

# Fluidized-bed reduction of fine iron ore by the *in situ* combustion of coal

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## SYNOPSIS

This paper describes an investigation into the prereluction of hematite fines in a fluidized-bed reactor as a means by which they can be prepared for direct smelting in a transferred-arc plasma furnace. A single-stage fixed fluidized-bed reactor using the *in situ* gasification of coal was employed.

It is shown that the hematite is reduced to magnetite at 1000 °C, the degree of reduction being limited by the inherent shortcomings of a single-stage reactor in which both the gasification and the reduction reactions have to occur. Methods by which the degree of reduction can be increased are suggested.

The gasification process was studied in some detail, and three different reducing agents are compared. The dominating effect of the Boudouard reaction on the behaviour of the fluidized bed is clearly illustrated. Non-coking coals of high reactivity are best suited to this process, a finding that would complement the trend away from coke as a metallurgical reducing agent.

## SAMEVATTING

Daar word verslag gedoen van 'n ondersoek na die voorreduksie van fynhematiet in 'n fluïedbedreaktor as 'n wyse om die erts voor te berei vir regstreekse uitsmelting in 'n oordraboogplasma-oond. 'n Vaste enkeltrapfluïedbedreaktor wat van die *in situ*-vergassing van steenkool gebruik maak, is vir die doel gebruik.

Hematiet is by 1000 °C tot magnetiet gereduseer. Die mate van reduksie word egter beperk deur die inherente tekortkominge van 'n enkeltrapreaktor waarin sowel die vergassings- as die reduksiereaksie moet plaasvind. Voorstelle word gemaak oor stappe wat geneem kan word om meer effektiewe reduksie te bewerkstellig.

Die vergassing van steenkool in die vergasser is in besonderhede bestudeer en drie verskillende reduseermiddels is met mekaar vergelyk. Die beherende invloed van die Boudouard-reaksie op die gedrag van die fluïedbed word duidelik geïllustreer. Die mees geskikte reduseermiddel vir hierdie tipe fluïedbed-reaktor is 'n nie-kookskool met 'n hoë reaktiwiteit. Die ontwikkeling van fluïedbedreakortegnologie is dus 'n verdere stap in die vervanging van kooks met steenkool as reduseermiddel, in die metallurgiese bedryf.

## Introduction

Large quantities of hematite fines (smaller than 5 mm) are currently being stockpiled in South Africa and, although lumpy iron ore (larger than 5 mm) is expected to be available for another 30 years or more<sup>1</sup>, there are several economic incentives for the development of a process that could use these fines as a feedstock.

A challenging prospect is the use of a fluidized-bed reactor for the prereluction of these fines, which could subsequently be charged hot to a transferred plasma-arc furnace for the completion of the reduction process. Meihack *et al.*<sup>2</sup> have shown that considerable savings in electrical energy can be achieved by the use of prereluced material as the feedstock to a plasma furnace.

The application of fluidized-bed technology in the pyrometallurgical industry is not new, and examples of its use include the reduction of iron ore with hydrogen<sup>3</sup>, the production of titanium tetrachloride<sup>4</sup>, and the roasting of various sulphide ores<sup>5</sup>. The combination of a fluidized-bed prereluction unit with a plasma-arc furnace is not a new concept either. The ELRED process<sup>5,6</sup>, which was proposed by ASEA of Sweden, comprises a recirculating fluidized-bed reactor in which the combustion of coal provides the heat as well as the reducing gas

required for the reduction of iron ore. This prereluction unit is coupled to a d.c.-arc furnace that uses a hollow carbon electrode through which the product of the fluidized bed is fed to the furnace. The prereluction stage has been successfully tested on pilot-plant scale at a throughput of about 400 kg of prereluced material per hour. The final reduction stage has been tested in a 25 t furnace at Domnarvet steelworks, Sweden<sup>5</sup>.

The gasification of coal *in situ* as employed in the ELRED process offers a challenging opportunity of replacing expensive reducing gas with a cheap source of fuel. It further creates the possibility that the range of coals suitable for iron-making can be enlarged. It is particularly important for the iron and steel industry in South Africa to move away from its dependence on coking coals: firstly, because coking coals are expensive and, secondly, because high-quality coking coal is in short supply<sup>7</sup>.

In the investigation described here, a single-stage, bubbling fluidized bed was chosen for evaluation as a potential prereluction unit, mainly because this type of fluidized bed is compact and therefore well-suited to laboratory-scale investigations, but also because it is fairly easy to operate. However, a single-stage bubbling fluidized-bed reactor gasifying coal *in situ* would not be considered for use as a self-contained prereluction unit on a production scale. A production plant should consist either of multistage units or a cascade of single stages. The emphasis in the present investigation was therefore

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not on the immediate development of a new process, but rather on an analysis of the principles involved in the *in situ* gasification of coal, the degree of reduction attainable in single-stage prereduction, and the intricate interaction between combustion and reduction.

#### Apparatus

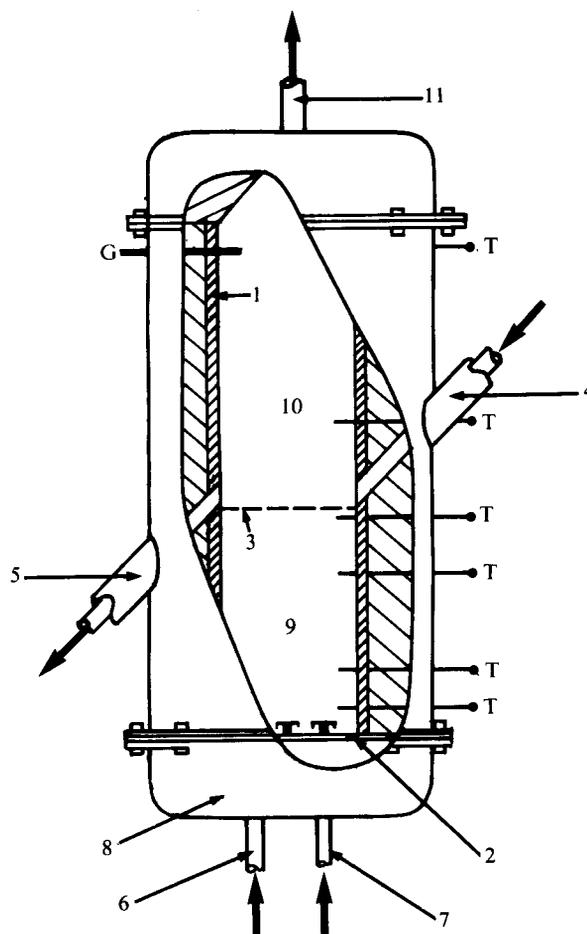
A schematic diagram of the fluidized-bed reactor used in the present investigation is shown in Fig. 1. The 'working part' of the reactor comprises an alumina tube with an inner diameter of 150 mm and height of 540 mm. The fluidized-bed overflow is 267 mm above the distributor plate, leaving a freeboard height of approximately 275 mm. A thermocouple is located at each of the following levels above the distributor plate: 30, 70, 170, 230, 330, and 515 mm. A multichannel digital thermometer displays one thermocouple reading at a time. The off-gas is sampled continuously and is analysed for oxygen, hydrogen, carbon monoxide, and carbon dioxide.

The feed materials are extracted from two sealed hoppers by screw feeders, and are gravity-fed into the reactor through the feed pipe. The product overflows through the product outlet into a sealed container. Air and liquified petroleum gas (LPG) are introduced via rotameters to the plenum chamber. LPG is combusted to preheat the reactor to a temperature at which coal combusts spontaneously (approximately 750 °C). The off-gas passes through a cyclone, where the dust is collected, and then through a burner where, as a safety measure, any remaining combustible gases in the off-gas are fully combusted.

#### The Gasification of Coal

The type of coal used for *in situ* gasification has a significant influence on the behaviour of a fluidized bed, since the characteristics of the coal determine the eventual temperature of the bed and the composition of the reducing gas. It is preferable to operate a fluidized bed at temperatures below 1000 °C so that sintering of the iron ore and coal particles is avoided. On the other hand, temperatures much lower than 1000 °C are detrimental to the kinetics of the reduction reactions. Furthermore, a ratio of carbon monoxide to carbon dioxide of at least 2,3 (at 1000 °C) is required before it is possible to reduce iron oxide to metallic iron (Fig. 2). It is therefore imperative that the rate of the reaction between carbon and oxygen be high enough for a sufficiently high temperature to be attained, while the Boudouard reaction ( $\text{CO}_2 + \text{C} \rightarrow 2\text{CO}$ ) should proceed at such a rate that the required amount of reducing gas is produced. A further complicating factor in this mass and energy balance is that the gas flow through the reactor should be sufficient for proper fluidization to occur.

The influence of the feed rate, reactivity, and volatile content of the coal on the behaviour of the fluidized bed was investigated. Three reducing agents were evaluated: Springbok no. 5 coal, Springbok no. 5 'char' (the carbonaceous product of the reduction experiments), and Collie coal. The chemical analyses and certain relevant properties of these reducing agents are summarized in Table I. The coal-evaluation experiments were conducted using silica sand with a particle size of 0,8 to 1,8 mm, instead of hematite ore, in the fluidized bed. This procedure was adopted so that the influence of the reduc-



- 1 Alumina tube
- 2 Distributor plate with bubble caps
- 3 Level of fluidized-bed surface
- 4 Feed pipe
- 5 Product outlet
- 6 Air inlet
- 7 Liquefied petroleum gas (LPG) inlet
- 8 Plenum chamber
- 9 Fluidized-bed area (below dotted line)
- 10 Freeboard area (above dotted line)
- 11 Gas off-take
- T Thermocouple
- G Gas-sampling probe

Fig. 1—The experimental fluidized-bed reactor

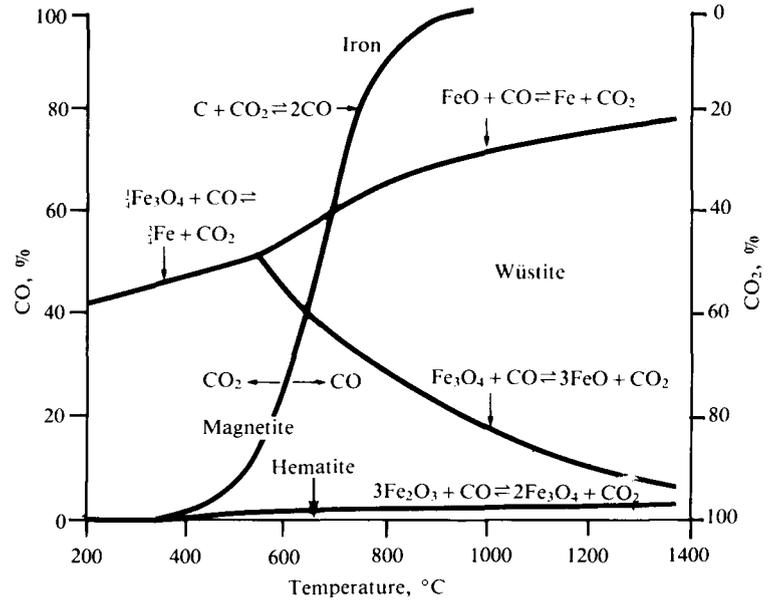
tion of hematite on the temperature of the fluidized bed and the off-gas composition could be established independently.

Fig. 3 illustrates the dependence of the bed temperature and the off-gas composition on the feed rate for Springbok no. 5 coal, while the dependence of the bed temperature and the ratio of carbon monoxide to carbon dioxide in the off-gas on the feed rates of the three reducing agents is shown in Fig. 4. Fig. 5 shows the temperature profiles through the reactor for the three reducing agents.

#### Effect of the Coal Feed Rate

Fig. 3 reveals that a sharp rise in the partial pressure of carbon dioxide, accompanied by a simultaneous decrease in the partial pressure of oxygen, occurs with increasing feed rate. This indicates that the carbon-combustion reaction,

**Fig. 2—Equilibrium diagram for the iron-carbon-oxygen system (after Stephenson<sup>6</sup>)**



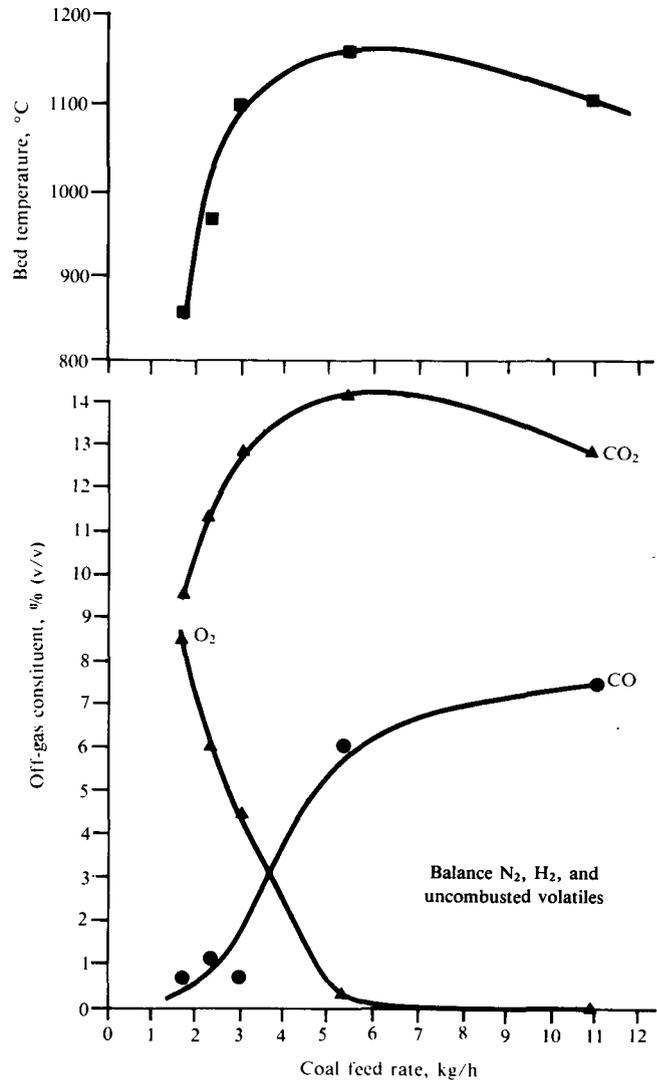
**TABLE I  
COMPOSITION AND PROPERTIES OF THE REDUCING AGENTS TESTED**

Reducing agent	Springbok no. 5 coal	Springbok no. 5 'char'	Collie coal
Comments	A slightly coking Transvaal coal	The carbonaceous product from the reduction experiments	A non-coking Western Australian coal
Particle size, mm.	1,0 to 1,9	Made from coal 1,0 to 1,9	1,0 to 1,9
Coal reactivity* (gC/min) (Iscor standard, 1000 °C) <sup>16</sup>	2,06	ND	3,70
Ash-fusion temp., °C	>1400	ND	>1400
Swelling index	About 3,5	ND	0
Composition, %			
Fixed C	55,6	61,2	50,9
Volatile matter	31,0	2,6	31,7
Ash	10,3	34,9	3,8
Moisture	3,1	1,3	13,6
Ash composition, %			
TiO <sub>2</sub>	1,13	ND	2,07
MgO	1,98		1,70
Al <sub>2</sub> O <sub>3</sub>	22,61		32,50
SiO <sub>2</sub>	59,12		37,70
CaO	3,85		1,47
Cr <sub>2</sub> O <sub>3</sub>	0,93		-
MnO	0,13		-
Fe <sub>2</sub> O <sub>3</sub>	3,79		20,90
P <sub>2</sub> O <sub>5</sub>	-		1,10
K <sub>2</sub> O	0,145		0,27
Na <sub>2</sub> O	0,038		1,06
Balance	6,30		1,20

ND = Not determined

\* Coal reactivity refers to the rate of the Boudouard reaction at a specified temperature

Silica feed rate 10,76 kg/h  
Air flowrate 21,5 kg/h



**Fig. 3—Effect of the feed rate of Springbok no. 5 coal on bed temperature (70 mm above the distribution plate) and on off-gas composition**

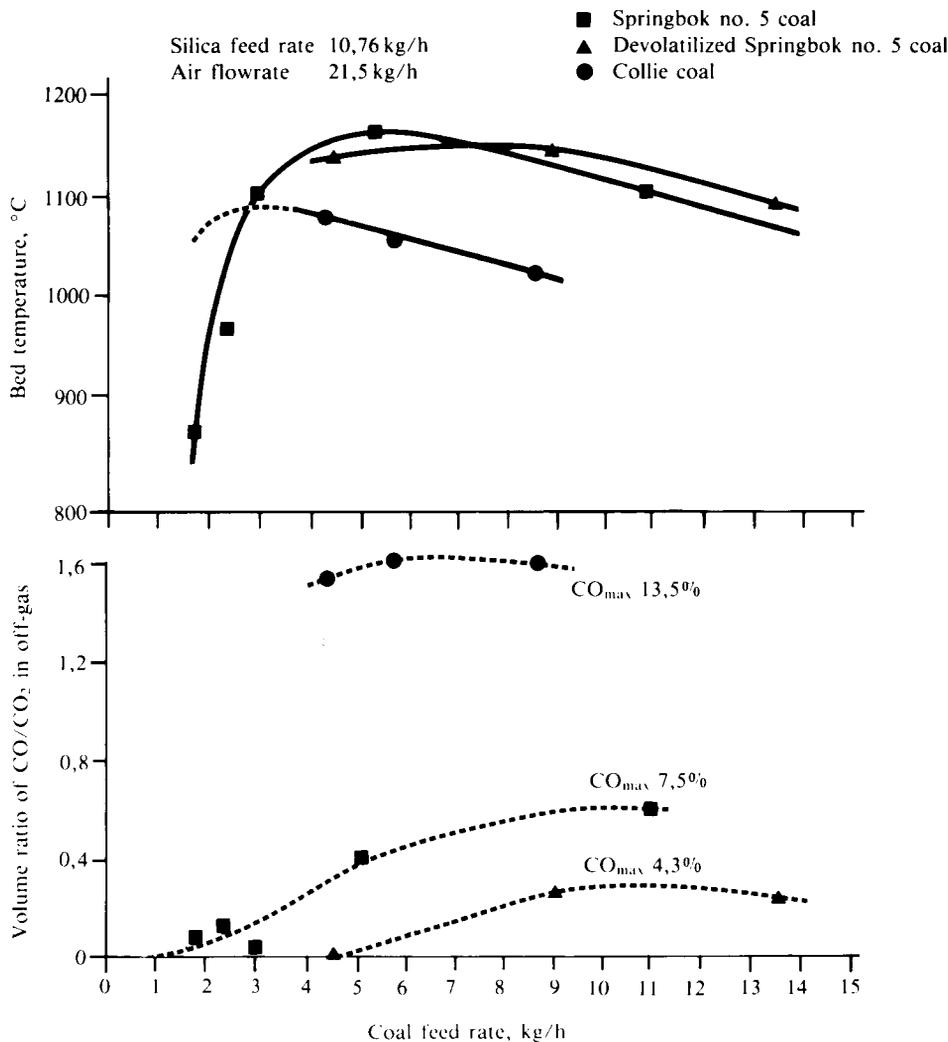


Fig. 4—Effect of feed rate on the bed temperature (70 mm above the distribution plate) and the ratio of carbon monoxide to carbon dioxide in the off-gas for three reducing agents



dominates the process at coal feed rates lower than 3 kg/h. This is confirmed by the accompanying rise in temperature, indicating that the heat of combustion provided by reaction (1) overrides the endothermic effect of the release and decomposition of the volatile materials, as well as that of the Boudouard reaction



At coal feed rates between 3 and 5,5 kg/h, reaction (1) still proceeds at a high rate, as shown by the rapid depletion of oxygen in the system. However, in this regime the Boudouard reaction gains in importance as carbon dioxide is converted to carbon monoxide, causing the rate of temperature increase to diminish. However, the combustion of the volatile materials, which consumes oxygen and releases heat, makes it difficult to quantitatively assess the relative rates of these reactions.

At coal feed rates higher than 5,5 kg/h, the oxygen in the system is depleted, carbon dioxide is consumed (carbon monoxide being released concurrently), and the temperature drops. The endothermic Boudouard reaction therefore dominates this regime, although the combustion of the volatile components of the coal may also contribute to the increase in the carbon monoxide content of the off-gas. In this regime, the temperature is also in-

fluenced, but to a lesser degree, by the heat capacity of the coal.

The difference in temperature distribution in the reactor as a function of coal feed rate is clearly shown by a comparison of the two curves for different feed rates of Springbok no. 5 coal in Fig. 5. At a feed rate of 1,72 kg/h, enough oxygen is available to facilitate combustion, even above the surface of the fluidized bed. Consequently, the temperature increases from the bottom to the top of the reactor. The combustion taking place within the fluidized bed is due mostly to the reaction of oxygen with fixed carbon, while that above the surface is attributable mainly to the reaction of oxygen with volatile matter. At a feed rate of 5,27 kg/h, more carbon is combusted, and hence the overall temperature in the fluidized bed is higher. Also, all the oxygen is consumed within the fluidized bed and the Boudouard reaction ensues, causing the shape of the two curves to differ considerably.

At a feed rate of 5,27 kg/h, a high-temperature zone develops close to the bottom of the fluidized bed, indicating that the exothermic combustion of carbon to carbon dioxide proceeds at a high rate in this region. This is in agreement with the findings of Cooke and Robson<sup>9</sup>, who observed that a zone with a temperature about 100 °C higher than that of the bulk of the bed exists near the bottom of a fluidized-bed combustor. The high

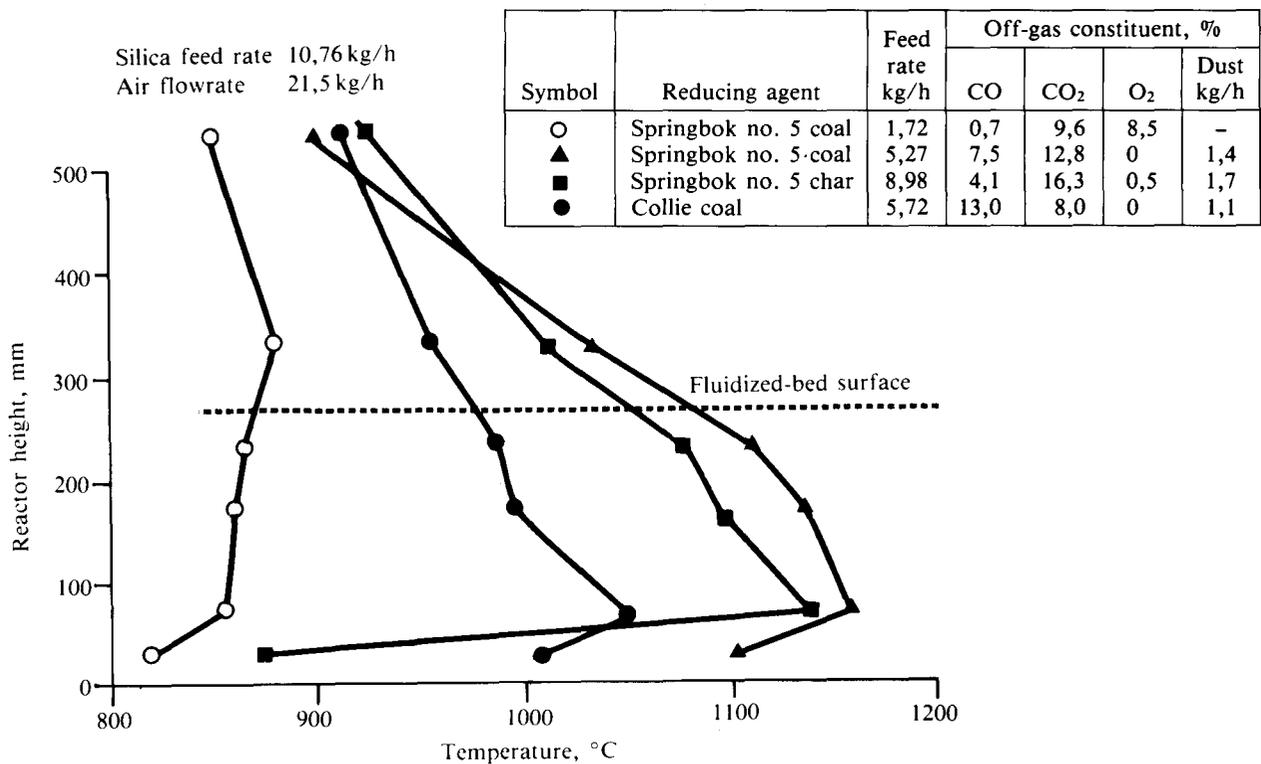


Fig. 5—Temperature profiles through the fluidized-bed reactor

temperature and excess carbon provide a strong driving force for the conversion of carbon dioxide to carbon monoxide, and this endothermic reaction results in a drop in bed temperature immediately above the high-temperature oxidizing zone. At the top of the fluidized bed, the temperature drops as a result of pre-heating of the feed material, devolatilization of the coal, and cracking of the volatile components of the coal. Above the surface of the bed, the temperature is determined by the endothermic reactions due to the cracking of volatile material and heat losses through the reactor containment.

#### Effect of Coal Reactivity

It is evident from Table I that the reactivity of Springbok no. 5 coal is lower than that of Collie coal. The reactivity relates specifically to the rate of the Boudouard reaction, and it is to be expected that this reaction will proceed at a much higher rate with Collie coal than with Springbok no. 5 coal, resulting in a higher rate of formation of carbon monoxide and a lower bed temperature. This is confirmed by the experimental findings reported in Fig. 4. A maximum of 13,5 per cent carbon monoxide was released during the gasification of Collie Coal, while only 7,5 per cent carbon monoxide was released during the gasification of Springbok no. 5 coal. Further evidence that the reactivity of the coal, and hence the Boudouard reaction, dominates the behaviour of the bed is provided by the fact that the maximum temperature of the bed, as well as the maximum release of carbon monoxide, occurs at lower feed rates for Collie coal, i.e. the Boudouard reaction proceeds at a higher rate at any given temperature for coal of higher reactivity (Fig. 4).

Inspection of the temperature profiles (Fig. 5) indicates that, for Collie coal and Springbok no. 5 coal at similar

feed rates, the coal with the higher reactivity causes a larger drop in temperature immediately above the zone of highest temperature because the Boudouard reaction proceeds at a higher rate. This stresses the dominating influence of the endothermic Boudouard reaction. In summary, not only is the atmosphere more reducing with the higher-reactivity coal (Collie coal), but the bed temperatures are also somewhat lower, yet high enough to ensure reduction of the hematite. Lower bed temperatures are advantageous because the softening of partially reduced iron ore will be less severe, reducing the possibility of clinker formation.

#### Effect of Volatile Content

An important conclusion can be drawn from a comparison of the bed temperature and the off-gas composition for Springbok no. 5 coal and those for its devolatilized equivalent. Fig. 4 shows that the temperature of the fluidized bed is virtually independent of the volatile content of the coal, but the char produces less carbon monoxide. A comparison of the temperature curves shows clearly that the endothermic decomposition of the volatile products does not dominate. However, at high feed rates of coal there is some indication that the release of volatile material does lower the temperature of the bed. The small difference in bed temperature that results from the use of standard coal, as opposed to devolatilized coal, is clear evidence of the dominant role played by the Boudouard reaction with respect to the temperature of the bed.

#### Coal Consumption

Because an excess of coal is required in a fluidized-bed process using the *in situ* gasification of coal in order to

produce reducing gas, it is to be expected that the coal will not be fully utilized, particularly in a single-stage design like that used in the present study.

The consumption of fixed carbon, degree of devolatilization, and degree of combustion of volatile materials for Springbok no. 5 and Collie coal are compared in Fig. 6. At coal feed rates lower than 3 kg/h, the consumption of fixed carbon increased with increasing feed rate for Springbok no. 5 coal, whereas the opposite trend was observed at higher feed rates. This apparent anomaly can be resolved if it is recalled (Figs. 3 and 4) that there was a sharp rise in temperature in this regime. As the feed rate increased, the temperature of the bed increased markedly, causing the carbon-combustion reaction to accelerate. The sharp decline in the oxygen content of the off-gas (Fig. 3) is further evidence that the carbon is gasified at an increased rate at higher coal feed rates, provided that the feed rate is lower than 3 kg/h.

It is clear that coal feed rates in excess of 4 kg/h should be avoided if a single-stage fluidized bed of the current design is to be operated economically. The consumption

of fixed carbon diminishes sharply at higher feed rates, with the result that unreacted coal forms a pronounced portion of the product stream. The sharp decline in the degree of combustion of volatile matter provides further proof that the increased amount of volatile matter made available at higher coal feed rates do not provide proportionally more reducing gas. At the high temperatures prevailing in the fluidized bed, the volatile materials are effectively driven off, but are not fully utilized as reducing gas, probably because the higher hydrocarbons are not fully cracked.

The combustion of volatile material is more complete for Collie than for Springbok no. 5 coal. When heated, Springbok no. 5 coal swells to form a char with a bulk density of about 500 kg/m<sup>3</sup>, whereas Collie coal does not swell and produces a char with a bulk density of about 800 kg/m<sup>3</sup>. It is to be expected that Collie coal would mix better into the bed than Springbok no. 5 coal owing to its higher density<sup>10</sup>, so that the volatile materials in the Collie coal are more likely to be combusted.

Although the reactivity of Collie coal is higher than

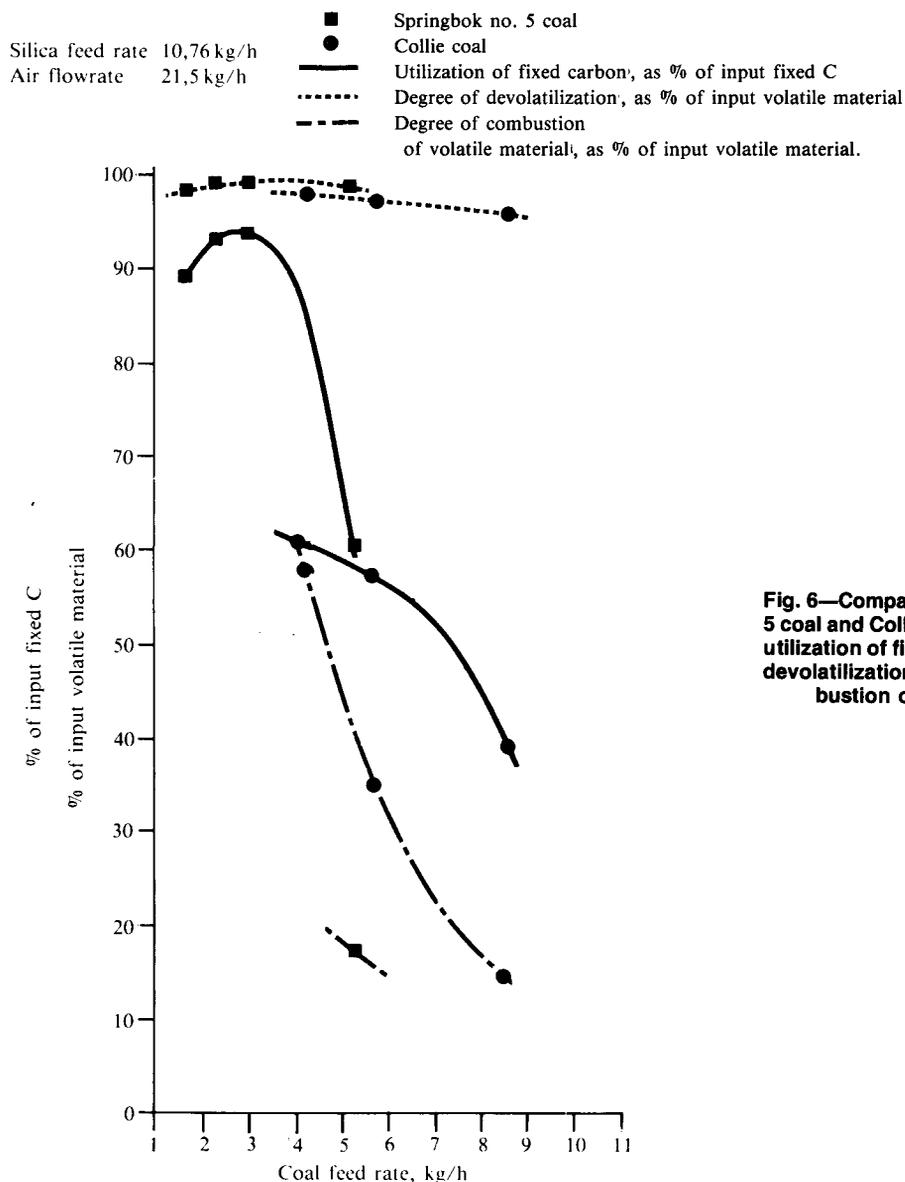


Fig. 6—Comparison of Springbok no. 5 coal and Collie coal in terms of the utilization of fixed carbon, degree of devolatilization, and degree of combustion of volatile matter

that of Springbok no. 5 coal, less fixed carbon is consumed. This may be due to the fact that the degree of combustion of the volatile components of Collie coal is higher and that, as a result, less oxygen is available for the gasification of the fixed carbon.

#### Stability of the Fluidized Bed

The fluidized bed frequently destabilized as a result of the formation of agglomerates (clinkers) when Springbok no. 5 coal or char was used, but not when Collie coal was used. It is essential that the mechanism of clinker formation be understood, because destabilization of the bed would lead to poor operational control.

Topper *et al.*<sup>11</sup> postulated that clinker formation in the high-temperature zone of a fluidized bed is primarily due to the tendency of a coal to swell. They suggested that char particles stick together when the burden passes through a regime of plastic flow at high temperature, and were able to relate this sticking behaviour to the swelling index. Should this postulate be correct, it is to be expected that Springbok no. 5 coal (with a relative swelling index of 3,5) should be prone to clinker formation, whereas Collie coal (with a relative swelling index of zero) should not have this tendency. This was found to be the case in the present investigation.

However, when char was used, clinker formation also occurred. This observation could hardly be related to swelling, and it would be difficult to envisage a relation between clinker formation and the swelling index of char as proposed by Topper *et al.* It is more likely that the sticking behaviour of char is the result of high-temperature softening.

During the formation of clinkers, the temperature in the lower zone of the bed rose sharply to above 1300 °C, and then fell to ambient temperature, while the temperature of the freeboard gas rose to about 1100 °C.

It is postulated that clinker formation starts when some char particles stick together in the high-temperature zone of the reactor. Owing to the relative inertia of the clinker, the heat generated by the combustion reaction in this area is dissipated less efficiently than in zones where the small particles are readily mixed. The first observable effect of clinker formation is consequently an increase in temperature in the hot zone. (Cooke and Robson<sup>9</sup> used similar reasoning to explain the overheating of the bottom part of a fluidized bed when the coal particles are too large to be fluidized sufficiently.) The temperature in the vicinity of the clinker rises sharply, probably exceeding the fusion temperature of the coal ash. The clinker now becomes even stickier, and grows as particles of char and silica become attached to it until these clinkers occlude the entire cross-section of the bed. Only a few channels are now available for the distribution of the incoming gas, with the result that the air is forced through these channels only, thereby increasing the gas velocity so that particulate material is blown into the freeboard area. The rise in the temperature of the freeboard gas observed experimentally during clinker formation is, to some extent, evidence that the entrained stream of coal (char) particles was combusted in accordance with this proposed mechanism.

#### Reduction of Hematite

Hematite fines from Iscor's Sishen Mine were reduced

in the experimental reactor in an attempt to determine the degree of reduction that is attainable in a single-stage fluidized bed in which the reducing gas is supplied by the *in situ* combustion of coal.

The ratio of carbon monoxide to carbon dioxide in the off-gas from Springbok no. 5 coal, which was combusted in the presence of silica, was about 0,5, and that in the off-gas from Collie coal was approximately 1,6 (Fig. 4). Wüstite would consequently be the only stable iron oxide phase when either of these coals was used as the reductant at a reaction temperature of 1000 °C (Fig. 2).

Springbok no. 5 coal was used as the reducing agent for hematite. (This was done despite the superiority of Collie coal as a reducing agent, since insufficient quantities of Collie coal were available.) In experiments designed to study the influence of retention time on reduction, the same iron ore was passed through the reactor three times in succession. LPG was continually introduced into the reactor to maintain the temperature below 1000 °C. During the third pass of the iron ore through the reactor, it was necessary to add nitrogen and to decrease the flow of air because temperature control of the fluidized bed became increasingly difficult. The experimental conditions are summarized in Table II.

The results of the reduction experiments are presented in Fig. 7. As the retention time was increased, the average reduction approached 11,1 per cent asymptotically. This

TABLE II  
CONDITIONS FOR THE REDUCTION OF HEMATITE

Iron-ore feed	Pass 1	Pass 2	Pass 3
	Hematite	Magnetic product from 1	Magnetic product from 2
Particle size, mm	0,5 to 1,0	12,88	12,31
Feed rate, kg/h	14,90		
Fe, %	65,80		
SiO <sub>2</sub> , %	3,22		
Al <sub>2</sub> O <sub>3</sub> , %	1,17		
Iron retention time, min	11	12	13
Coal	Springbok No. 5	Springbok No. 5	Springbok No. 5
Particle size, mm	1,0 to 1,9	1,0 to 1,9	1,0 to 1,9
Feed rate, kg/h	5,27	5,27	5,27
Fixed C, %	55,60		
Volatile matter, %	31,10		
Moisture, %	3,10		
Ash, %	10,30		
Feed rate of LPG, kg/h (NI/min)	1,87 (16,0)	1,87 (16,0)	2,80 (24,0)
Flowrate of nitrogen, kg/h (NI/min)	—	—	9,2 (121)
Flowrate of air, kg/h NI/min	21,7 (279)	21,7 (279)	14,5 (187)
Temperature, °C	950 to 1000	950	950
Approx. composition of off-gas, %			
CO	7,5	4,5	3,5
CO <sub>2</sub>	11,3	10,4	7,0
CO:CO <sub>2</sub>	0,66	0,43	0,50
H <sub>2</sub>	ND	5,0	3,8

ND = Not determined

Temperature 950 to 1000 °C  
 CO:CO<sub>2</sub> in off-gas Approx. 0,5

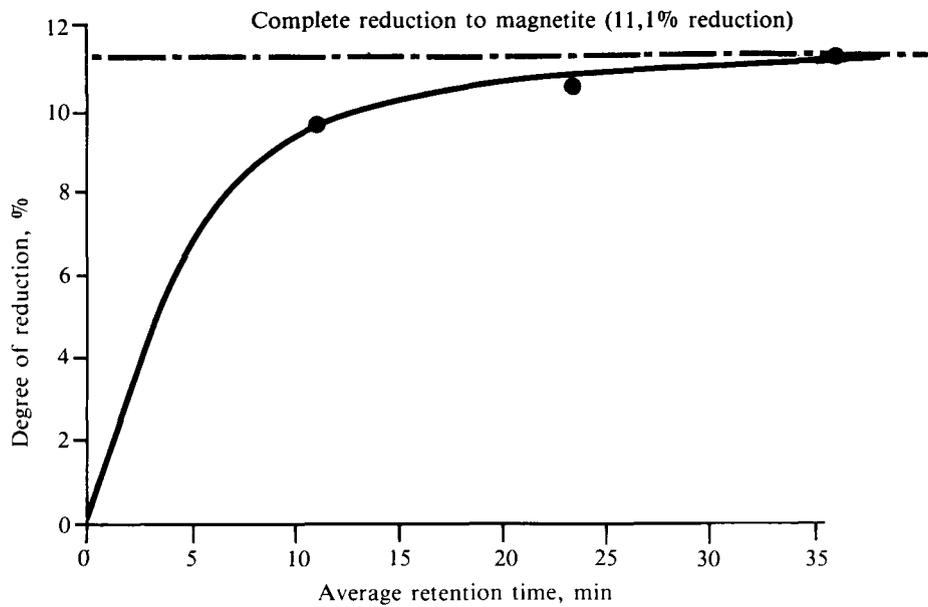


Fig. 7—Reduction of hematite

is equivalent to the complete reduction of hematite to magnetite, and indicates that the oxygen potential in the fluidized bed is not low enough for the reduction of magnetite to wüstite. This is in contrast to the equilibrium conditions suggested by the temperature and the ratio of carbon monoxide to carbon dioxide in the off-gas, which indicate that wüstite should form (Fig. 2). However, the measured ratio of carbon monoxide to carbon dioxide does not necessarily reflect the reducing conditions in the fluidized bed: firstly, because the combustion of volatile material can cause carbon monoxide to form even above the surface of the fluidized bed; and, secondly, because an oxidizing zone exists immediately above the distributor plate.

A mineralogical investigation of the partly reduced particles revealed that the hematite ore originally contained both dense and porous particles, the latter constituting some 82 per cent of the total. The dense particles exhibited typical topochemical-reduction behaviour (Fig. 8). The porous particles were reduced more homogeneously (Fig. 9), evidently because the reducing gas could penetrate the particles through the pores. After the second pass of the iron ore through the reactor, all the porous particles were fully reduced to magnetite. Although some of the dense particles were fully reduced to magnetite, others were reduced to less than 25 per cent magnetite.

Sporadic clinker formation occurred during the second and third successive passes of the iron ore through the reactor. These clinkers were very sticky at the operating temperature of the fluidized bed, and much more difficult to break than the silica-coal clinkers referred to earlier. When examined mineralogically, the clinkers were found to possess the structure shown in Fig. 10. This photomicrograph shows lath-shaped iscorite crystals with fayalite inclusions penetrating wüstite grains, which have magnetite rims. The presence in the structure of iscorite ( $5\text{FeO} \cdot \text{Fe}_2\text{O}_3 \cdot \text{SiO}_2$ ) is interesting, because this phase has been described only once before in the literature—by

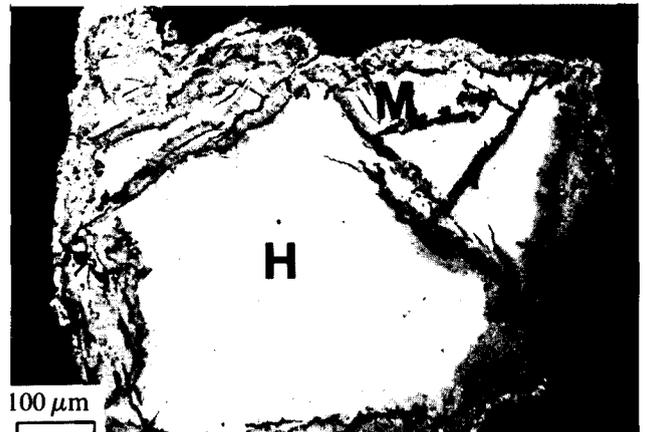


Fig. 8—Topochemical reduction of a solid particle of hematite (H) to magnetite (M)

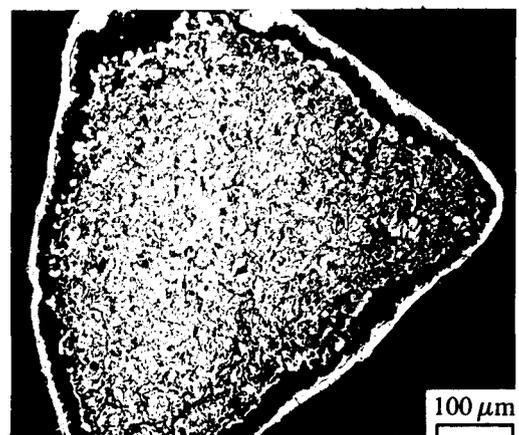


Fig. 9—Porous particle of hematite fully reduced to magnetite

Smuts *et al.*<sup>12</sup>. The iscorite phase was obtained in samples removed from the base of a reheat furnace held continuously at 1300 °C.

The following mechanism of clinker formation within the bed containing iron ore is proposed with reference to the phase diagram shown in Fig. 11.

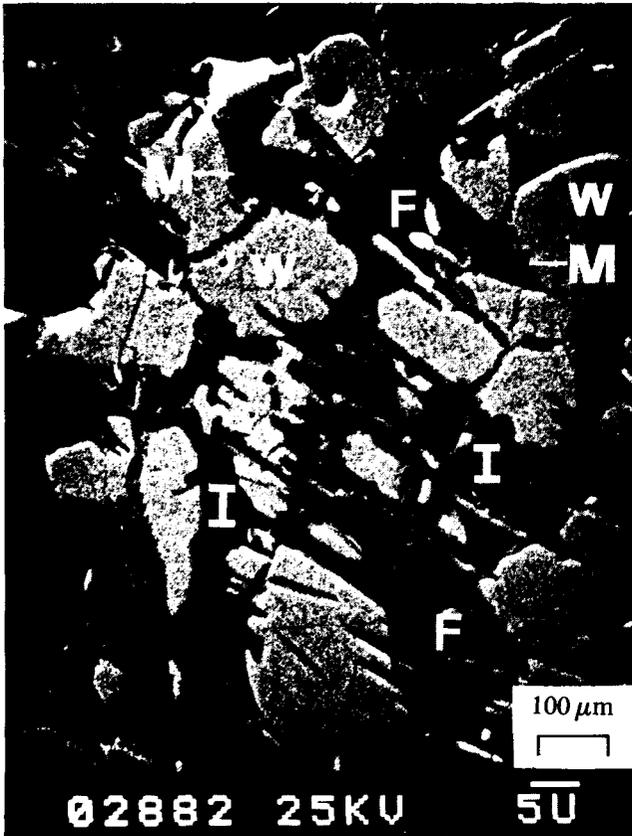


Fig. 10—Lath-shaped iscorite crystals (I) with fayalite inclusions (F) penetrating magnetite-rimmed (M) wüstite grains (W)

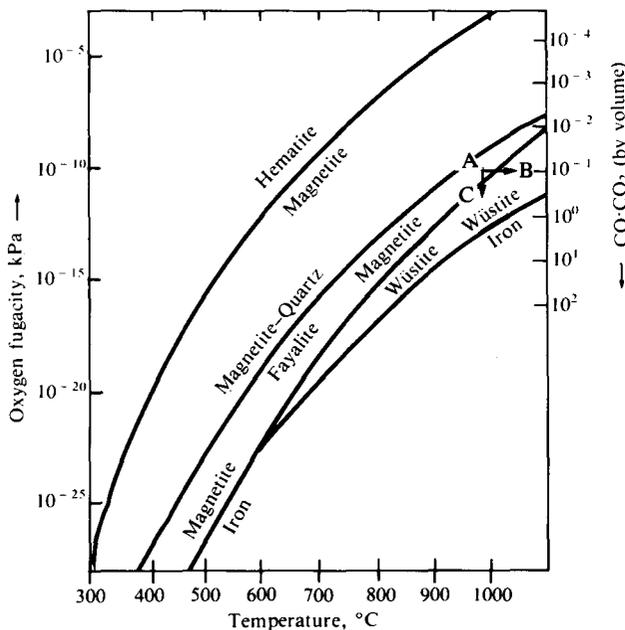
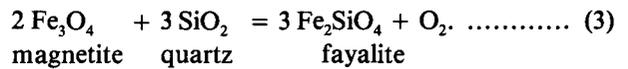
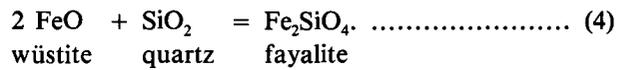


Fig. 11—Equilibrium diagram for the iron-silicon-oxygen system after Eugster *et al.*<sup>13</sup> The ordinate on the right-hand side was constructed from data presented by Muan and Osborn<sup>14</sup>

Because of the coking properties of the coal used in the present investigation, some char particles tend to stick together initially to form a small clinker. The clinker consequently becomes overheated because the heat is not dissipated efficiently, causing the coal ash to soften and particles of magnetite to attach themselves to the clinker. This mechanism of clinker formation is probably more prevalent just above the distributor plate, which, during normal operation, represents the hottest zone in the fluidized bed (Fig. 5). Although the clinker is contained in this relatively oxidizing zone just above the distributor plate, a rise in temperature would cause the ratio of carbon monoxide to carbon dioxide inside the clinker to increase, because the clinker contains carbon, and the formation of carbon monoxide from carbon and carbon dioxide may eventually cause the magnetite in the clinker to be transformed to wüstite (A-B or A-C in Fig. 11). However, fayalite may also be formed in association with the quartz contained in the coal ash:



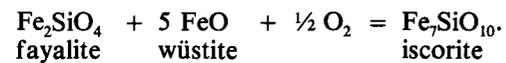
It is also thermodynamically feasible that fayalite may form by a reaction between wüstite and quartz, as follows:



(Wüstite is represented by the simplified formula FeO.)

If the assumption that the softening of the coal ash contributes to clinker formation is valid, the implication is that the temperature within the clinker was higher than 1400 °C (Table I). Also, since iscorite was observed mineralogically and previous investigators<sup>12</sup> have shown that iscorite forms only at temperatures in the region of 1300 °C, the temperature within the clinker must have been at least about 1300 °C. It was evident from the mineralogical investigation of the clinker that fayalite formed in the fluidized bed (Fig. 10). Since the liquidus temperature of fayalite<sup>15</sup> is only 1205 °C and the clinker clearly reached a higher temperature, the liquid fayalite must have contributed to the cementation of grains of iron ore, thereby promoting clinker formation.

It can be inferred from Fig. 11 that a decrease in temperature (B-A), probably accompanied by a decrease in the ratio of carbon monoxide to carbon dioxide (C-A), results in the transformation of wüstite to magnetite, which would explain the presence of the magnetite rims observed on the edges of the wüstite grains (Fig. 10). Similarly, the formation of iscorite would be expected to be an oxidizing reaction and, because of the association of iscorite with fayalite and wüstite (Fig. 10), the following reaction for the formation of iscorite is envisaged:



### Discussion

In the course of the present study, it was found that the coking properties of a given coal contribute significantly to the formation of clinker. Hence, it seems that only non-coking coals should be considered for *in situ* combustion in a fluidized-bed reduction process.

A further prerequisite for the acceptance of coal in this process is that the ash-fusion temperature should be in excess of 1400 °C so that melting, or even excessive softening, of the ash can be avoided. It was proposed, and observed, that fayalite may form by the reaction of silica with magnetite or wüstite. Nevertheless, the silica content of the coal ash is not considered to be of crucial importance, provided the requirements with respect to coking properties and ash-fusion temperature are met. A reducing gas with a ratio of carbon monoxide to carbon dioxide of at least 2,3 must be produced, because this is the minimum ratio needed for the reduction of iron ore to metallic iron at 1000 °C.

The present configuration of the fluidized-bed reactor and the coal used limited the degree of reduction of hematite to 11,1 per cent. This low degree of conversion can possibly be attributed to the following factors.

A gas of low reducing strength is produced in the fluidized bed. Apart from the properties of the coal that influence the production of reducing gas, two other factors may be of importance: the short retention time of the gas in the fluidized bed (less than 0,2 second), which does not allow sufficient carbon monoxide to be produced, and insufficient utilization of the volatile materials. Also, because an oxidizing zone is present in the fluidized bed just above the distributor plate, reduced particles of iron ore may be re-oxidized.

### Conclusion

Fine iron ore can be reduced with coal in a fluidized-bed reactor. However, modifications are needed to the design of the reactor used in this study to improve the efficiency of the process. For example, the retention time of the gas in the fluidized bed would increase if the bed were deeper; the utilization of the volatile materials would improve if provision were made for the coal to be fed into the bottom, rather than the top, of the fluidized bed; reduced particles of iron ore could be prevented from coming into contact with the oxidizing zone in the bottom of the reactor if the fluidized bed were divided into two compartments by the incorporation of a perforated horizontal baffle-plate. This, in effect, would transform the single-stage reactor into a two-stage co-current fluidized-bed reactor.

The coal used as both fuel and reductant in a fluidized-bed reactor should be non-coking and should have a high ash-fusion temperature, as well as a high reactivity. The use of the fluidized-bed process would complement the

trend away from the use of coke as a metallurgical reducing agent.

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