

Metallized Burden Preparation

MONDAY MORNING, NOVEMBER 29, 1965

The session on Metallized Burden Preparation convened at 9:00 am. Chairmen were J. C. Agarwal, division chief, process engineering, U. S. Steel Corp., Monroeville, Pa., and M. C. Chang, research group supervisor, Youngstown Sheet and Tube Co., Youngstown, Ohio.

The Purofer Direct Reduction Process

by Ludwig von Bogdandy, Heinz-Dieter Pantke, and Ulrich Pohl

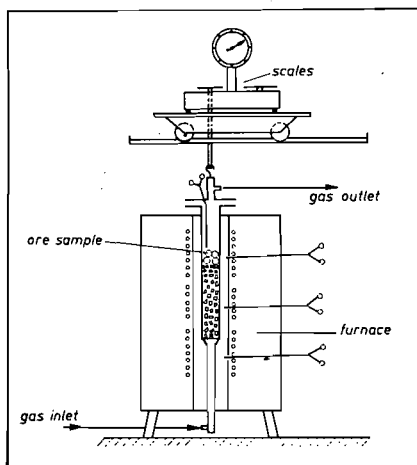


Fig. 1—Equipment for investigation of reduction speed.

The increasing availability of natural gas in many parts of the world and, moreover, the fact that great quantities of natural gas are flared for need of adequate use make direct reduction of iron ores with natural gas an economically and technically interesting proposition. Furthermore, in Europe it must be taken into account that the advent of big quantities of natural gas raises new problems for the integrated steel plants possessing coke ovens. Coke-oven gas is produced there as a by-product of metallurgical coke for the blast furnaces. This coke-oven gas has, up to now, been widely used in the chemical industry and in gas distribution for household use. Both groups are now slowly changing to fuel oil and natural gas so that a higher consumption of coke-oven gas is a must for most European steel plants. Thus, coke-oven gas is also considered as a sound basis for direct reduction.

The product of such a *direct reduction* process should be sponge iron in some lumpy form which can be used as a cooling medium in BOF's or as raw material for electric steelmaking furnaces. Furthermore, sponge iron can, in some

LUDWIG VON BOGDANDY, HEINZ-DIETER PANTKE, and ULRICH POHL are with Hüttenwerk Oberhausen AG, Oberhausen, Germany.

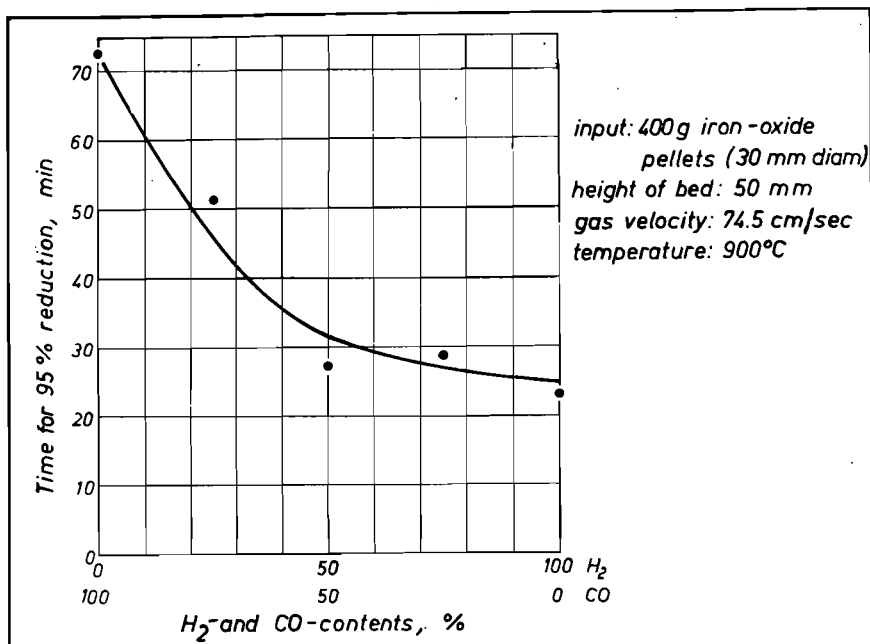


Fig. 2—Reduction time (95% reduction) as a function of gas composition (CO/H₂).

places, be used as a charging material for blast furnaces, taking some of the metallurgical load out of the furnace and resulting in higher efficiency for blast furnaces.

PRELIMINARY LABORATORY WORK

In order to shorten the time and to cut expenditures for development work, extensive laboratory studies were done, first to determine the optimum working conditions for a direct reduction process that would fulfill the above mentioned conditions and result in the highest efficiency possible, e.g. maximum output of iron and minimum gas consumption.

Fig. 1 shows the apparatus for measuring the reduction of various ores, pellets, and sinter as a function of the possible variation of working conditions. Some typical results are shown in Fig. 2 (effect of H₂ content in the reducing gas), and in Fig. 3 (effect of the grain size of various ores). The effects of temperature and gas pressure were also investigated. A very complicated relation was found between the reduction speed and the grain size.

Based on known physical chemical principles, the explanation could be found that $dR/dt \sim 1/d^n$, (where R = degree of reduction, d = particle diameter, t = time, and n = constant), and depending on the chemical step, which is rate controlling, n comes out as between 0 2.

Fig. 4 shows, for one example, the complete set of relations between reduction speed, temperature, degree of reduction, and gas composition. Based on these results, the optimum results can be expected at relatively high temperatures (limited only by the softening point of the ore), at H₂ contents of at least 50%, pressure near atmospheric, CO₂ and H₂O contents of the gas as low as possible, and grain size of the ore between 10-60 mm according to their reducibility. Smaller grain sizes than 10 mm will not increase the reduction speed very much, except at very low particle sizes of less than 0.1 mm. The proof for these conclusions is given in several published investigations.¹⁻⁷

Natural gas or even coke-oven gas, as such, cannot be used as a re-

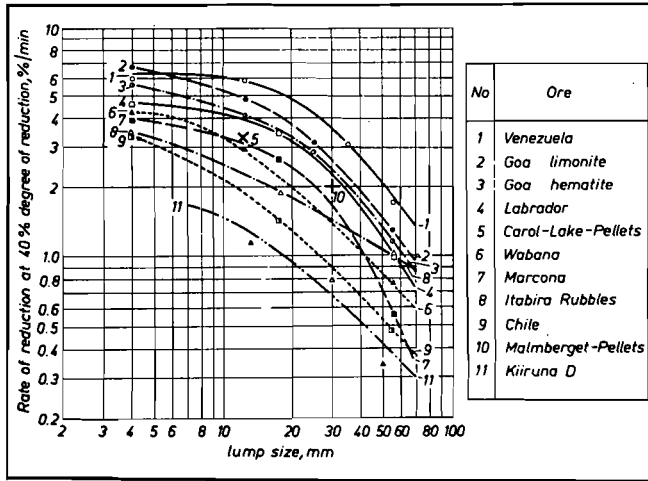


Fig. 3—Rate of ore reduction as a function of lump size (reducing gas: H₂, 800°C).

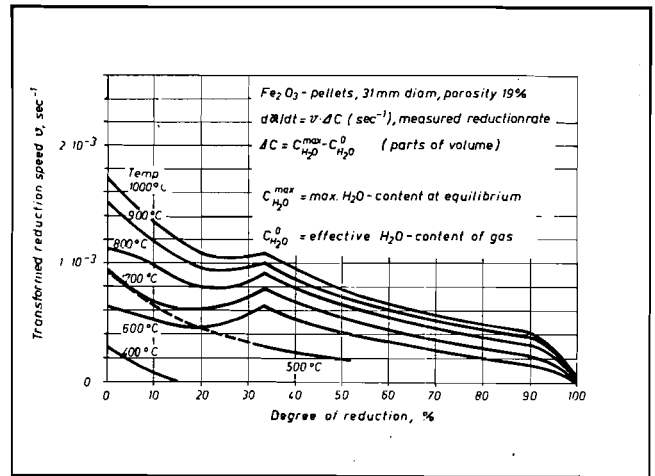


Fig. 4—Transformed reduction speed as a function of temperature and degree of reduction.

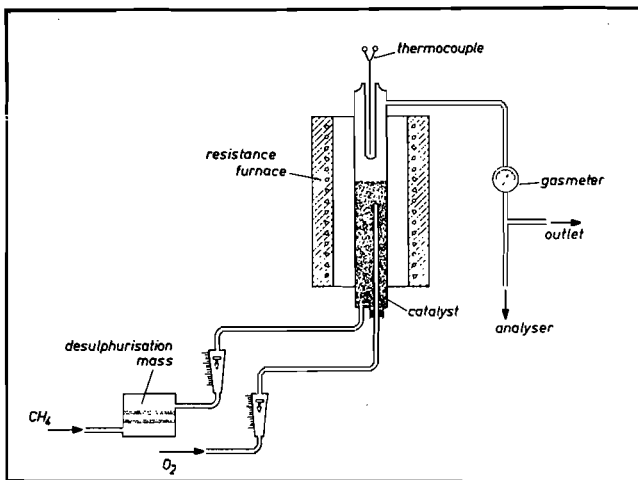


Fig. 5—Laboratory equipment for gas reforming trials.

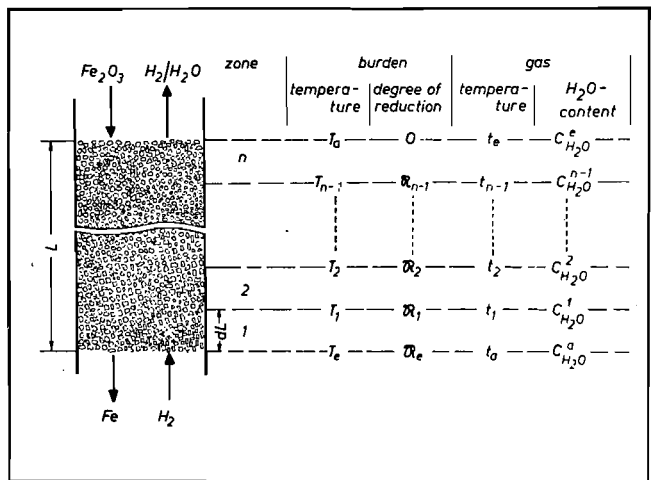


Fig. 6—Mathematical derivation of reduction in a shaft furnace.

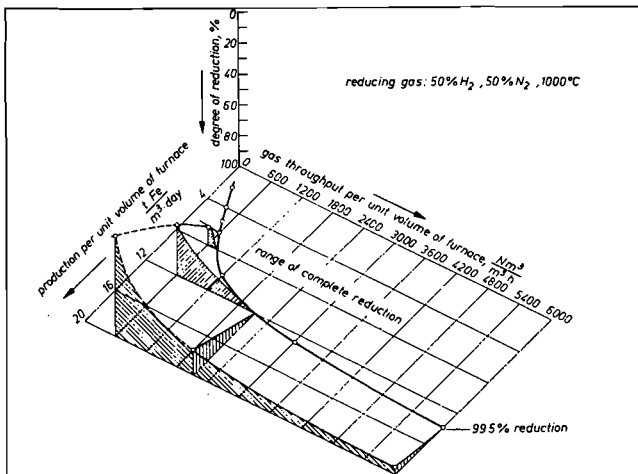


Fig. 7—Relation between degree of reduction, output of sponge iron, and throughput of reducing gas.

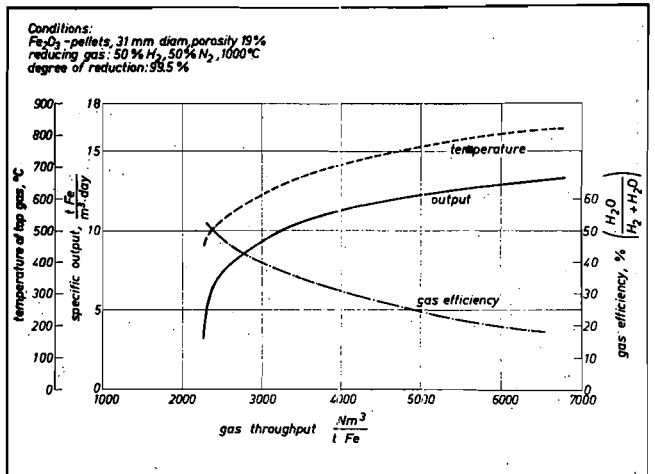


Fig. 8—Specific output as a function of specific gas throughput.

ducing agent. Only H₂, CO, or a mixture of both eventually also with N₂ are suitable for the reduction. It is necessary, therefore, to reform these gases according to such reactions as CH₄ + ½ O₂ = CO + 2H₂. The chemistry of the reforming reactions is well known, and the chemical industry makes use of them to produce synthesis gas. Contrary to the known gas reforming processes, it is neces-

sary for economic reasons to get the reforming done in one stage, i.e. to get a suitable reduction gas (CO, H₂, N₂) of about 1000°C in one heat with very low contents of H₂O, CO₂, and CH₄, without the need of cooling, washing, and reheating procedures. The optimum conditions for this reaction were found by extensive work with very simple laboratory equipment shown in Fig. 5.

MATHEMATICAL MODEL

It was clear from the beginning that, for financial reasons, the technical and economical risk for the first big-sized plant had to be kept to a minimum. To facilitate this last step, mathematical models on a physical chemical basis were developed to evaluate the advantages and disadvantages of the different principles for a direct reduction

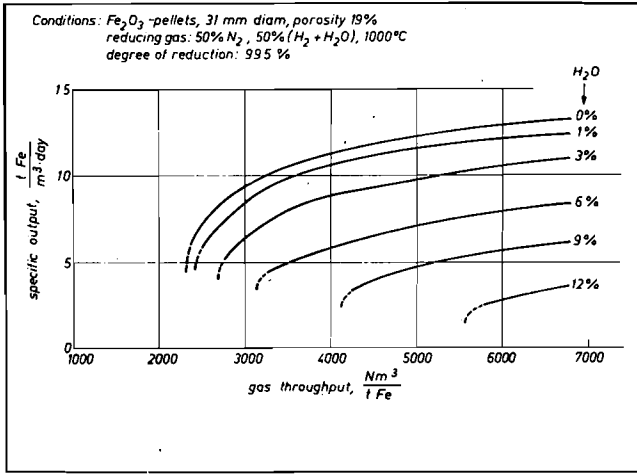


Fig. 9—Influence of H₂O-content in the reducing gas on the productivity.

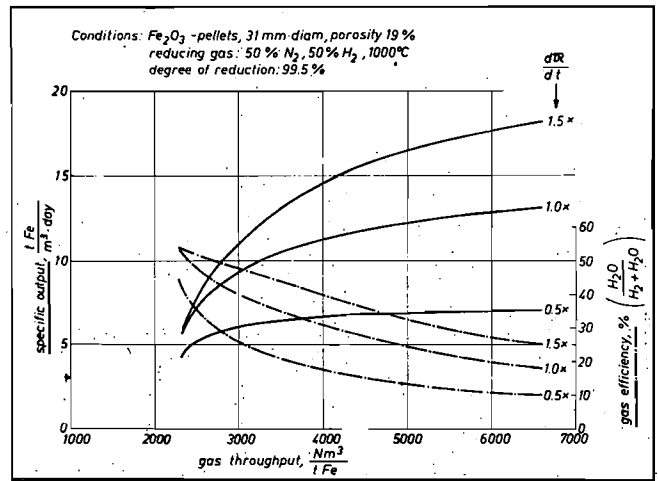


Fig. 10—Influence of the reducibility on the productivity.

process. After thorough investigation of batch processes in stationary retorts, fluidized beds, travelling grates, and rotating furnaces, it was found that the shaft furnace with counter-current flow of solids and gases should give optimum results, e.g. highest productivity per unit of volume and cross section of the vessel, and the lowest possible consumption of reducing gas.

The principle of the mathematical model for the shaft furnace is, therefore, demonstrated. For simplification, the reducing gas is considered to be pure hydrogen (Fig. 6). For a special investigated type of Swedish pellets of moderate reducibility, the reduction speeds are given in Fig. 4. For an arbitrarily chosen throughput of iron, temperature, and degree of reduction in the lowest layer of the furnace, the reduction speed with fresh hydrogen of 1000°C can be calculated by using the data in Fig. 4. As a consequence of reduction and heat transfer, the gas will have a lower temperature and a certain H₂O content after penetration of the lowest layer. This provides the starting conditions for the next layer. By going step by step through the reacting vessel and by iteration (a trial and error method with arbitrarily chosen values for the solution), the correct course of temperature of gas and solids, degree of reduction, and H₂O content of the gas are found. These steps were performed in an electronic computer.⁸ Fig. 7 shows one example of the results, and Fig. 8 contains the very important relation between specific output of iron per unit of volume of the vessel and gas consumption per ton of iron.

Remarkably high throughputs seem attainable at relatively low gas consumption. A realistic figure of 10 tpd Fe/m³ means that the shaft, for a daily production of 1000 tons of sponge iron, must not be greater than 100 m³, i.e. 3.5 m average diameter and 10 m high. Fig. 9 shows the detrimental effect of even small H₂O contents in the fresh reducing gas

on the productivity. The influence of the reducibility is shown in Fig. 10.

PILOT PLANTS

After several intermediate steps,^{6,10} a 25 tpd plant was erected. Fig. 11 shows schematically the gas flow in the installation. The performance of the plant was excellent.

Natural gas and rest gas (an intermediate between natural gas and coke-oven gas) were used. The gas compositions before and after the reforming stage are given in Table I.

Most of the reduction work was done with Swedish pellets, the reduction characteristics of which were shown in Fig. 4. Table II shows the chemical analysis before and after reduction in the pilot plant.

The sponge iron came out in the same outer appearance as the charged ore.

The ores shown in Table III were tested. These ores differed mainly in the gas temperature that could be applied without sintering in the lowest zone. This temperature was found to be in the range of 900° to 1050°C. Fig. 12 shows the comparison between the actual data of the pilot plant and the theoretical prediction from the mathematical model. Within the order of magnitude there is agreement, but still not satisfactorily close enough. The reason is thought to lie in the uneven distribution of the reducing gas, which is flowing into the vessel from the circumferential tuyeres and possibly may not penetrate the whole cross section of the furnace. Some improvement seems desirable in this respect, and will—as we hope—be attained by the development which is underway.

But, the productivity and the degree of gas utilization achieved are of a relatively high standard. With larger plants, even better results seem possible, due to lower heat losses per ton of iron. Plants of 300-500-tpd sponge-iron capacity now seem to be possible and are in preparation.

APPLICATION OF THE PUROFER PROCESS

Sponge iron will normally be converted into pig iron or liquid steel. The use in the blast furnace is well known and need not be discussed here.

Three further types of application were studied thoroughly.

Smelting of Sponge Iron in a Rotating Vessel¹¹

Fig. 13 shows an experimental 3-ton rotating furnace, which was fired with oil. The main obstacle of the smelting process was found in the reoxidation of the sponge iron. This could be overcome by a suitable charging technique which avoided the contact between the sponge iron and the oxidizing furnace atmosphere. Depending on the smelting conditions, it was possible to produce pig iron, steel, or iron of intermediate analysis (Table IV). The lining of the vessel was made of silica, and the relatively high contents of phosphorus and sulfur in the steel are the consequence. This way of making steel from sponge iron seems promising but needs further investigation.

Use of Sponge Iron in the BOF Steelmaking Process

Sponge iron can be used as a cooling medium substituting the normally used scrap in the BOF¹²⁻¹⁴. The advantage will be the easier charging. This possibility should be interesting for BOF steel plants that must make use of their coke-oven gas and are short of scrap, which will normally happen if continuous casting is used.

A good balance of gas and energy is reached when all of the coke-oven gas is used for production of sponge iron. The produced amount is just sufficient for the BOF. The excess blast furnace gas can then be used for heating the coke ovens

Table I. Analysis of Used Gases Before and After Reforming With Oxygen.

	H ₂ %	CO %	CH ₄ %	CnHm %	CO ₂ %	N ₂ %
Rest gas	9.8	11.4	54.1	1.9	0.9	21.0
Nat. gas	0.8	0.0	97.8	0.0	0.6	0.8
Reformed gas						
from rest gas	58.1	26.7	1.8	0.0	0.8	11.8
from nat. gas	63.1	25.4	4.7	0.0	1.7	4.8

Table II. Swedish Pellets, Chemical Analysis Before and After Reduction.

	Fe _{total} %	Fe _{met} %	SiO ₂ %	P %	S %	total gangue %
Ore	68.85	0.0	0.41	0.014	0.013	0.98
Sponge iron	95.30	90.50	0.57	0.020	0.010	1.36

Table III. Chemical Analysis of Tested Ores.

	Fe %	SiO ₂ %	P %	S %	total gangue %
Venezuela ore	64.8	1.37	0.11	0.055	3.57
Goa ore	59.9	2.70	0.08	0.035	8.43
Kiiruna-D ore	59.0	2.72	2.03	0.030	11.42
Itabira-Rubbles	68.4	0.80	0.06	0.010	1.30
Sinter	52.8	8.20	0.06	0.038	26.97

Table IV. Analysis of Iron and Steel Produced From Sponge Iron.

	C %	Si %	Mn %	P %	S %
Pig iron	2.76	0.22	0.18	0.065	0.039
Intermediate iron	0.35	0.01	0.02	0.280	0.020
steel	0.06	0.08	0.02	0.070	0.020

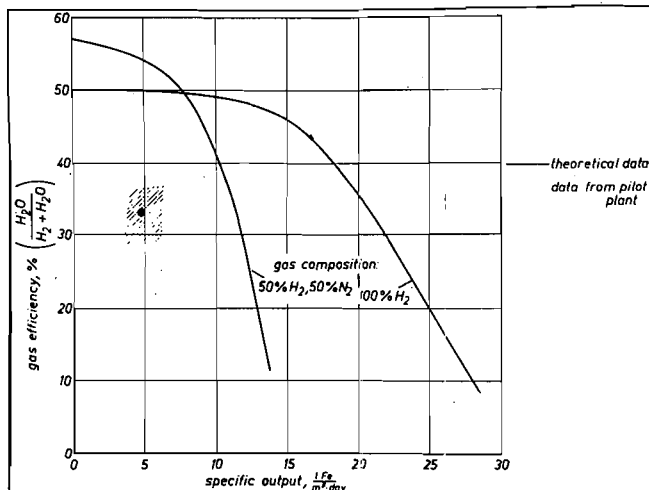
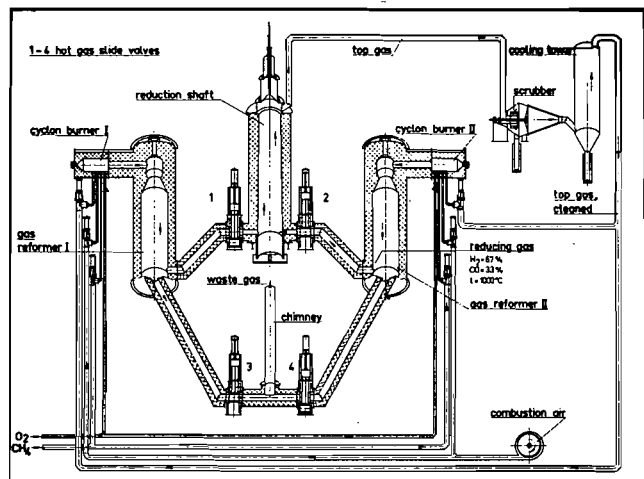


Fig. 12—Comparison between theoretical prediction and actual performance.

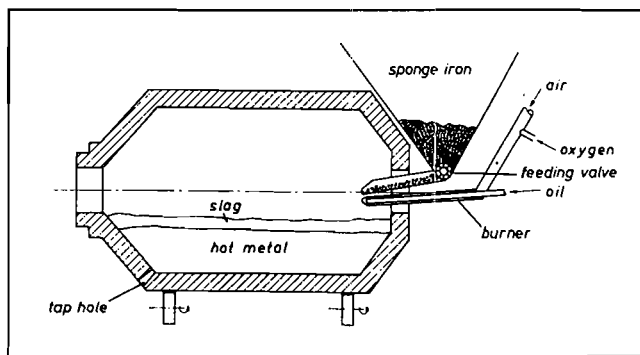


Fig. 13—Rotary furnace for smelting sponge iron.

$$c = c_{Fe} (p_{Fe} + 11.92 \cdot p_{gas} + proc + m + cap); \text{ \$/sht sponge iron}$$

Declaration:

- c_{Fe} = Fe-content of sponge iron
- p_{Fe} = costs of ore at the shaft furnace, \\$/sht Fe
- p_{gas} = costs of natural gas, \\$/10⁶ Btu
- $proc$ = processing costs, \\$/sht Fe
- m = costs of maintenance, \\$/sht Fe
- cap = capital charges and interests, \\$/sht Fe

Fig. 11 (left)—Gas flow of the 25-tpd Purofer pilot plant.

Fig. 14 (above)—Production costs of sponge iron.

and making electric power. There will be practically no surplus of energy; the plant will be nearly balanced.

Use of Sponge Iron in the Electric Steel Furnace

Steel can be made in electric furnaces on the basis of a high proportion of sponge iron (up to 100% in the charge)¹⁷⁻²⁰, without loss in productivity. The combination of the Purofer process and electric furnace steelmaking seems interesting for small and medium steel plants where scrap is short and natural gas supply is plentiful.

ECONOMICS

Based on 10 years of experimental work, the following conclusions can be drawn:

A 500-tpd sponge iron plant erected in Germany—excluding foundations, license, and insurance—will cost \$2.5 million.

The specific consumption of natural gas or coke-oven gas calculated

in heat units will be:

$$11.92 \cdot 10^6 \text{ Btu/short ton Fe.}$$

The approximate production costs of the sponge iron are shown in Fig. 14.

From this formula it is possible to make a first estimate on the probable cost of sponge iron in relation to the prevailing local conditions. For the following examples the cost of sponge iron (degree of reduction 90-95%, Fe-content 93%) is given:

Venezuela	23.97 \$/sht
Europoort	29.33 \$/sht
Ruhr district (Germany)	34.49 \$/sht

From this, it seems that the production of sponge iron is economically very interesting in Venezuela and countries with similar conditions, and also on the European coast where supply with natural gas is possible, while in the Ruhr district, the classical steelmaking area of Germany, the gas price at the moment is still too high to encourage mass production of sponge iron.

REFERENCES

- ¹ L. v. Bogdandy: *Arch. Eisenhuettenwesen*, 1961, vol. 32, pp. 275-286.
- ² E. W. Thiele: *Ind. Eng. Chem.*, 1939, vol. 31, pp. 916-920.
- ³ E. Wicke: *Z. Electrochem.*, Ber Bunsenges. phys. Chemie, 1956, vol. 60, pp. 774-782.
- ⁴ L. v. Bogdandy and W. Janke: *Z. Electrochem.*, Ber. Bunsenges. phys. Chem., 1957, vol. 61, pp. 1146-1153.
- ⁵ L. v. Bogdandy and H. G. Riecke: *Arch. Eisenhuettenwesen*, 1958, vol. 29, pp. 603-609.
- ⁶ O. Knacke: *Arch. Eisenhuettenwesen*, 1959, vol. 30, pp. 581-584.
- ⁷ L. v. Bogdandy, H. P. Schulz, B. Wuerzner, and I. N. Stranski: *Arch. Eisenhuettenwesen*, 1963, vol. 34, pp. 401-409.
- ⁸ L. v. Bogdandy and R. Wartmann: *Arch. Eisenhuettenwesen*, 1965, vol. 36, pp. 221-236.
- ⁹ L. v. Bogdandy: *Stahl und Eisen*, 1962, vol. 82, pp. 869-883.
- ¹⁰ H. D. Pantke: *JOURNAL OF METALS*, 1965, vol. 17, no. 1, pp. 40-44.
- ¹¹ J. T. Wasmuht: *Dissertation TH Aachen*, 1965.
- ¹² O. Masi and P. Cannizzo: *J. I. S. I.*, 1962, vol. 200, pp. 199-211.
- ¹³ K. G. Speith, H. vom Ende, and O. Steinhauer: *Stahl und Eisen*, 1963, vol. 83, pp. 961-971.
- ¹⁴ E. Ploekinger and K. Borowski: *Techn. Mitt. Krupp*, 1960, vol. 18, pp. 1-8.
- ¹⁵ R. F. Rinesch: *Congrès International sur les Acieries à l'Oxygène*, p. 13, Le Touquet, France, Sept. 23-25, 1963.
- ¹⁶ P. Feder: "Recent advantages in ferrous metallurgy", *Chem. Eng. Progress, Symp. Ser.*, 1963, vol. 59, pp. 135-141.
- ¹⁷ "New Iron Ore Reduction Techniques," p. 8, publ. by Corp. Venezuela Guyana, Caracas 1962.
- ¹⁸ J. L. Stalhed: *JOURNAL OF METALS*, 1957, vol. 9, pp. 246-249.
- ¹⁹ W. R. Weaver: *Iron Age*, 1960, vol. 185, pp. 18-24.
- ²⁰ J. L. P. Ayala and R. C. Jaime: *Iron and Steel Engineer*, August 1963, pp. 69-76.