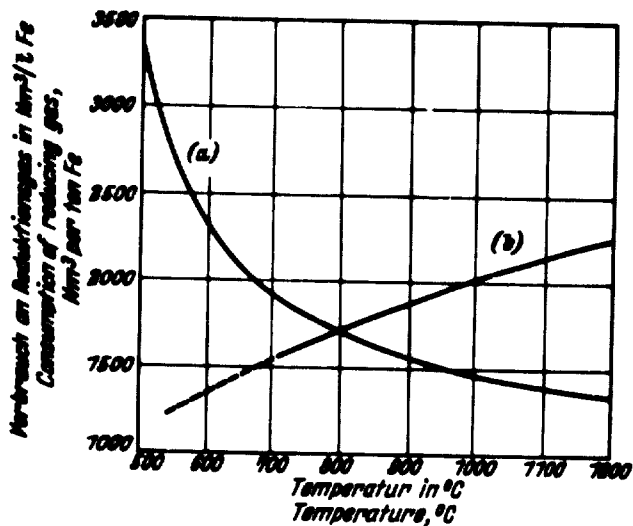
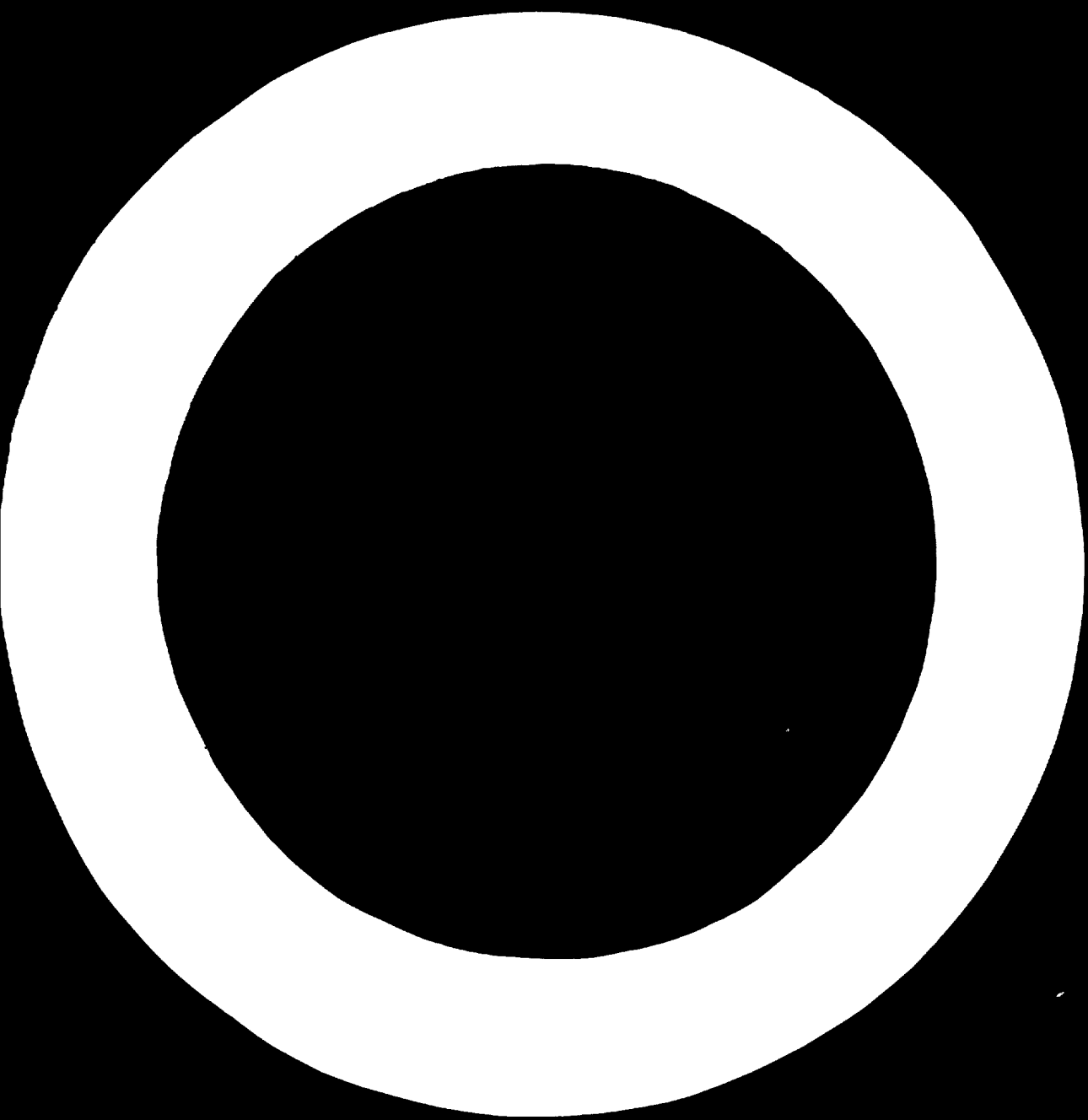


Figure 3

Bild 3. Reduktionsgasverbrauch im Gleichgewicht bei isothermer Reduktion von Eisen(III)-oxyd zu Eisen in Abhängigkeit von der Temperatur  
 Figure 3. Consumption of reducing gas in equilibrium in the isothermal reduction of iron(III)-oxide to iron as a function of temperature  
 a) Wasserstoff - hydrogen, b) Kohlenmonoxyd - carbon monoxide



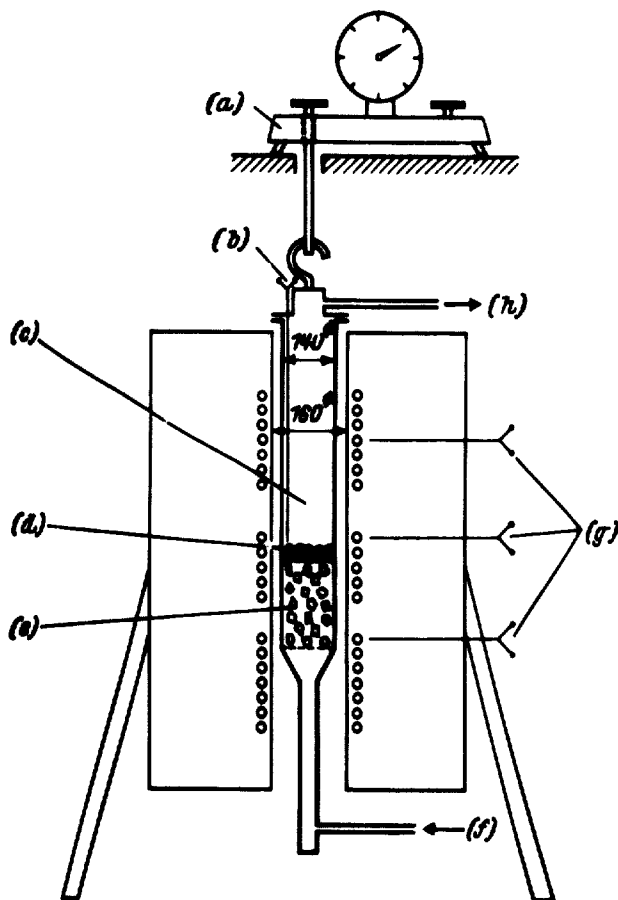


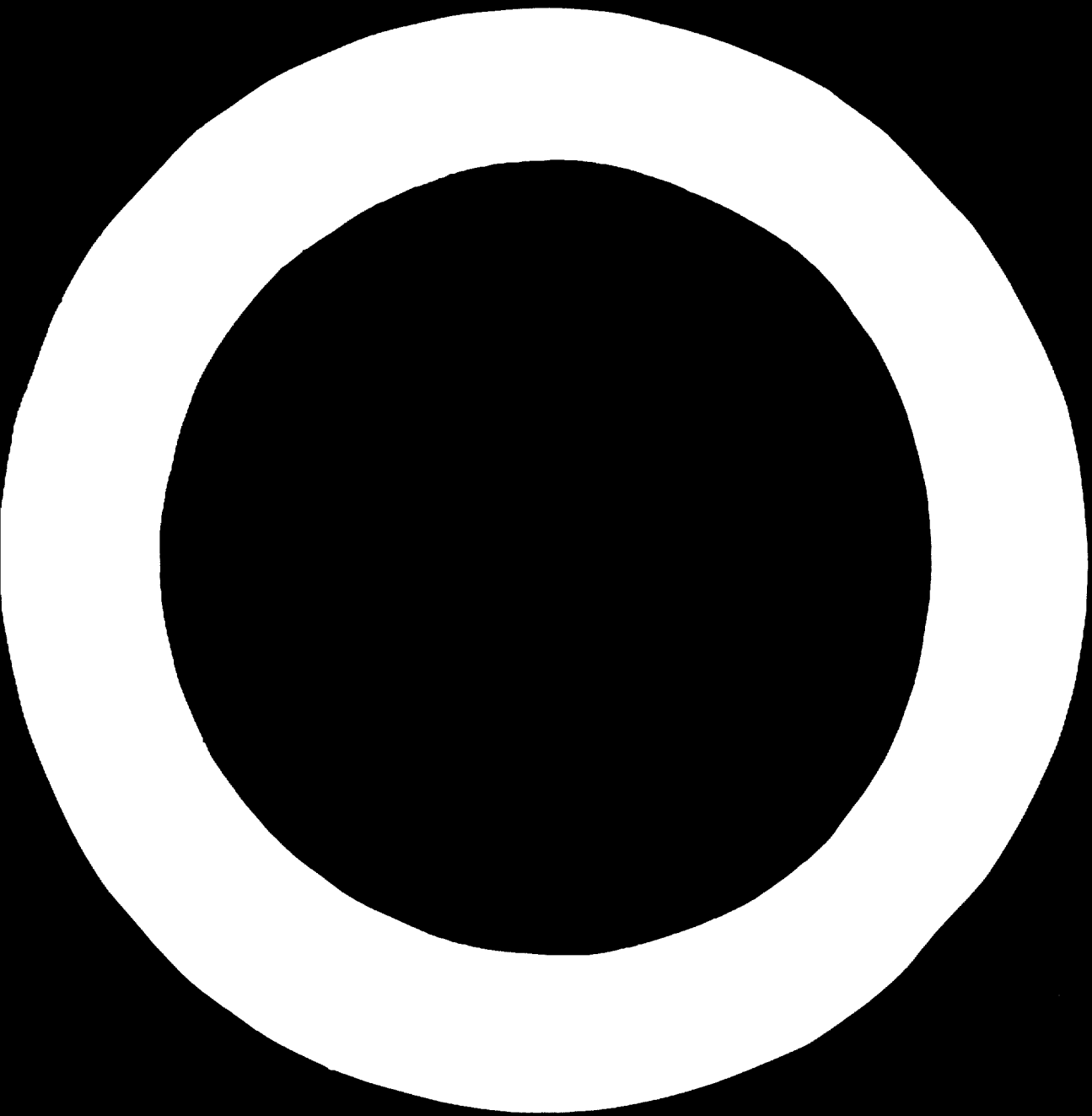
Figure 4

Bild 4. Einrichtung für Reduktionsversuche  
Figure 4. Equipment for reduction tests

- a) Waage — scale
- b) Thermoelement (Messung der Temperatur im Reduktionsraum) — thermocouple (measurement of temperature in reduction chamber)
- c) Reduktionszone (h = 400 mm) — reducing zone (h = 400 mm)
- d) Eisen(III)-oxyd-Pellets (30 mm Dmr.) — iron(III)-oxide pellets (30 mm dia.)
- e) Vorwärmszone (Kupferringe; h = 200 mm) — preheating zone (copper rings; h = 200 mm)
- f) Gasintritt — gas inlet
- g) Thermoelemente zur Steuerung der drei Ofenwicklungen — thermocouples for controlling the three furnace windings
- h) Gasaustritt — gas outlet

Figure 5 shows that hydrogen-rich gases reduce at a much higher rate than carbon monoxide. The highest rates of reduction have already been reached at 50 %  $H_2$ . The predominant influence of the temperature also on the speed of reaction with hydrogen is shown in Figure 6. Carbon monoxide also reduces more rapidly at higher temperatures. Increasing the gas pressure in the reaction vessel did not accelerate the reaction significantly (Fig. 7).





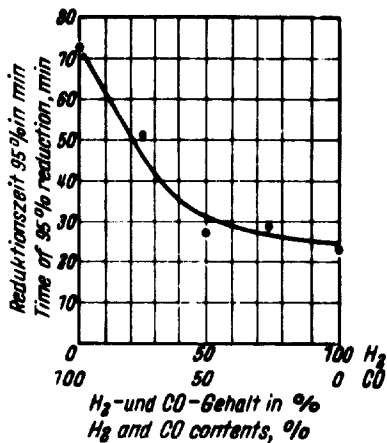


Bild 5. Reduktionszeit (95% Reduktionsgrad) in Abhängigkeit vom Wasserstoff- und Kohlenmonoxydgehalt des Gases.

Figure 5. Time of reduction (degree of reduction: 95%) as a function of hydrogen and carbon monoxide content of gas

Versuchsbedingungen: Einsatz: 400 g Eisen(III)-oxyd-Pellets; Schütthöhe: 50 mm; lichter Gefäßdurchmesser 70 mm; Temperatur: 900 °C; Strömung: 40 NI/min  
 — Test conditions: input: 400 g iron(III)-oxide pellets; height of bed: 50 mm; inside dia. of vessel 70 mm; temperature: 900 °C; flow: 40 NI per min

Figure 5

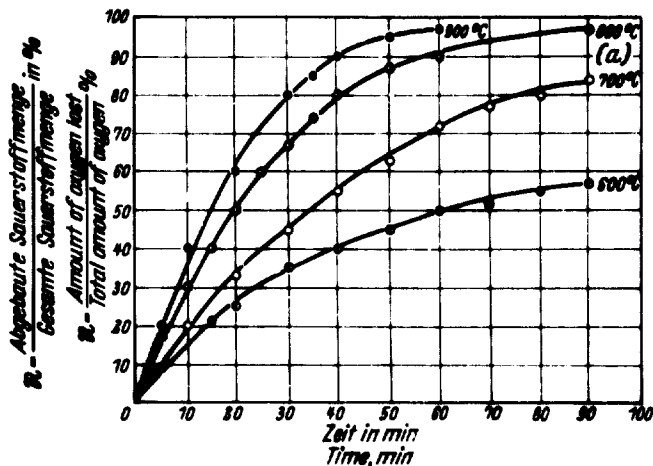


Bild 6. Reduktionsgrad in Abhängigkeit von Zeit und Temperatur. Isotherme Reduktion von Eisen(III)-oxyd-Pellets (30 mm Dmr.) mit Wasserstoff

Figure 6. Degree of reduction as a function of time and temperature. Isothermal reduction of iron(III)-oxide pellets (30 mm dia.) by hydrogen  
 a) Reduktionstemperatur — reduction temperature

Figure 6

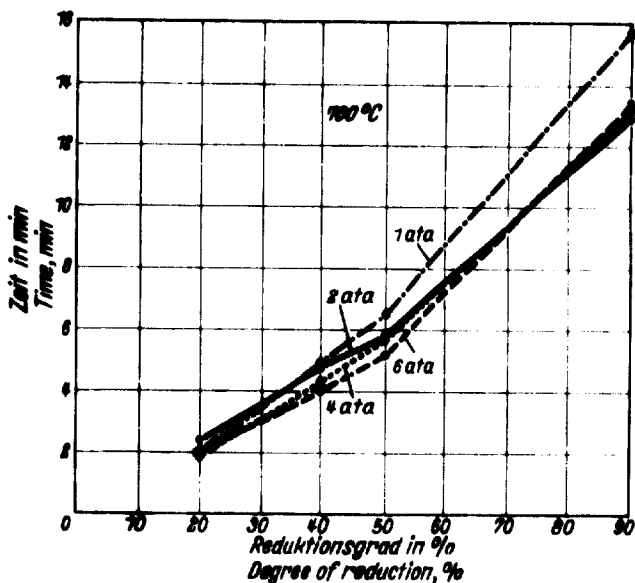


Bild 7. Reduktionsdauer für verschiedene Reduktionsgrade in Abhängigkeit vom Druck

Figure 7. Time of reduction for different degrees of reduction as a function of pressure

Reduktionsgas: Wasserstoff; Einsatz: Eisen(III)-oxyd-Pellets (3 bis 5 mm Dmr.) — reducing gas: hydrogen; input: iron(III)-oxide pellets (3–5 mm dia.)

Figure 7



It is proved by these results that favourable conditions for the industrial reducing process can be obtained by a maximum gas temperature and by hydrogen contents in excess of 50 %. An upper temperature limit is given only by softening of the ore. The stability of the partially and completely reduced ore is increased by raising the hydrogen content and lowering the carbon monoxide content of the reducing agents.

A limit lies at about 1000 to 1100°C. On the other hand an increase of pressure is not necessary, a fact which greatly simplifies the construction of large-scale plants.

A further question concerns the flow rate of the gas. The more gas per unit time which passes through the ore charge, the higher is the reduction rate; at the same time the consumption of gas per ton of iron is also increased, as less of the gas reacts with the charge. A transposable measure of the gas supplied per unit time, referred to unit weight of the charge, is the time taken for the gas to traverse the column; this is given by the quotient of the charge volume (empty tube) and the gas supply per unit time (referred to 0°C and normal pressure). Figure 8 shows the development of output and gas consumption/t Fe as a function of time of traverse of the gas, based on the laboratory experiments. It follows from the diagram that a relatively good throughput is possible for the minimum consumption of hydrogen with gas transit times of about 7 sec.; this time of transit is chosen as basis for the larger plants.

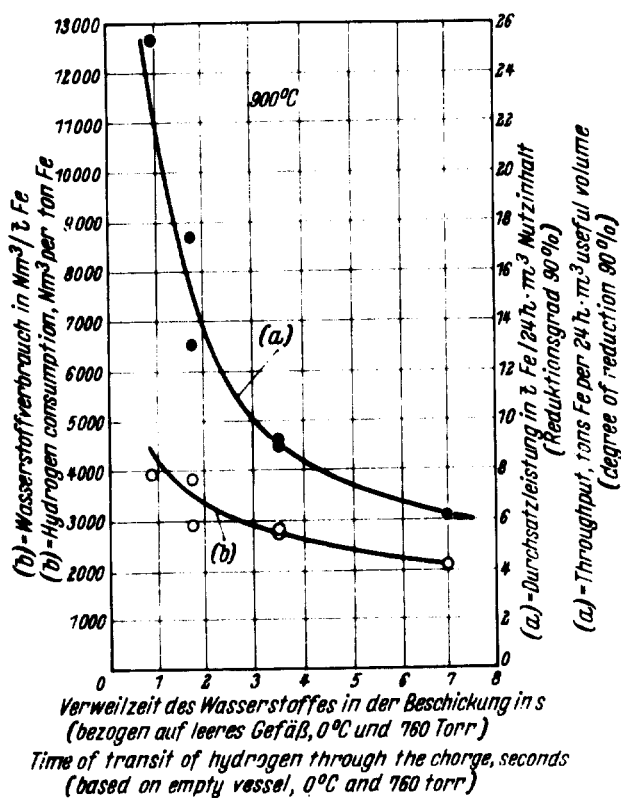
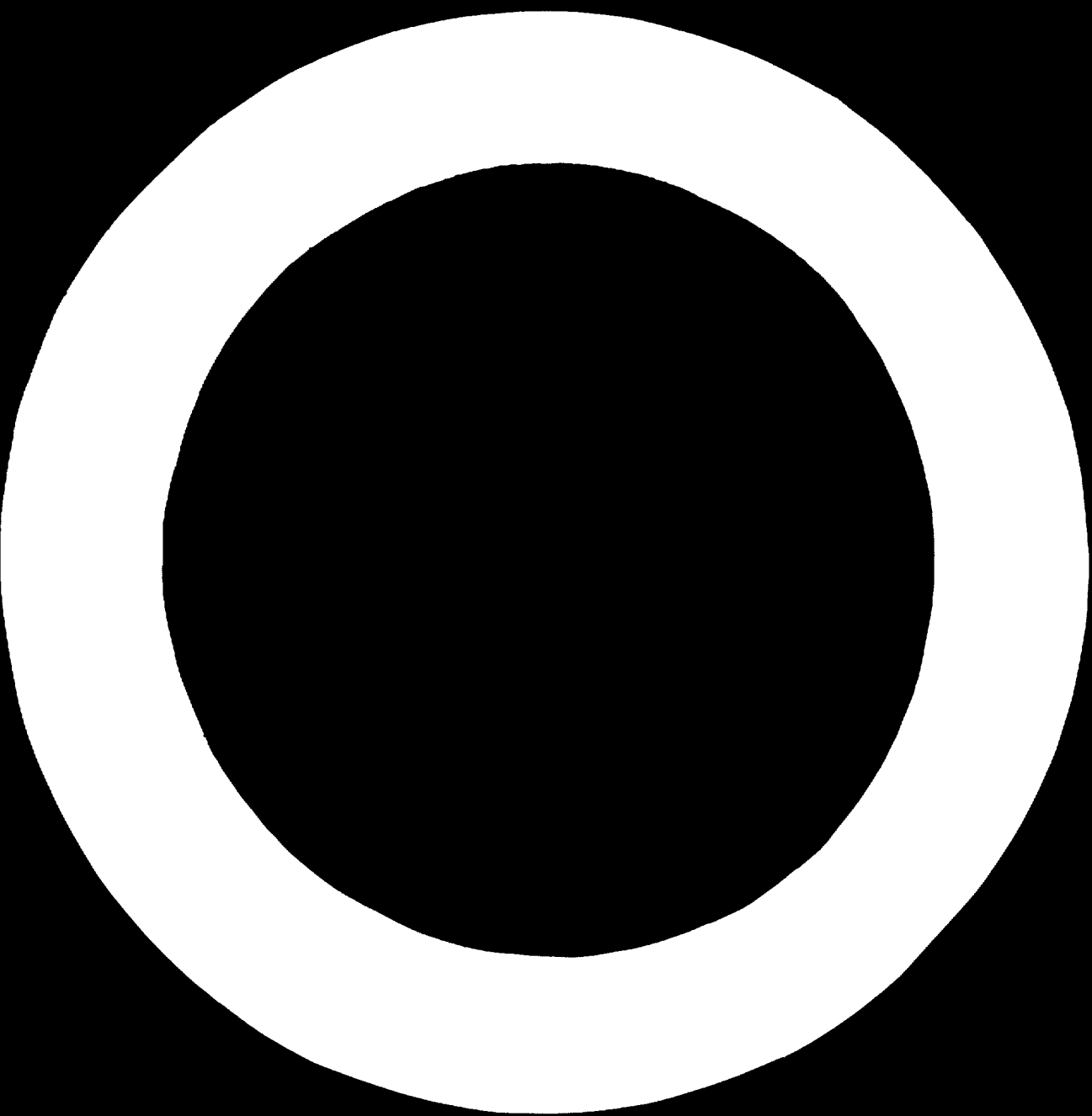


Figure 8

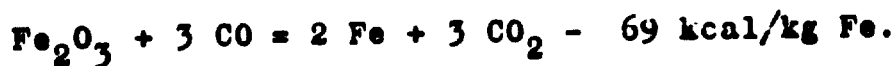
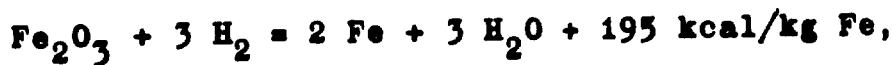
Bild 8. Leistung und Wasserstoffverbrauch eines isotherm geführten Reduktionsofens bei der Reduktion von Pellets (30 mm Dmr.)

Figure 8. Output and hydrogen consumption of an isothermally operating reduction furnace reducing pellets (30 mm dia.)

- a) Durchsatzleistung — throughput
- b) Wasserstoffverbrauch — hydrogen consumption



It is known that the reduction of iron (III)-oxide with hydrogen, in contrast to carbon monoxide, proceeds endothermically:



It must be noted, however, that the heat of reaction represents only a comparatively small proportion of the total heat consumption. Larger amounts of heat are necessary for heating the ore to the reducing temperature to vaporize the ore moisture, and to cover the radiation losses. The amounts of heat for reduction, vaporization, radiation and heating the charge must be covered from the sensible heat of the reducing gas. Figure 9 shows that hydrogen at 1000°C still contains sufficient heat, even with a low throughput of hydrogen per ton of iron, to supply the necessary amounts of heat; even then an excess of heat remains, which appears as a high top gas temperature. As shown by Figure 9, the conditions are less favourable when the hydrogen enters at a lower temperature, for example 600°C.

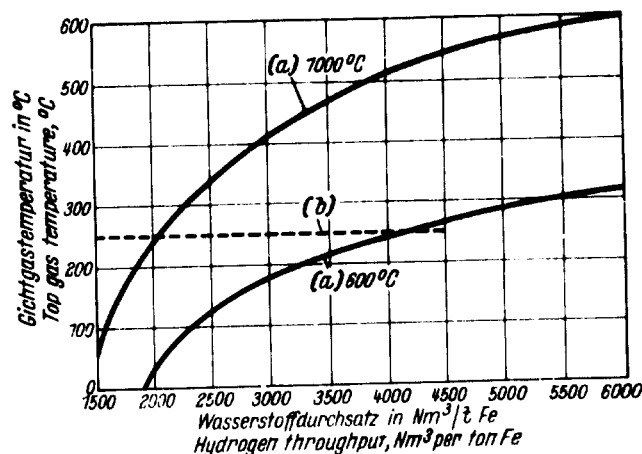
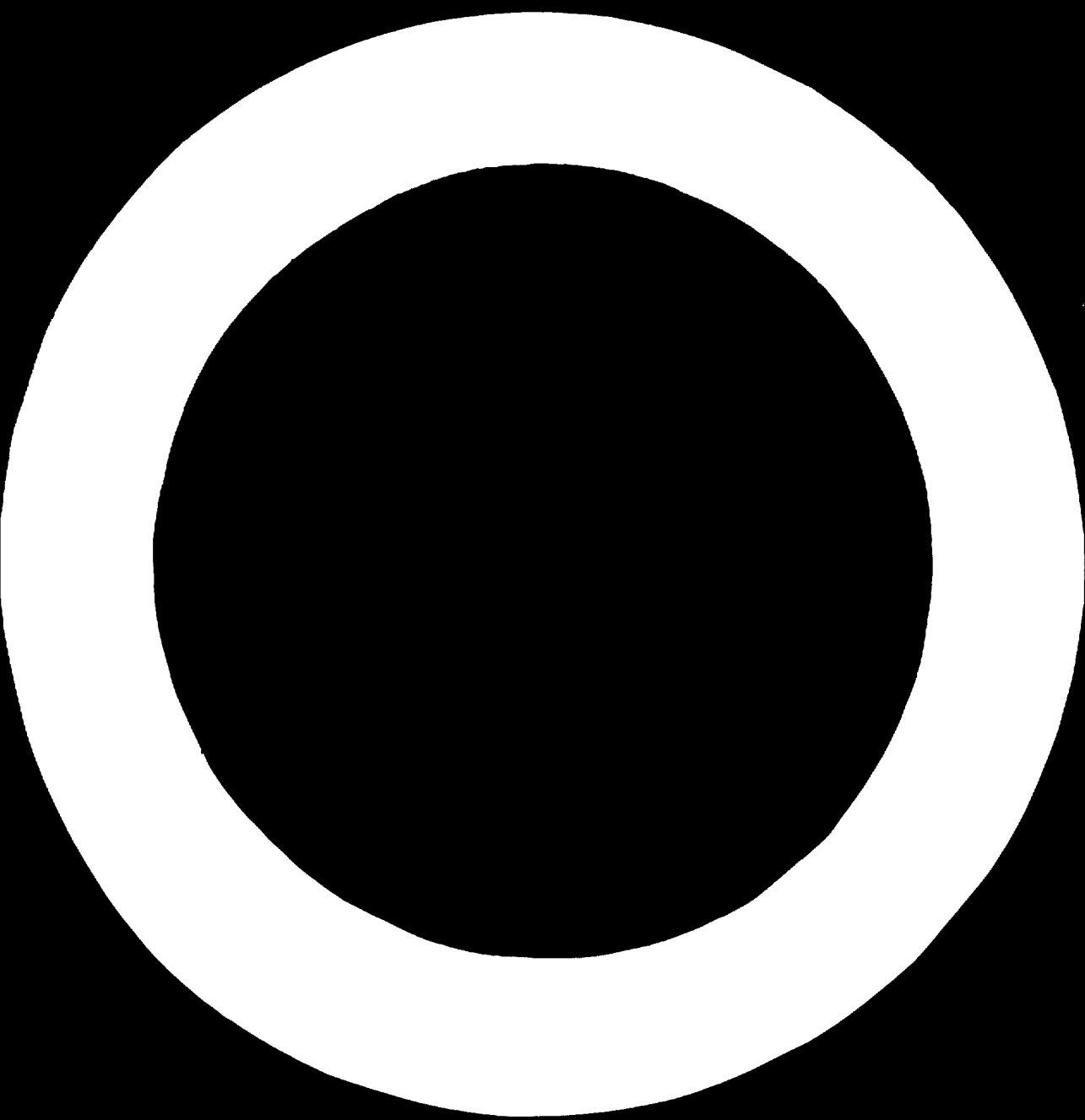


Bild 9. Theoretische Gichtgastemperatur in Abhängigkeit von Durchsatz und Eintrittstemperatur des Wasserstoffs (10% Abstrahlungsverluste, 3% Nässe im Erz. Erzreduktion nach  $\text{Fe}_2\text{O}_3 + 3 \text{H}_2 = 2 \text{Fe} + 3 \text{H}_2\text{O}$ )

Figure 9. Theoretical top gas temperature as a function of throughput and inlet temperature of hydrogen (10% radiation losses, 3% moisture in ore. Ore reduction according to  $\text{Fe}_2\text{O}_3 + 3 \text{H}_2 = 2 \text{Fe} + 3 \text{H}_2\text{O}$ )

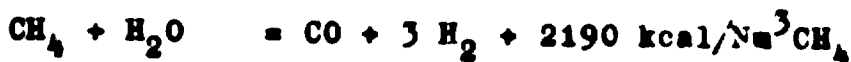
- a) Eintrittstemperatur — inlet temperature
- b) untere Grenze der Gichtgastemperaturen von Reduktionsanlagen — lower limit of top gas temperatures of reduction plants

Figure 9



Principles underlying the transformation of methane into mixtures of hydrogen and carbon monoxide

This transformation proceeds in accordance with the following equations:



At temperatures above 800°C the equilibrium at normal pressure lies completely to the right. A difficulty of these reforming reactions is the formation of soot. The equilibrium diagram (Fig. 10) shows that high temperatures and a high hydrogen/carbon monoxide ratio are suitable for repressing soot formation; but this cannot be prevented altogether, as the reducing gas is cooled by the reduction.

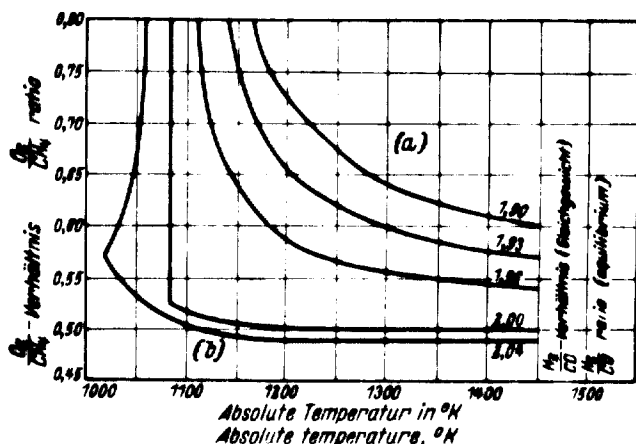


Figure 10

Bild 10. Temperaturabhängigkeit des Verhältnisses von O<sub>2</sub>:CH<sub>4</sub> bei verschiedenen Verhältnissen von H<sub>2</sub>:CO im Spaltgas (Gesamtdruck: 760 Torr)

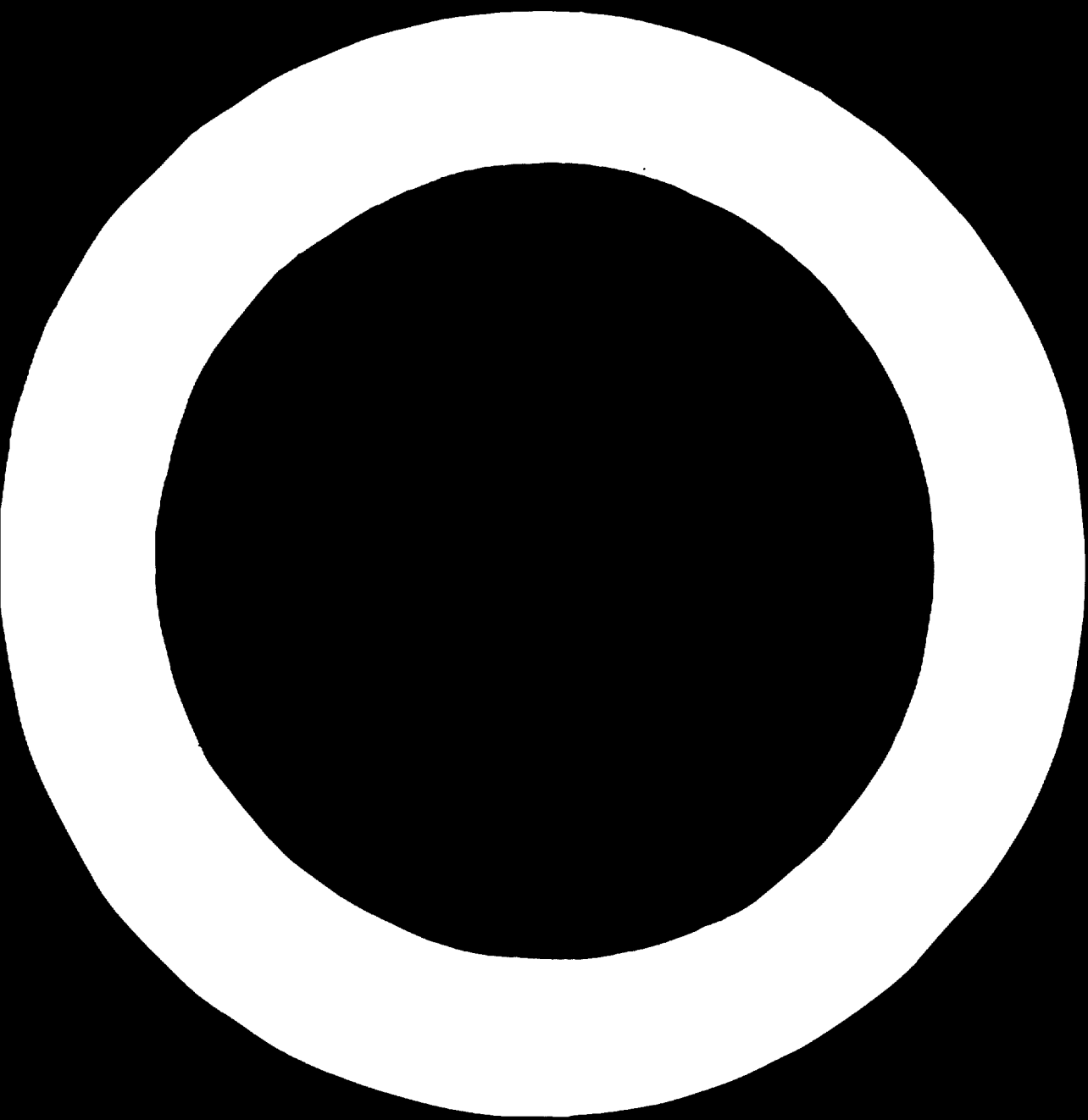
Figure 10. Temperature dependence of the ratio of O<sub>2</sub>:CH<sub>4</sub> given various ratios of H<sub>2</sub>:CO in reformed gas (total pressure: 760 mm Hg)

- a) keine Kohlenstoffabscheidung - no carbon precipitation
- b) Kohlenstoffabscheidung - carbon precipitation

Our own investigation in the laboratory was directed to the problem of reforming methane in such a way that a mixture of hydrogen and carbon monoxide low in carbon dioxide and steam is formed in a single step at 1000 to 1100°C, which is immediately suitable for reduction. So far these conditions have not been fulfilled by the known processes.

The chemical industry has developed and uses several processes for methane reforming to produce synthesis gas, all based on the above mentioned reactions mostly with nickel





catalysts. These processes are characterized by a surplus of reforming agent to get as low as possible a content of  $\text{CH}_4$  which would disturb the following synthesis reactions. Thus the reformed gas contains appreciable amounts of  $\text{CO}_2$  and  $\text{H}_2\text{O}$  which have to be washed out, before the gas can be used for the reduction of iron ore.

None of these reforming processes seemed useful to us, as it is necessary to undertake the following complicated heat-treating of the gases:

- a) heating and reforming  $> 800^\circ\text{C}$ ,
- b) cooling, washing of  $\text{CO}_2$  and  $\text{H}_2\text{O}$
- c) reheating to  $> 800^\circ\text{C}$  for the reduction.

The investigation was carried out in the equipment shown in Figure 11. Here, methane-containing gases from coke oven gas, to pure methane, with sulphur contents between 50 to  $500 \text{ mg/Nm}^3$ , were reformed by oxygen, carbon dioxide and steam. The reaction tube was charged with different types of filling materials and catalysts; the emerging gaseous products were analysed in the Orsat apparatus, the water vapour and soot contents collected in suitable filters, and determined by weighing. After numerous unsuccessful trials a catalyst was found which gave the desired gas composition at  $1000^\circ\text{C}$ , i.e. a mixture of hydrogen and carbon monoxide at  $1000^\circ\text{C}$  that is suitable for the reducing process without further treatment. This catalyst was found to be insensitive to sulphur, as well as to frequent alternations between oxidizing and reducing atmospheres. In contrast to the known processes these properties enable semi- and full-scale plants to be greatly simplified, thereby reducing the capital costs.

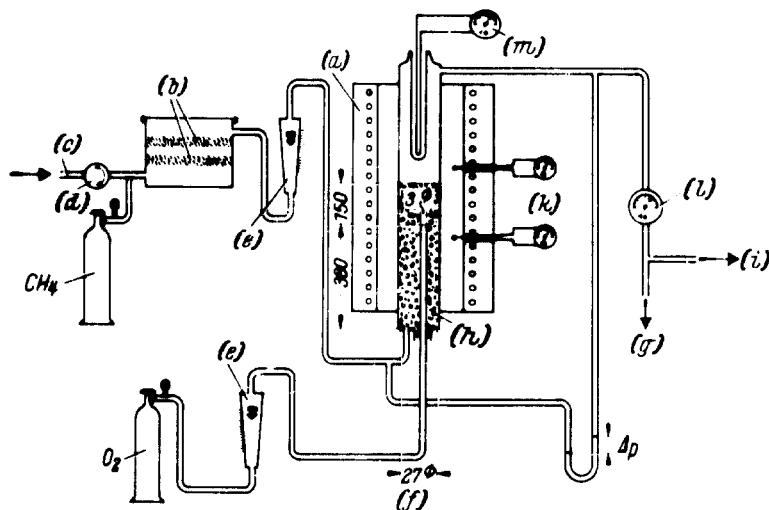
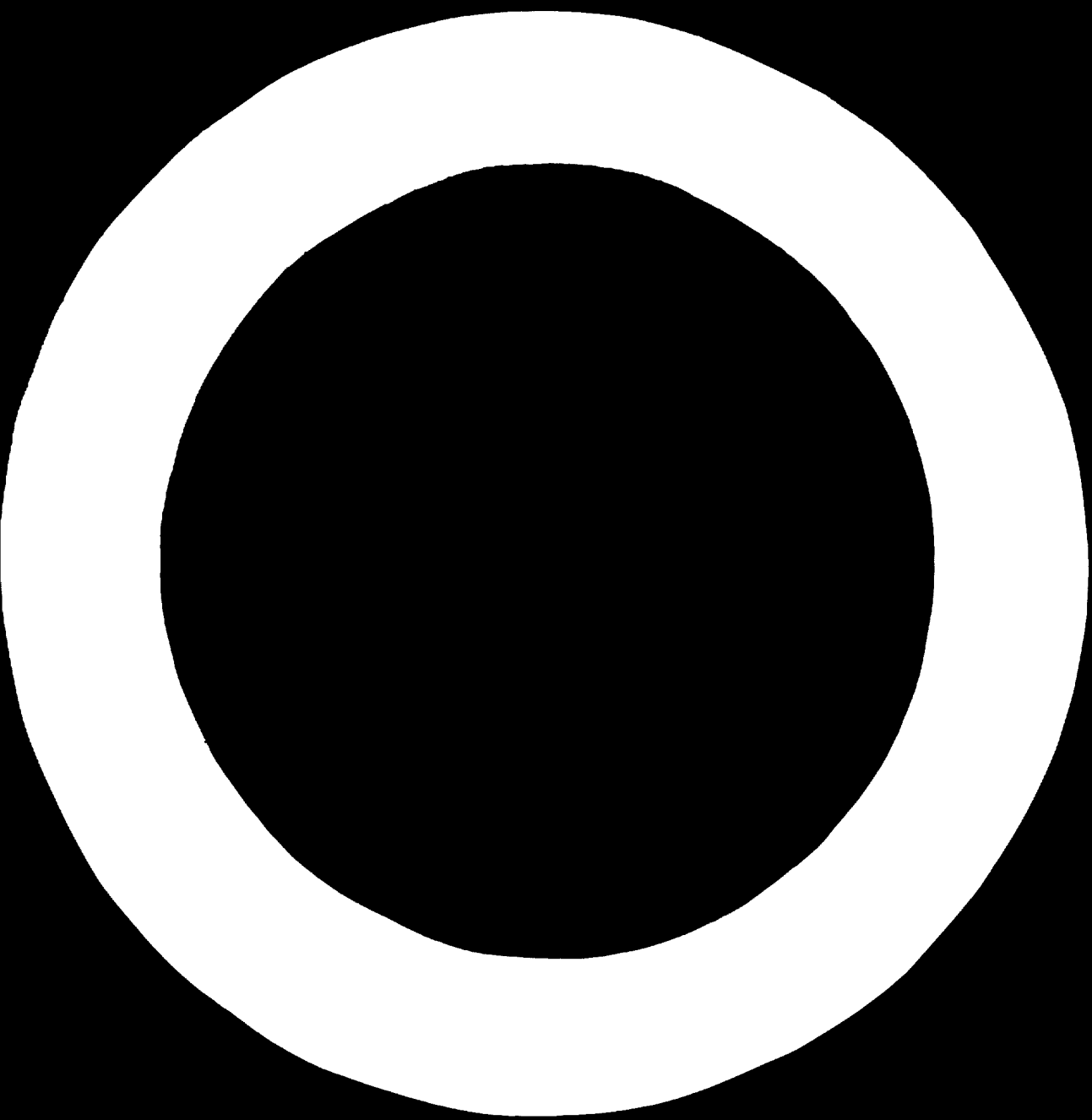


Figure 11

Bild 11. Versuchseinrichtung für die Gasaufbereitung  
Figure 11. Laboratory equipment for gas preparation

- |   |  |
|---|--|
| a) Widerstandsofen — resistance furnace                     | g) Untersuchung der chemischen Zusammensetzung — chemical analysis |
| b) Entschwefelungsmasse — desulphurisation mass             | h) Katalysator 3 bis 5 mm — catalyst size 3 to 5 mm                |
| c) Koksöfengas oder Restgas — coke-oven gas or residual gas | i) Austritt — outlet   |
| d) Gebläse — blast  | k) Regler — control  |
| e) Rotameter — rotameter                                    | l) Gasuhr — gasmeter   |
| f) Durchmesser (innen) — inside diameter                    | m) Gastemperatur — gas temperature                                 |



#### 4. Construction of the pilot plant

The laboratory experiments have established the most important operating conditions i.e.: decomposition of the natural gas with oxygen or steam at the catalyst at 1000 or 1100°C, with the production without further treatment of a hydrogen-carbon monoxide mixture suitable for reduction, containing more than 50 % H<sub>2</sub>; reduction to commence at 1000 to 1100°C at normal pressure; time of transit of the gas through the ore charge about 7 sec. The process is divided into a gas reformer stage and an ore reducing stage, with operating conditions which have been laid down in some detail. Considerable latitude still remains however in the design of industrial plant, which must be narrowed down by further experiment and consideration, in order to find the best solution.

#### 4a. Construction of the gas reformer

The reforming of the natural gas into carbon monoxide and hydrogen takes place at the catalyst at 1000 to 1100°C. As the catalyst had proved to be sufficiently resistant to alternations of reducing and oxidizing atmospheres, it could also be used simultaneously as a regenerative heat carrier. The catalyst is first heated to about 1200°C; then the oxygen-methane or steam-methane mixture is passed in through a pre-mixing chamber and is reformed to a hydrogen-carbon monoxide mixture at about 1000 to 1100°C. The temperature of the catalyst falls gradually to 1000°C, and is then reheated. By installing two alternately operating gas reactors an uninterrupted production of the desired mixture at the correct temperature range is ensured. The waste gas from the reducing furnace is used for heating. Figure 12 shows details of the gas reformer for the pilot plant. The soot problem is also solved by the interplay between reforming and catalyst reheating. Each deposit of soot which forms during the reforming period is burnt during reheating; in this way it is both removed and utilized as a source of heat.

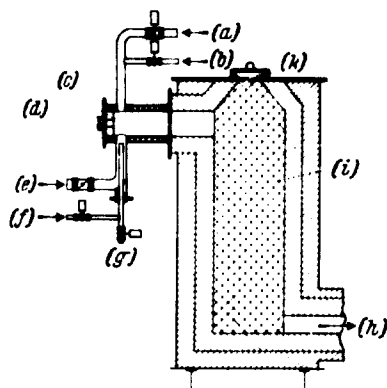
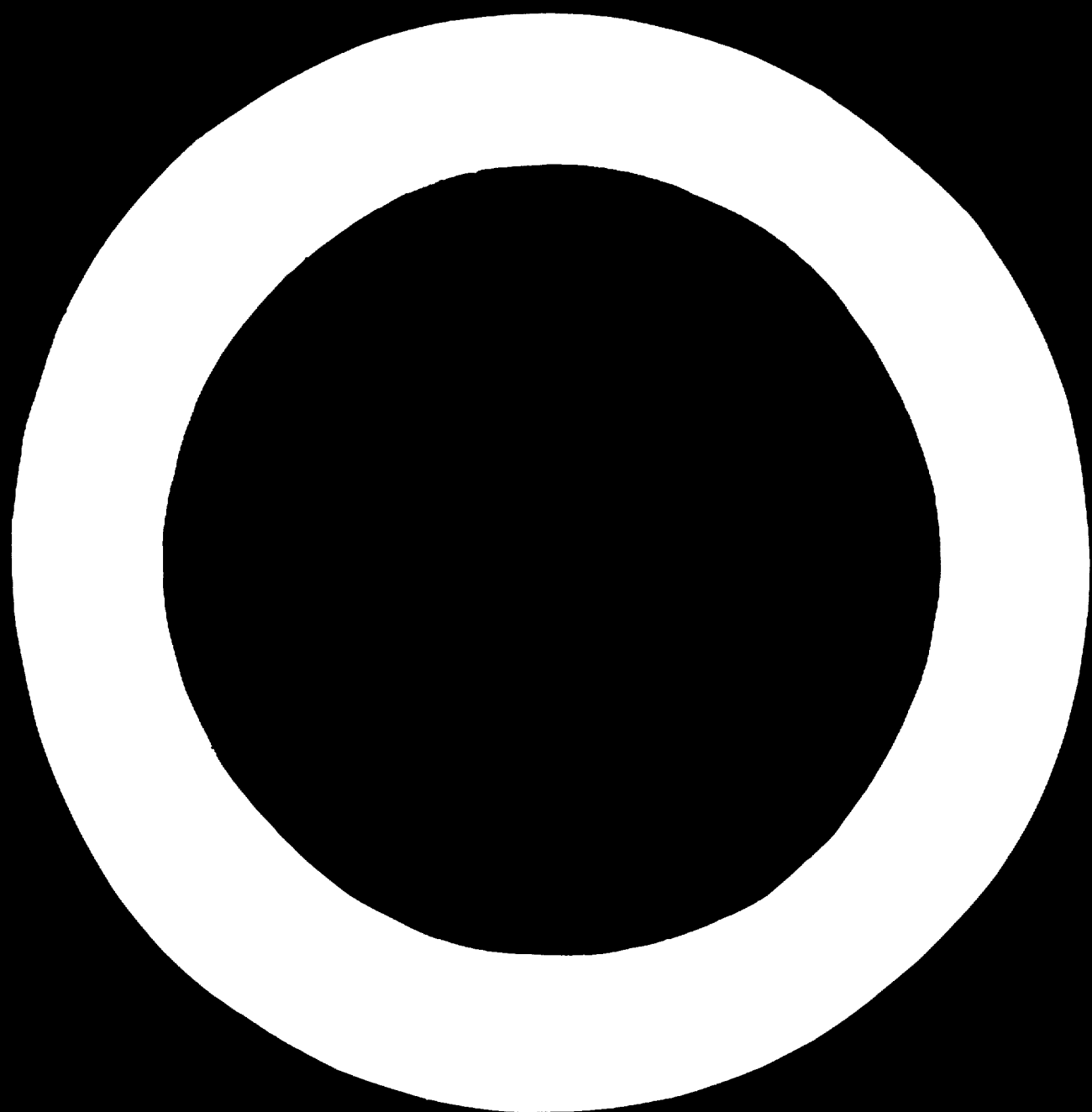


Figure 12

Bild 12. Schema des Gasumsetzers  
Figure 12. Cross-section of gas reformer

- a) Gichtgas vom Reduktionsschachtofen -- top gas from reduction shaft furnace
- b) methanhaltiges Gas -- methane gas
- c) Zyklonbrenner -- cyclone burner
- d) Schauloch -- peepsight
- e) Gebläseluft -- air blast
- f) Wasserdampf -- steam
- g) Sauerstoff -- oxygen
- h) Reduktionsgas, Kohlenmonoxyd-Wasserstoff (1000 bis 1100 °C) -- reducing gas, carbon monoxide hydrogen (1000 to 1100 °C)
- i) ...



#### 4b. Construction of the reduction furnace

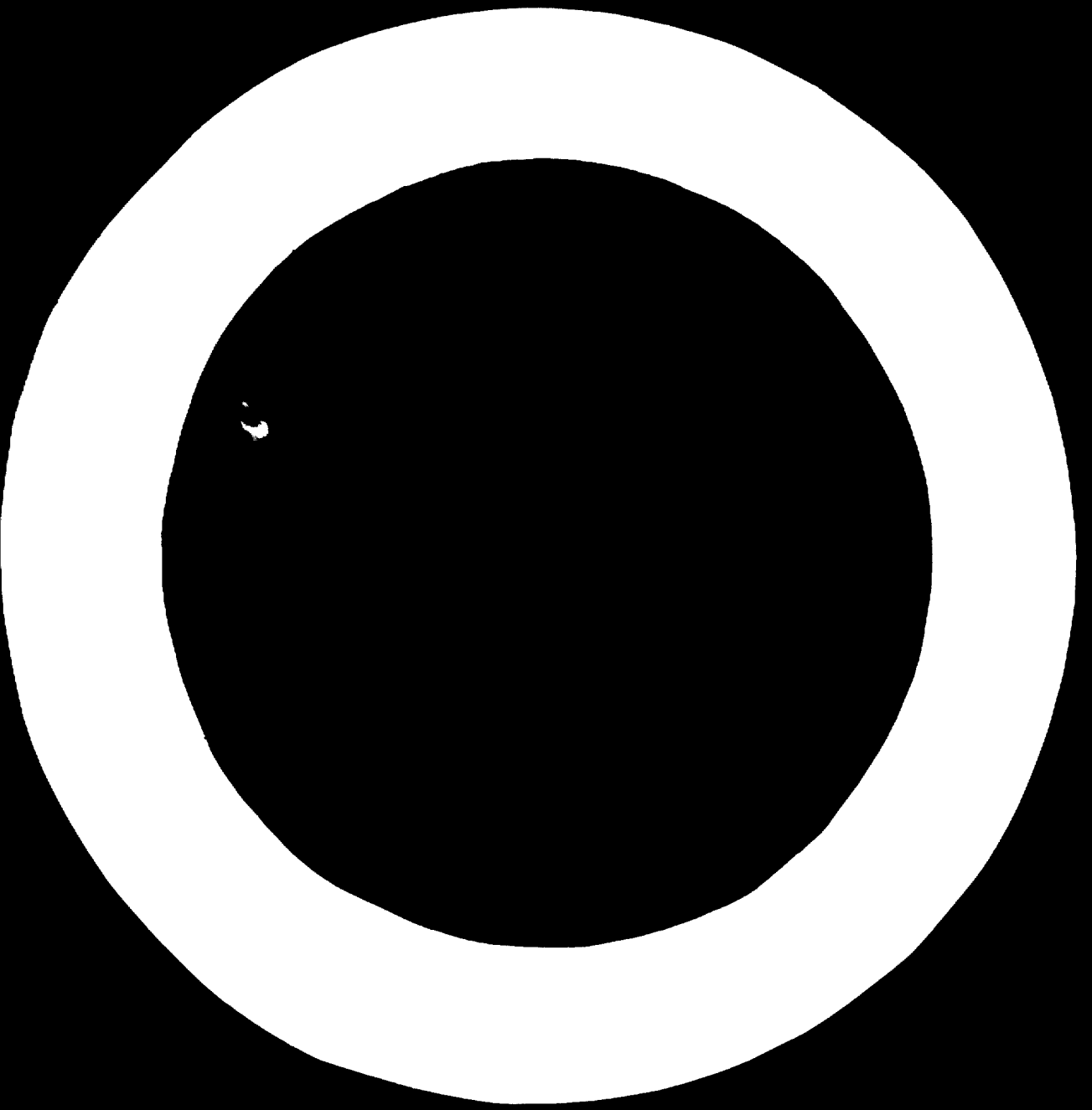
It has already been shown that sponge iron processes with a lump ore charge are superior to fluidized-bed processes operating with finely-divided ores, in respect of throughput and treatment of the reduced product. But several technical possibilities exist even with lump ore, such as retorts, travelling grate, rotary container, and shaft furnace. All these possibilities were experimentally investigated, the travelling grate and the shaft furnace being tried out on the semi-industrial scale. The final selection was the shaft furnace. With this unit the highest throughputs per m<sup>3</sup> of effective volume can be attained and by using the counterflow principle, the best possible thermal and chemical utilization of the reducing gases, comparable with those in the blast furnace, is achieved.

Over and above this, the shaft furnace has the advantage that many of the structural characteristics which have proved satisfactory in the classical blast furnace can be adopted. Figure 13 shows details of the reduction shaft as constructed for the pilot plant.

The ore enters from the top through a double-bell seal into the shaft, and travels gradually downwards, thereby being heated to temperature and reduced. At the lower end of the column the finished sponge iron is discharged by special equipment and ejected into a drum. The drum is cooled from outside with water. The sponge iron, cooled under a reducing atmosphere, is discharged at the end of the drum. Figure 14 gives a general view of the pilot plant in its present form.

The first pilot plant was put into operation early in 1959. The gas reformer was already in its present form; reduction was conducted on a travelling grate. This plant ran until the end of 1960 with only brief interruptions, and produced 700 kg of sponge iron per day. Subsequently the travelling grate was replaced by the shaft furnace. On the 8th of February, 1961, the pilot plant started up with the reduction shaft, and now produced 2500 kg sponge iron per day with the same supply of gas in unit time from the unmodified gas reformers. The superiority of the counterflow principle in the shaft furnace is clearly evident from these results.

Apart from charging, all the operations of the plant are controlled automatically. An illuminated circuit diagram shows the condition of the plant at any time (Fig. 15). The gas reformers are mechanized and so is the discharge of the reduced material from the shaft into the cooling drum and its ejection from the drum.



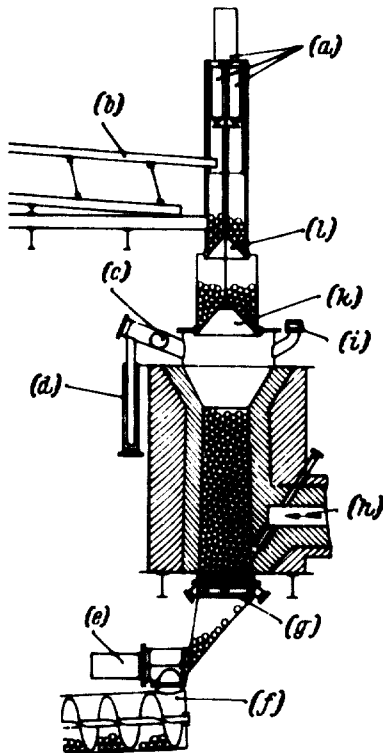


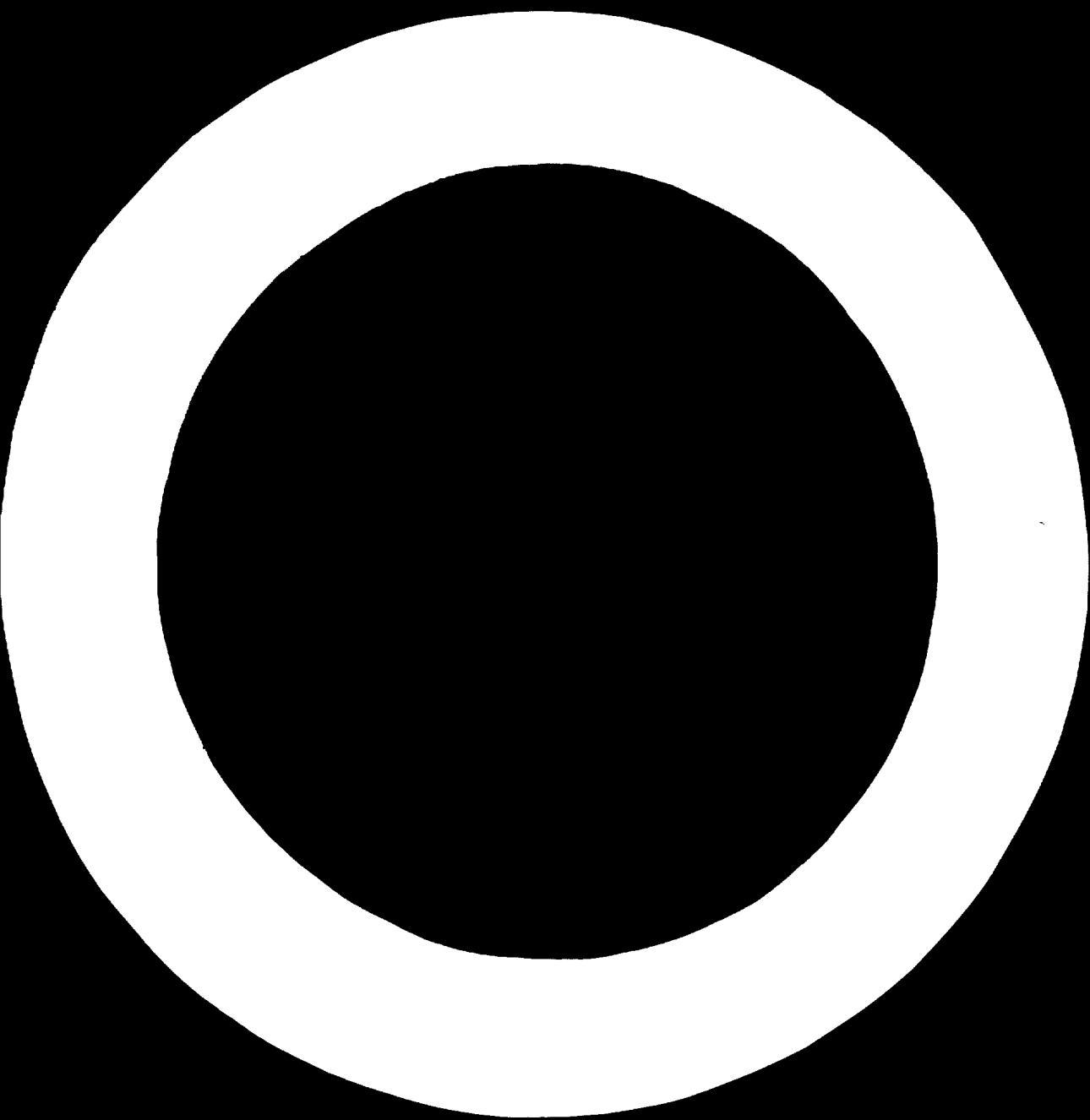
Figure 13

- Bild 13. Reduktionsschachtofen  
 Figure 13. Reduction shaft furnace
- a) Antriebszylinder — driving cylinder
  - b) Schwingrinne — feeder vibrator
  - c) Austritt des Gichtgases — outlet for top gas
  - d) Wassertasse — water seal
  - e) Plattenverschluß — gate
  - f) Kühltrommel — cooling drum
  - g) Austrag — discharge
  - h) Zufuhr des Reduktionsgases (1000 bis 1100 °C) — supply of reducing gas (1000 to 1100 °C)
  - i) Explosionsklappe — safety valve
  - k) Unterglocke — lower bell
  - l) Oberglocke — upper bell



Bild 14. Gesamtansicht der halbtechnischen Reduktionsanlage  
 Figure 14. Overall view of pilot reduction plant





5

5

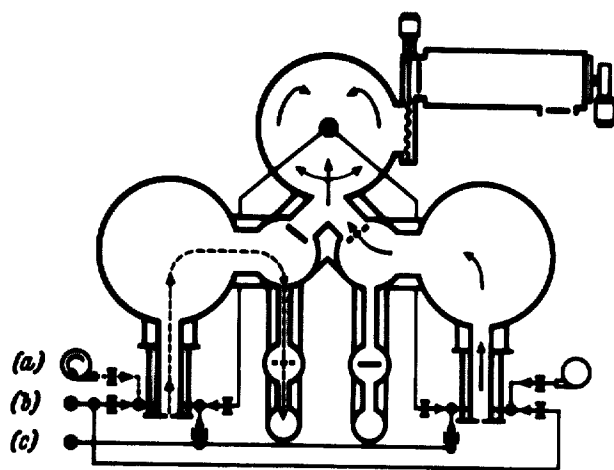


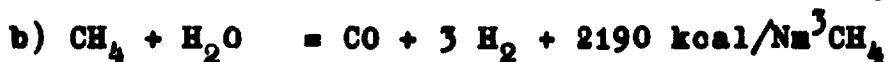
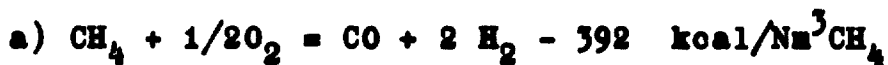
Bild 15. Schaltchema der Reduktionsanlage  
 Figure 15. Switch layout of reduction plant  
 a) Brenniuft - combustion air b) Sauerstoff - oxygen  
 c) Frischgas - fresh gas

Figure 15

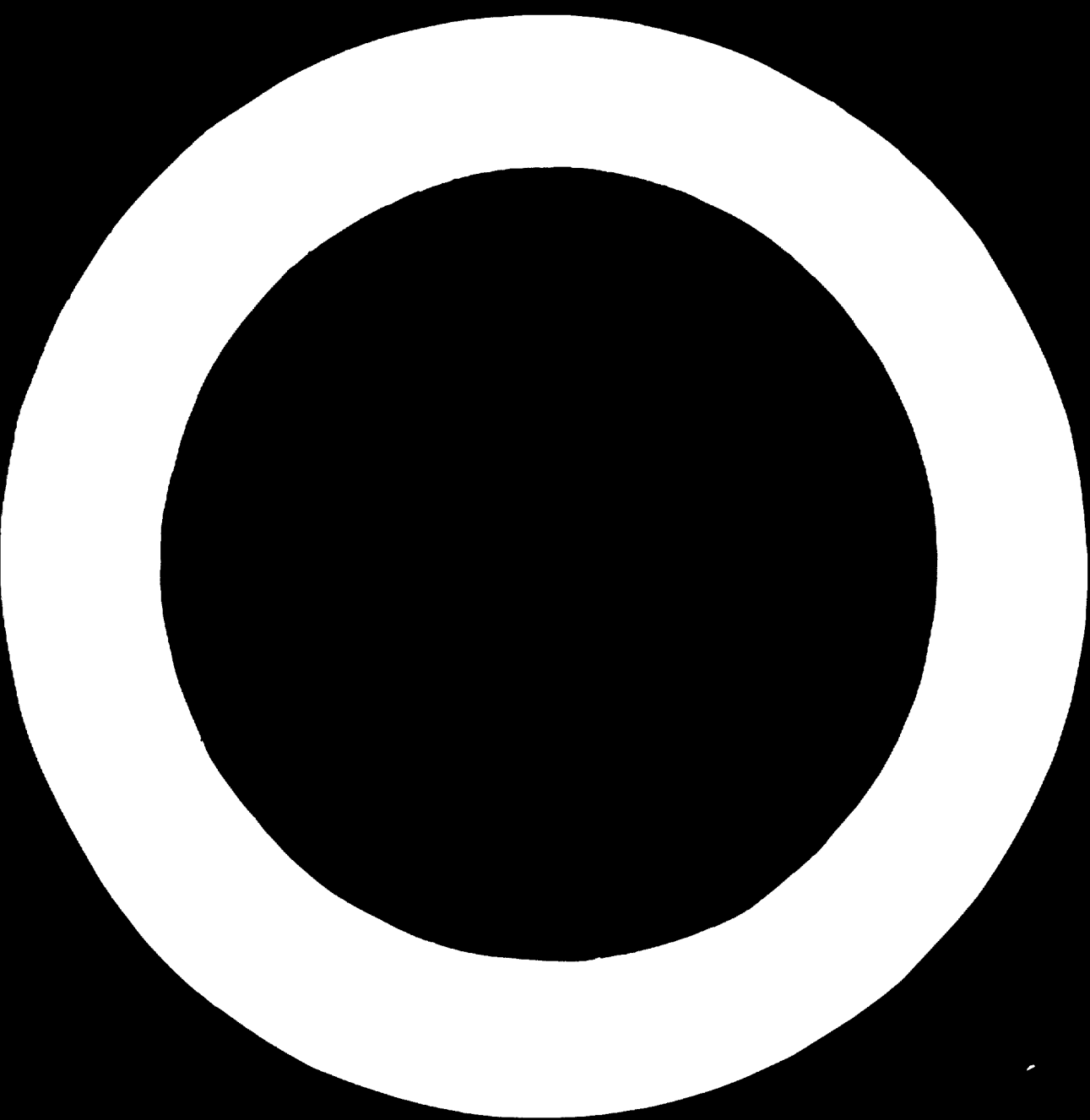
## 5. Results of experimental section

### 5a. Gas reforming

The manufacture of reducing gas is based on the reactions:



In reaction a) only 3 Nm<sup>3</sup> CO-H<sub>2</sub> mixture is produced from each Nm<sup>3</sup> CH<sub>4</sub>, and oxygen is required as a reforming agent; at the same time comparatively little heat is used. Air can be used instead of oxygen, but in this case the reducing gas contains about 45 % N<sub>2</sub>, which is a disadvantage for the subsequent use of the gas. Reforming with the cheaper steam provides 4 Nm<sup>3</sup> CO-H<sub>2</sub> mixture per Nm<sup>3</sup> methane, but the amount of heat required is several times greater. Both oxygen and steam were tested in the pilot gas reformers, and the reactions proceeded satisfactorily in each case. The following fuels were used:



	CO <sub>2</sub>	C <sub>n</sub> H <sub>m</sub>	CO	H <sub>2</sub>	CH <sub>4</sub>	N <sub>2</sub>	net calorific value, kcal/Nm <sup>3</sup>
	%	%	%	%	%	%	
Coke oven gas	1,6	2,4	4,8	58,3	26,6	6,0	4329
Residual gas (from a chemical plant)	0,9	1,9	11,4	9,8	54,1	21,0	5545
Natural gas	0,6	0,0	0,0	0,8	97,8	0,8	8406

Reforming proceeded satisfactorily throughout the year on 24-hour operation, as can be seen from the following gas compositions (monthly average values).

The gas was always mixed in exactly stoichiometric, i.e. CH<sub>4</sub> : H<sub>2</sub>O = 1 : 1 and CH<sub>4</sub> : O<sub>2</sub> = 2 : 1.

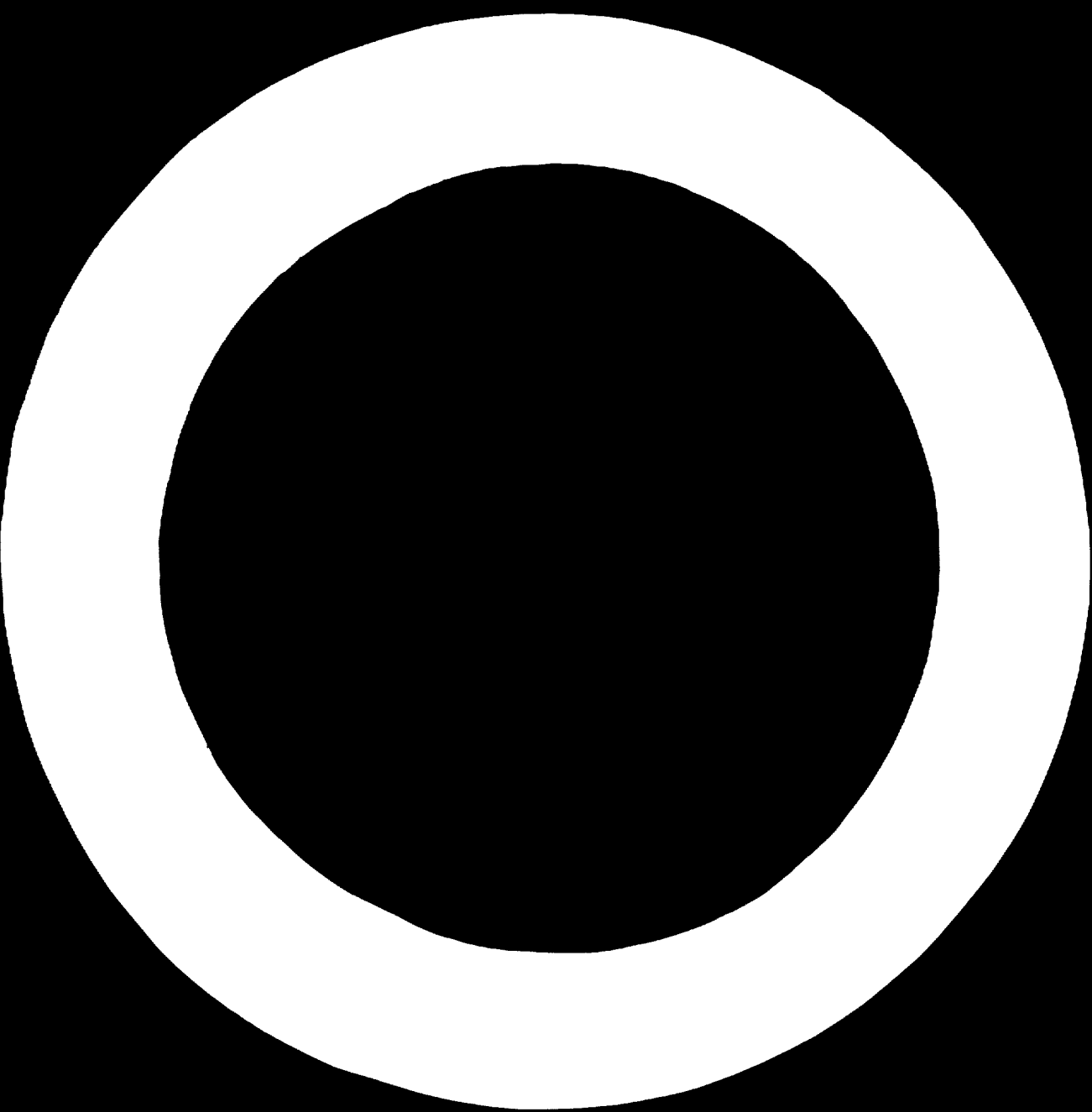
Reformed gas from:	CO <sub>2</sub>	CO	H <sub>2</sub>	CH <sub>4</sub>	N <sub>2</sub>
	%	%	%	%	%
Residual gas + O <sub>2</sub>	0,8	26,7	58,1	1,8	11,8
Natural gas + O <sub>2</sub>	1,7	25,4	63,1	4,7	4,8

The steam content was less than 25 g/Nm<sup>3</sup>. The temperature of the reformed gas lay between 1050 and 1100°C. In normal operation residual gas and oxygen were used, as these media were always available in the plant. Natural gas had to be obtained in containers of 1000 Nm<sup>3</sup> capacity. The ratio of hydrogen to carbon in the reformed gas did not correspond exactly to the crude gas charged. This is accounted for by the formation of soot: however, the soot does not interfere with the operation of the plant, and it is unnecessary to remove it. The soot-containing reducing gas permeates the charge, and most of it is finally used to reheat the gas reformers. In this way the heat content of the soot is fully utilized.

To investigate the sensitivity of the catalyst to sulphur, coke oven gas containing about 500 mg S/Nm<sup>3</sup> was charged experimentally for several weeks. As was expected the nickel-free catalyst was not damaged by this treatment.

#### 5b. Reduction

Iron (III)-oxide pellets and Venezuela ore with the following compositions were used as experimental ores:



	Fe	SiO <sub>2</sub>	P	combined water	moisture
	%	%	%	%	%
Iron (III)-oxide Pellets	68,9	0,57	0,019	0,0	0,30
Venezuela ore	64,8	1,37	0,11	5,49	7,89

Figure 16 gives survey of the reducibility of these materials. The rate of reduction in hydrogen at 800°C is plotted as a function of lump size. The pellets, about 30 mm in diameter, are relatively difficult to reduce. In long term trials extending over a month, it was possible to show, in spite of this fact that reduction was satisfactory, with high utilization of the reducing gas. The size of Venezuela ore lay between 6 and 100 mm.

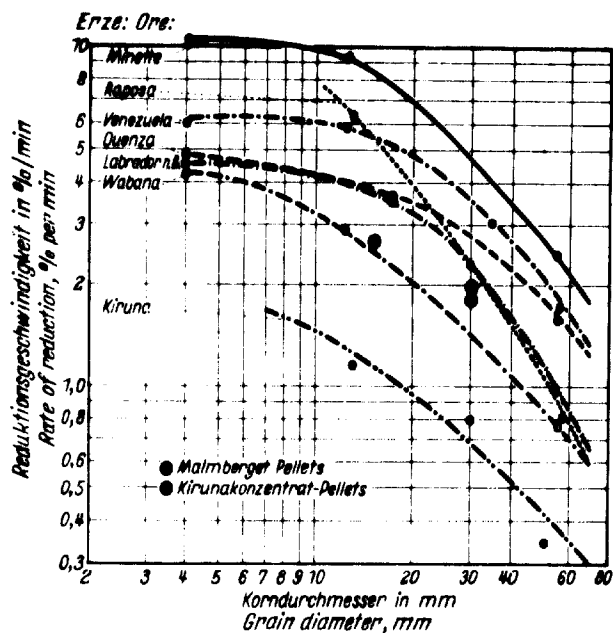
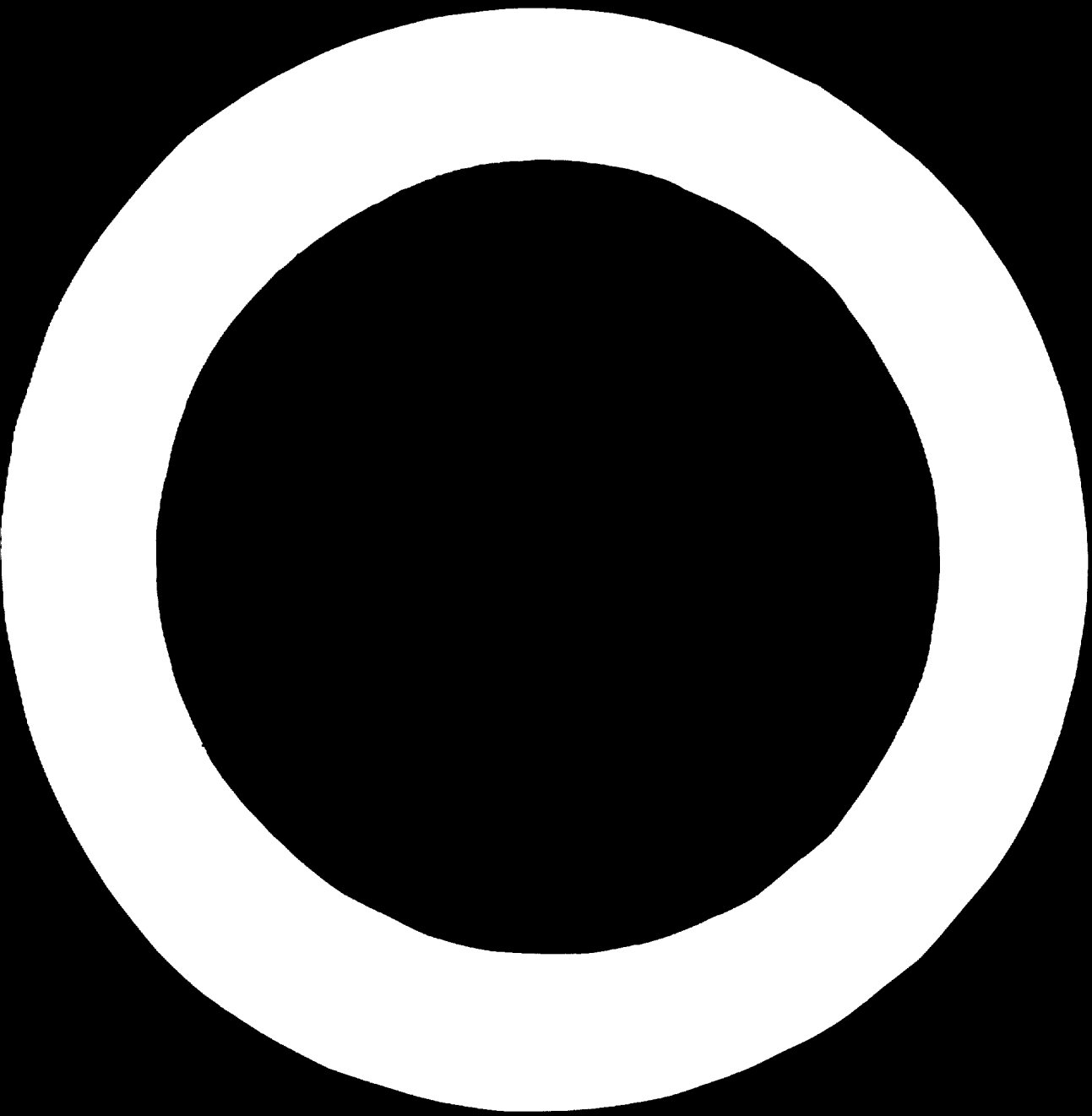


Bild 16. Zusammenhang zwischen Stückgröße und Reduktionsgeschwindigkeit bei Erzen  
Figure 16. Relationship between lump size and rate of reduction of ores

Figure 16



The monthly average composition of the reduced ore was as follows: total Fe 96,0 %, metallic Fe 91,2 %, C 1,40 %, P 0,015 % and S 0,012 %. The external form of the sponge iron corresponded to that of the charged ore.

Production on average was 2453 kg Fe/24 h with an effective furnace content of 0,66 m<sup>3</sup> (tuyere plane to the upper edge of the charge). The specific gas throughput of the furnace was 2570 Nm<sup>3</sup> CO + H<sub>2</sub>/t Fe, i.e. the chemical utilization of the gas amounted to 23,4 %. The non-utilized portion of the gas is partly burnt to reheat the gas reformers; an excess of heat still remains however, equal to about 25 % of the total quantity of top gas in the small pilot plant. This proportion will increase in larger plants due to lower specific heat radiation and preheating of the combustion air. The top gas not required for combustion was blown off in the pilot plant, as recovery would have been uneconomical. With larger plants the gas would be compressed and recirculated through the operation gas reformer to give additional reducing gas. Complete purging of the reaction products carbon dioxide and steam from the circulating gas is not necessary, as these constituents can be used with advantage as a reforming agent for the methane of the crude gas, thereby reducing the amount of oxygen required.

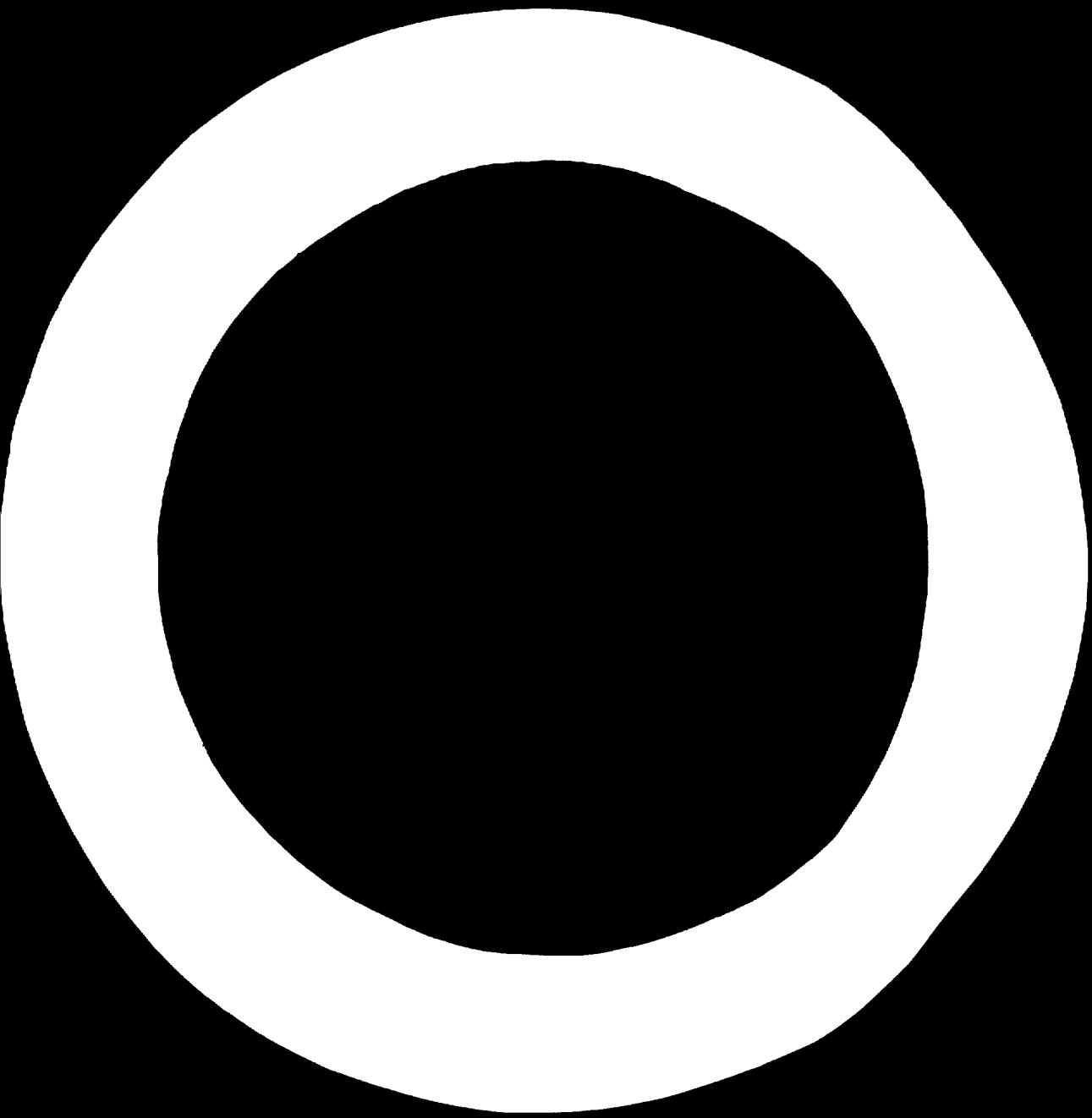
The total production of the pilot plant was around 2000 metric tons of sponge iron.

## 6. Use of the sponge iron produced

Sponge iron is a high-grade charge material for steelmaking processes, for example for electric furnaces, open hearth furnaces and it is especially suitable as a cooling agent for pneumatic steel processes. Sponge iron replaces scrap, and also has the advantage of being more convenient to charge, and of having very low contents of the undesirable trace elements, such as arsenic, copper, chromium and nickel, which unfortunately are almost always present in the circulating scrap.

Our trials confirmed that sponge iron can readily be melted down. A special technique in rotating furnaces seems very suitable for this melting work to produce liquid pig iron from the sponge which can be converted to steel in the well-known pneumatic processes. The details will be published elsewhere. At the present cost of iron carriers, the manufacture of sponge iron is an economic proposition for the steelworks, provided that the cost of heat from the gas does not exceed half that derived from coke (each being referred to the net calorific value). In the Federal German Republic the cost of heat from gas is too expensive for the production of sponge



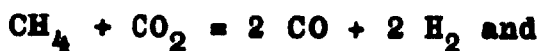


iron, both at present and in the foreseeable future. However, the relationships of energy costs are different in countries with a large supply of natural gas; in many cases heat from gas costs only a small fraction of heat from coke. In these countries the manufacture of sponge iron as a raw material for steel would seem to have great potentialities.

In order to cover the costs of the Oberhausen experimental plants, most of the sponge iron produced was processed to iron powder. To do this the following steps were necessary: decarburizing anneal, crushing, screening, and in some cases a special anneal to improve the quality of the sponge iron. It would go beyond the scope of this paper to explain these relationships in detail; up to the present several 100 tons of iron powder have been produced by this method and sold.

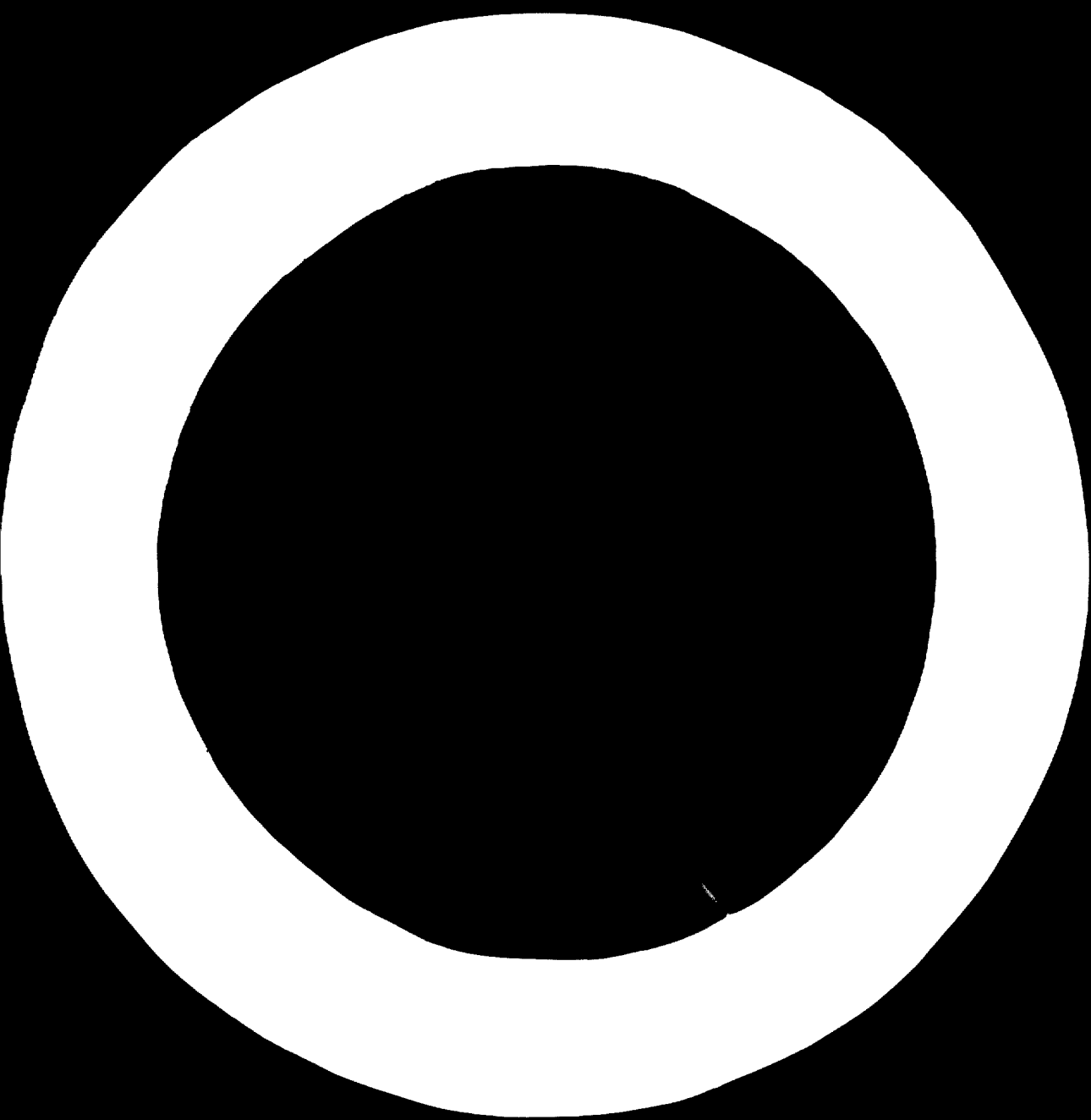
## 7. Transformation to the industrial scale

The manufacturing schedule of a large-scale plant is derived from the results of the pilot plant (Fig. 17). The sketch shows gas reformer I in operation and reformer II in the reheating position. The fresh reducing gas, heated to 1000 to 1100°C, passes up the shaft in counterflow to the ore. The used reducing gas is divided; part is used for heating the gas reformer, the rest (circulating gas) is fed back into the gas reforming system. About half the reaction steam is washed out; finally the gas is compressed. A small part is injected below as cooling gas into the discharge system of the reduction shaft. The remainder is returned to the gas reformer, together with fresh natural gas and reforming agent. The carbon dioxide and steam contents of the circulating gas act as a reforming agent in accordance with the reactions:

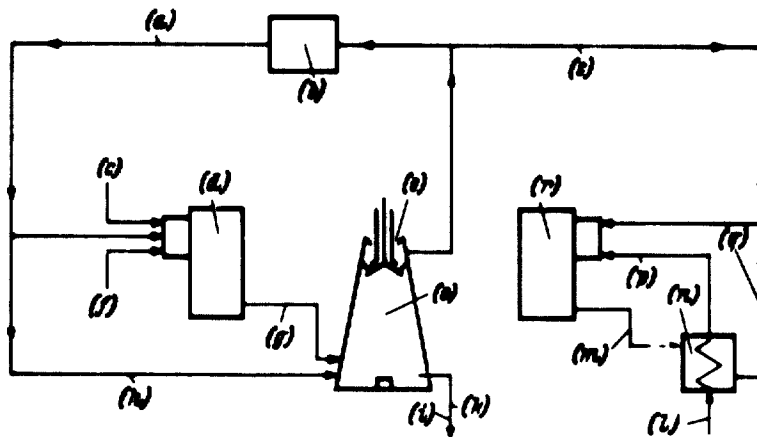


The consumption of fresh oxygen or steam is thereby reduced. The process schedule is basically the same for oxygen and steam; only the large-scale layout of the gas reformer is changed.

Since the beginning of 1960 work has continued on the transfer of the reduction process to the industrial scale; as a result plans for large-scale plants are now in existence. The outer appearance of these plants is very similar to that of the blast furnace. However, a much higher throughput is achieved size for size, as hydrogen is a stronger reducing agent than the usual carbon monoxide in the blast furnace.



A plant now being under construction is shown in figure 18. The daily output of this semi-industrial furnace will be about 25 tons.



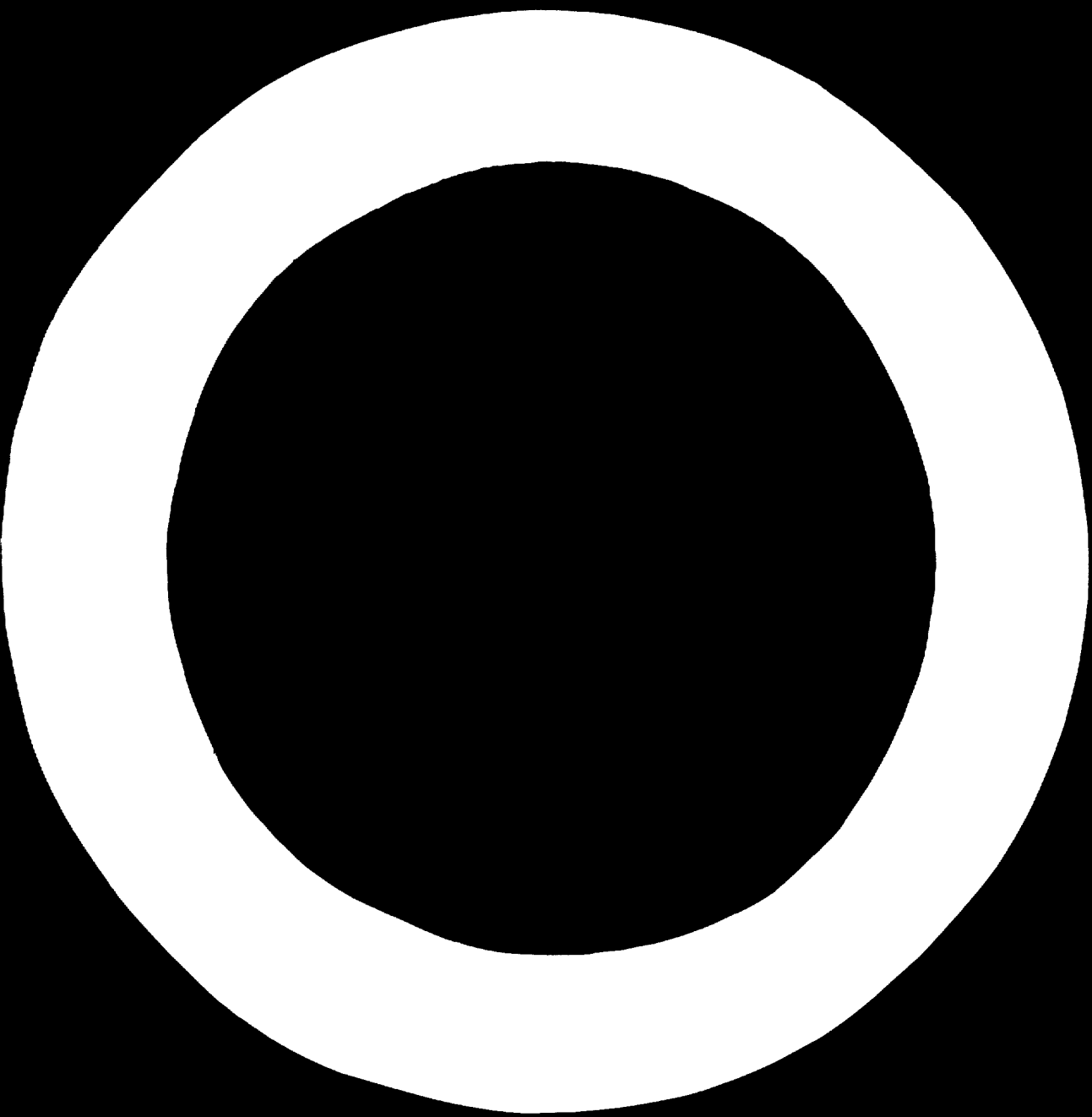
Die Spaltöfen I und II arbeiten abwechselnd — The reformer furnaces I and II work alternately

Bild 17. Fließbild einer betrieblichen Reduktions-Anlage mit Reduktionsgas-Erzeugung aus Erdgas und Sauerstoff

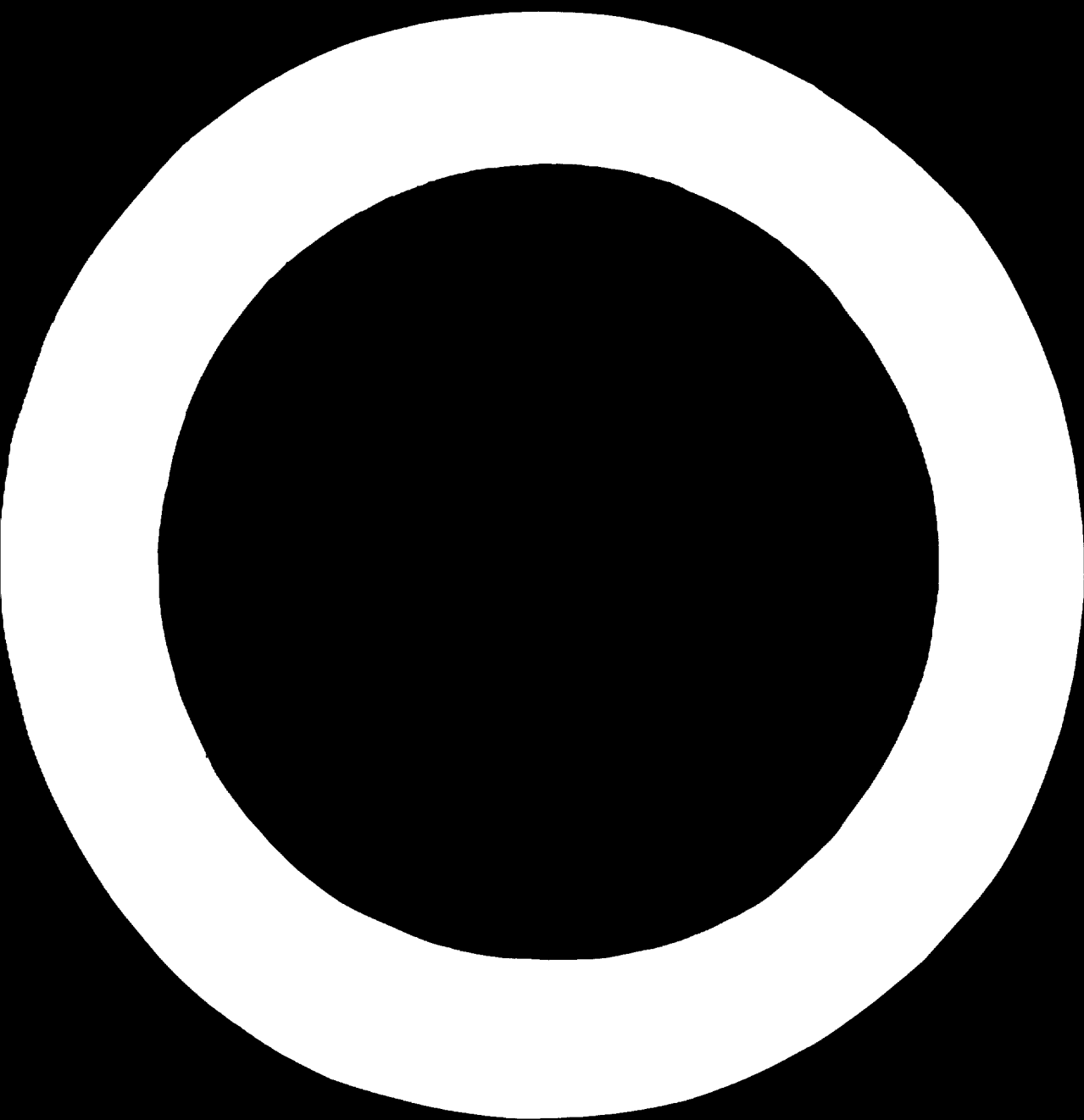
Figure 17. Flow diagram of an industrial reduction plant with production of reducing gas from natural gas and oxygen

- |  |   |
|--|---|
| a) rückgeführtes Dichtgas — recovered top gas  | l) Eisenschwamm (97% Fe, 2% C; 300 °C) — sponge iron (97% Fe, 2% C; 300 °C)   |
| b) Kondensation des Reaktionwasserdampfes auf 35 °C (Taupunkt) und Verdichten — condensation of reaction water vapour to 35 °C (dew-point) and compression | k) zur Kühltrommel — to cooling drum  |
| c) Methan ( $H_u = 8880 \text{ kcal/Nm}^3$ ) — methane (calorific value = 8880 kcal/Nm <sup>3</sup> )  | l) Wind (Druck 300 mm W.S.) — blast (pressure 300 mm w.g.)  |
| d) Spaltöfen I — reformer furnace I  | m) Abgas (1000 °C) — waste gas (1000 °C)  |
| e) Einsatz: Erz ( $\text{Fe}_2\text{O}_3$ ) — input: ore ( $\text{Fe}_2\text{O}_3$ )   | n) Rekuperator — recuperator  |
| f) Sauerstoff — oxygen   | o) Reduktionssochacht — reduction shaft   |
| g) Reduktionsgas (33% CO, 67% H <sub>2</sub> ; 1000 °C) — reducing gas (33% CO, 67% H <sub>2</sub> ; 1000 °C)  | q) Abgas (500 °C) — waste gas (500 °C)  |
| h) Kältegas — cooling gas  | p) Heißwind (600 °C) — hot blast (600 °C)   |
|  | r) Spaltöfen II — reformer furnace II   |
|  | s) Dichtgas zur Verbrennung (300 °C; $H_u = 2140 \text{ kcal/Nm}^3$ ) — top gas for combustion (300 °C; calorific value = 2140 kcal/Nm <sup>3</sup> ) |

Figure 17







### 3. Consideration of capital costs and operating costs

The detailed constructional work, based on the pilot plant results, and on experience with the erection of the semi-industrial plant, shows that the investment costs of a 500 tpd Purofer plant will not exceed those of a comparable blast furnace of the same capacity. In this comparison the oxygen plant is included.

But capital costs are not the decisive factor however. The economic superiority of the new process is due mainly to the fuel costs.

There are many countries in South America and the Near East without large supplies of coking coal, but where considerable reserves of natural gas and ore deposits are available. In these countries large quantities of natural gas are set free, which cannot be fully utilized by the incompletely developed economy, and must to a large extent be burnt to waste. As this loss cannot be carried permanently, the countries producing natural gas on a large scale are consequently trying to replace coke by this gas, so as to be able to transform indigenous and mostly high-grade ores into iron, and thereby build an economic foundation for their own steelmaking industry.

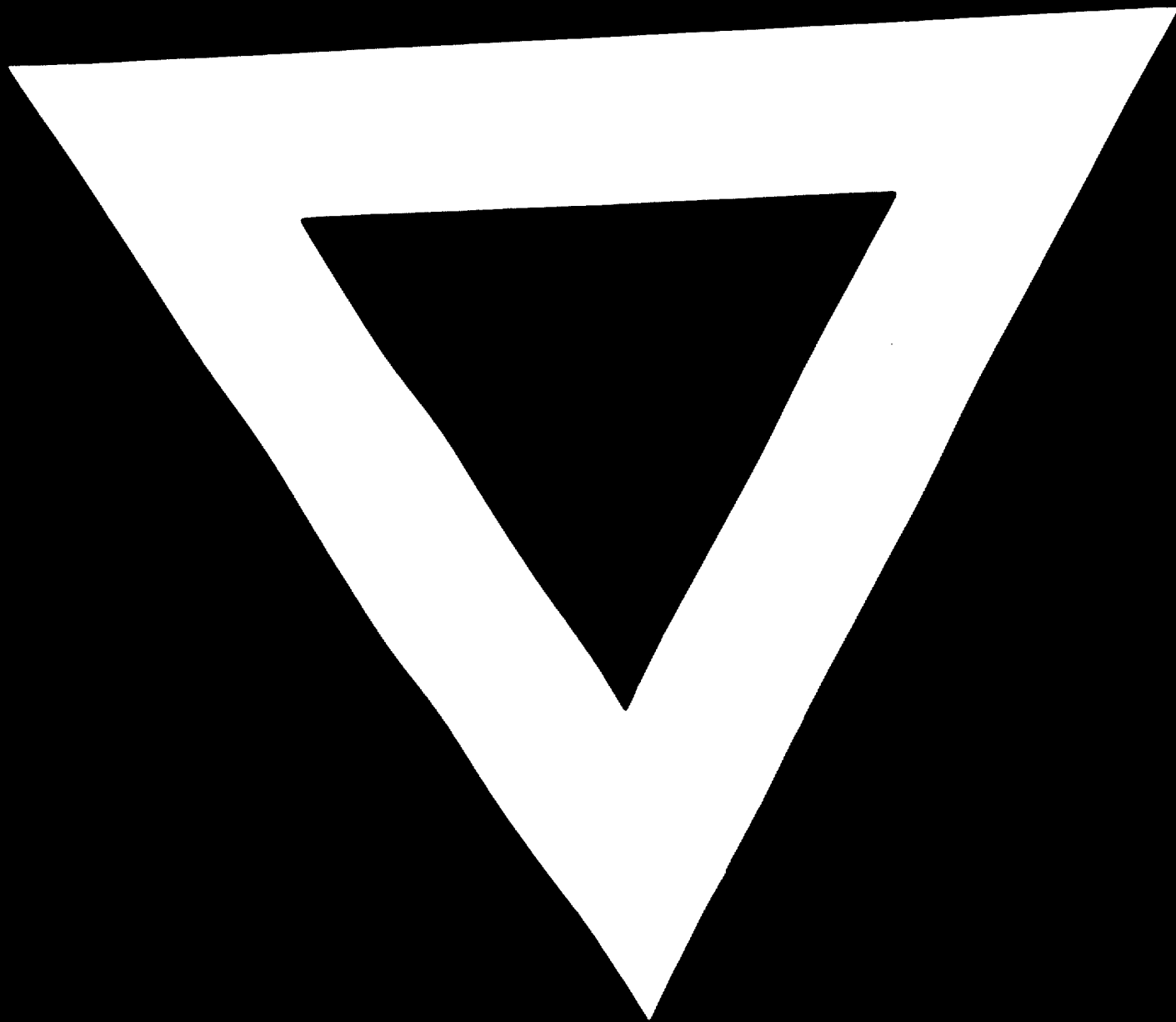
Coke is very expensive in the developing countries, as most of it has to be produced from imported coal. On the other hand natural gas costs less than 0.5 cts/Nm<sup>3</sup>.

As the standard of comparison to assess the economics of the Purofer process pig iron manufacture was selected, because pig iron is the usual raw material for further iron and steel production in countries with plentiful supplies of ore and a shortage of scrap. There are several possibilities of melting down sponge with suitable equipment under natural gas to make synthetic pig iron. An economic comparison between iron manufactured in the traditional blast furnace and synthetic pig iron from sponge shows, that operating costs mainly depend on fuel costs. They arrive

- 2.50 US\$/ton of pig iron in combined sponge-pig iron plants
- 17.50 US\$/ton of pig-iron in the blast furnace

(assuming that the blast furnace is located in a coal importing country). Such a start could not be overtaken. The synthetic pig iron will cost about 25 US\$/ton, which would be about 15 US\$/ton below the cost for pig iron made in the blast furnace in the same locality. The results so far obtained present a comparatively favourable picture for countries rich in natural gas, in respect both of capital and operating costs.





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