

VIEWPOINT

History of the iron furnace using the physical-chemical blast furnace model

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Abstract. The physical–chemical blast furnace model first built by Michard and Rist has proven to be a very efficient tool to predict and drive the present blast furnaces. It mostly describes the iron blast furnace as a counter current gas–solids dual heat and oxygen exchanger. The onset of coke gasification (or else of iron oxide direct reduction), leads to a two-zones exchange model. This theory is used to look back at the operation of the earlier and smaller iron furnaces such as the early XIXth century charcoal blast furnace or the much older low shaft furnaces. It shows the interest of using physical-chemical models to better understand the operation of past production tools.

Keywords: blast furnace / low shaft furnace / history / physical chemistry / archeology

1 Introduction

The energy transition is underway especially as it deals with the decarbonation of the economy, it is strongly associated with blast furnaces that are still producing 1,5 to 2 tons CO₂ per ton steel. The iron blast furnace is therefore probably experiencing a crucial moment in its history today. This is the right time to look back into the past and retrace its history:

- the modern blast furnace and the emergence of the theory of its operation; a real epistemological breakthrough,
- the charcoal blast furnace which has been the dominant model of pig iron production for several centuries in Europe and much longer in China,
- the low shaft furnaces or bloomeries that operated long before our era.

We shall do this by going back through history taking advantage of the most recent work to shed light on a still very uncertain past.

2 The modern blast furnace model: an epistemological breakthrough

We shall, therefore, rely on the pioneer work, of the recently deceased French engineers Jean Michard and André Rist (Fig. 1) who established the operating model of the blast furnace, as a result of theoretical work and practical

laboratory and industrial investigations, a true epistemological breakthrough, that has shifted the practice of the industrial blast furnace operation far from empiricism and from a statistical vision to a scientific approach based on physical chemistry and chemical engineering capable of driving the furnace operation in real time as well as predicting performance and predicting operating conditions under very different input parameters [1].

Their approach is based on the publication in the early 1960s of the results of vertical samplings in temperature and gas composition into blast furnaces, mainly in the Soviet Union, Belgium, France and Germany, which revealed the existence of a constant temperature zone called thermal reserve zone and a zone with constant gas composition called chemical reserve zone. On either side of these areas of thermal and chemical equilibrium, where the exchanges of materials and energy are very low, the rapid variations in temperature and in the composition of the gas show the significant intensity of the heat and oxygen exchanges [2–4].

Michard and Rist thus concluded to view the blast furnace as a series of two reactors and to separately describe these two reactors and their interconnection, which constitutes the basis of the blast furnace model.

Thus, the blast furnace is a double heat and oxygen exchanger in two zones (Fig. 2):

- The lower zone or processing zone in which the materials heat up from the reserve temperature of about 950 °C to the final temperature of the hot metal and the slag and undergo their final reduction from wustite to iron. In addition, the coke, inert in the upper zone, becomes reactive through the strongly endothermal

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Jean Michard (1926-2018)



André Rist (1927-2021)

Fig. 1. J. Michard and A. Rist. The founding fathers of the blast furnace model.

Boudouard reaction $C + CO_2 \rightarrow 2CO$. It is this reaction that accounts for the stabilisation of temperatures. The slowing down of reduction is the consequence of the approach of the reduction equilibrium between the gas and wustite/metallic iron. This processing zone is the only place where the energy necessary for the operation of the blast furnace is produced by the combustion of coke in front of the tuyeres. It is therefore its energy balance that determines the energy needs of the blast furnace.

- The upper zone or preparation zone in which materials charged at the top heat up to the temperature of the reserve zone and reduce from hematite to wustite. It is a recuperator of the energy of the gases coming out of the lower zone which is mainly used to preheat the materials charged at the top.

The separation into these two connected reactors (an epistemological breakthrough, one that completely modifies points of view and constitutes a real scientific breakthrough, as Bachelard would say) integrates the work of the French scientists Chaudron, Boudouard, Le Chatelier, of the German Reichardt and the Russian Kitaïev [6–9] (Fig. 3).

This vision of the blast furnace is reflected in two diagrams that interact in a dual way and have become famous all over the world [10–13] (Fig. 4).

- The Rist diagram on the left in which the exchanges of oxygen between gas and solids are represented by a straight line describing in its left zone the production of gas by the reaction of carbon on the abscissa with the blast and with iron oxides by direct reduction and in its right-hand zone the use of gas by the indirect reduction of iron oxides by the CO produced at the tuyeres and by direct reduction on the ordinate. We will underline that the slope of the line $\mu = C/Fe$ is the carbon fuel rate of the blast furnace in carbon atoms per iron atom.
- The Reichardt diagram describing heat exchanges between the gas generated at its flame temperature at the tuyeres and the solids heating up in counter current with the gas and to provide heat required by the reactions of reduction of iron oxides and coke gasification. The temperature on the abscissa, the enthalpy exchanged between gas and solids on the ordinate. The gap between the blue curve of gas and the red curve of solids shows the driving force of exchange; the efficiency of the exchange is characterized by the parameter u the ratio of the heat capacity flow rates of solids and gas.
- To which we add the Chaudron diagram which defines the equilibria of reduction of iron oxides.

The three enthalpy-temperature Reichardt diagrams presented here show the extent of the improvement of the blast furnace from the 1950s until 1982 when the lowest in the world coke rate with all coke operation was reached at Fos (Fig. 5).

The reduction in energy needs was the result of several decisive actions:

- The elimination of highly demanding energy side reactions such as low temperature decomposition of hydrates and carbonates contained in ores thanks to the agglomeration of the ores. The thermal pinch of the temperature enthalpy diagram thus moved from 650 °C to 950 °C, to the next endothermal reaction, that of coke gasification starting at about 950 °C, therefore drastically reducing the energy requirements of the lower processing zone.
- The possibility, consequently, of increasing the blast temperature thanks to the recycling in the hot stoves of part of the latent energy contained in the top gas, while continuing to lower the thermal needs by lowering the weight of slag, by reducing the heat losses and improving the reducibility of materials.
- The blast furnace coke consumption predicted by the model and realized in practice was divided by a factor greater than 2 during this period.

3 The charcoal blast furnace revisited: physical chemistry applied to Ebelmen work

We are now taking a step back in time, almost two centuries back, thanks to Ebelmen, a scientist and engineer unfortunately little known as he died at the age of 38 but whose results of pioneering measurements reported by Salvétat in 1855 allowed characterizing precisely the operation of charcoal blast furnaces at the beginning of the XIXth century (and may be much earlier). Ebelmen was indeed able to use the new analytical methods developed by chemists to characterize the composition of the materials and that of the gas at different levels of the furnace during the descent of the burden. Although thermocouples did not exist at that time, he was even able to assess the temperature range of materials from the color of the extracted boxes containing charcoal and ore extracted from various levels of the vessel [15–17] (Fig. 6).

Ebelmen's work was carried out on several French and Belgian blast furnaces. We will focus on the results that were obtained on Clerval blast furnace for which large amount of data collected during measurement campaigns in 1840 and 1841 are available. This blast furnace has a total height of 8 m (a shaft height of 5.7 m) and a volume of 12.9 m³.

Ebelmen not only recorded the operating results of the furnace, but he also charged boxes containing ore and charcoal which he removed from the furnace for analysis when they reached varying depths.

Ebelmen noted the depths reached by the boxes of coal and ore at various times. The rate of descent of materials seems constant over time and about 0.7 m per hour at the furnace top. It agrees with the prediction of the plug flow descent model of the burden which calculates the height reached as a function of the charged volume and from the

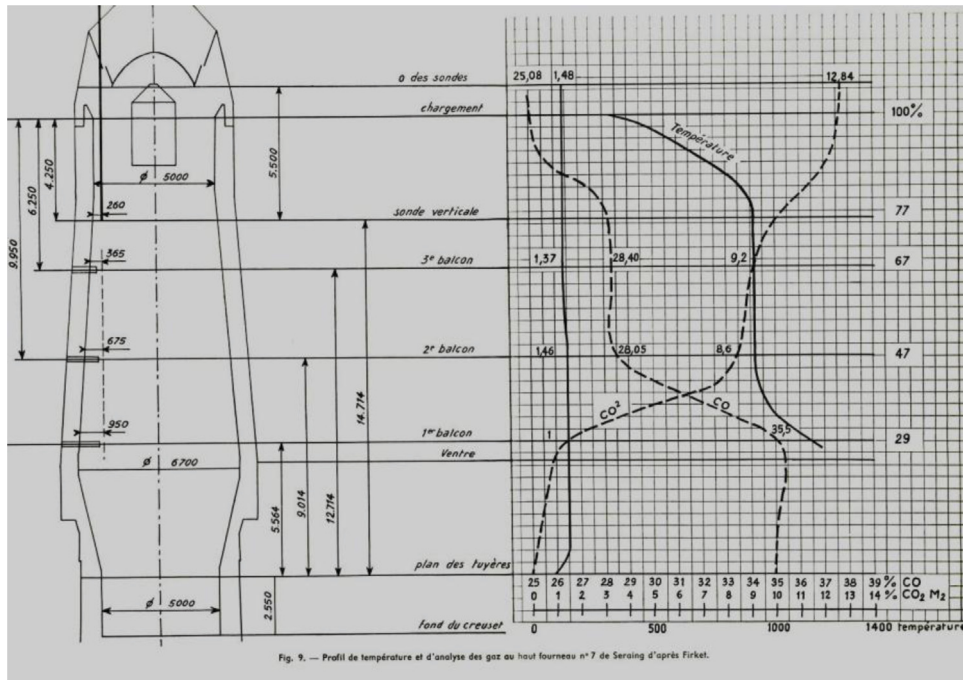


Fig. 2. Vertical samplings into the blast furnace [4].

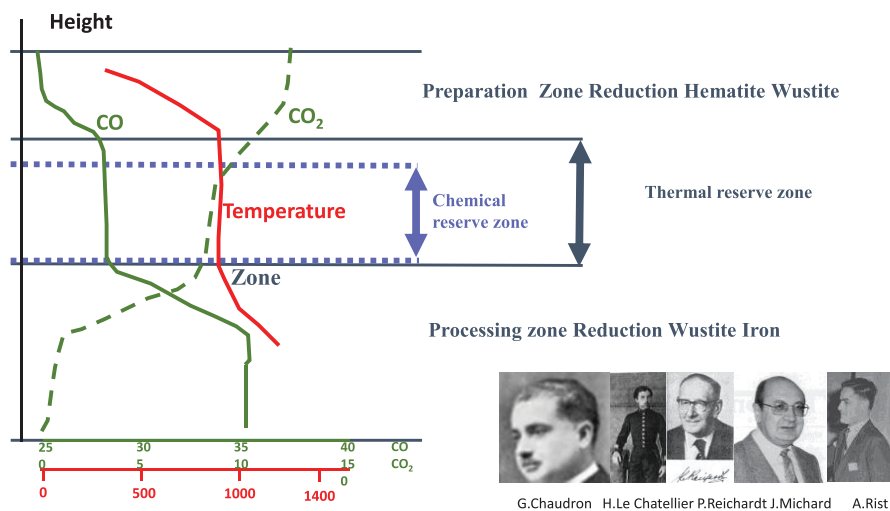


Fig. 3. Thermal and chemical reserve zones.

profile of the blast furnace. The slight difference between the model and the measurement is linked to the following three factors (Fig. 7):

- degradation of the materials by reaction and heating and decrease in the void ratio,
- softening and partial melting of the materials in the lower zone,
- slight deviation from the plug flow by peripheral extraction of charcoal by combustion with nozzles.

Here we show an important parameter: the residence time of the materials which can be calculated from the volume of the furnace in relation to the volume of materials

charged. This is the time allotted to carry out, partially or totally, the targeted oxygen and heat exchanges.

Temperature was estimated from the color of sampling boxes containing ore and charcoal pieces and removed after reaching different levels (for example, dark red was 700 °C whereas bright white was 1100 °C). Vertical soundings show the change in the temperature of the materials during their descent into the furnace with a temperature plateau at about 700 °C, the temperature of the reserve zone (Fig. 8).

They also give the evolution of the composition of each of the two ores (acidic and basic) evaluated by their degree of oxidation $Y = O/Fe$ oxygen atom per iron atom, from the hematite Fe_2O_3 $Y = 1.5$ to the wustite FeO $Y = 1.05$.

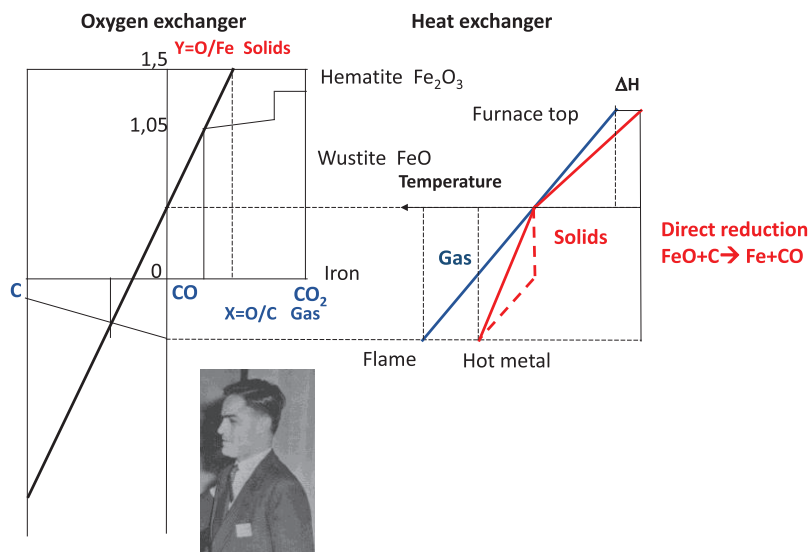


Fig. 4. A dual oxygen and heat exchanger connected Rist (right) and Reichardt diagrams.

They also provide valuable information on the reduction equilibria of limestone ores in which the presence of lime modifies the composition of iron oxides and where the chemical analysis suggests the formation of calcium ferrites such as $\text{CaO} \cdot 3\text{FeO} \cdot \text{Fe}_2\text{O}_3$. Oxidation degree $Y = 1, 2$ at O/ at Fe (the dotted line corresponds to the calcium ferrite C.3W.F reduction equilibrium line) (Fig. 9).

It shows an overall frame picture of the charcoal blast furnace operation with two exchange zones like the blast furnace of Michard and Rist but where the temperature of the reserve zone of 700°C , clearly lower than that observed on coke blast furnaces (950°C) is determined by the endothermic decomposition reactions of carbonates contained in the ore and limestone and volatiles contained in charcoal. The duration of the temperature and oxygen exchanges is greater than 2 h.

We can thus draw the theoretical Rist and Reichardt diagrams based on this model of the blast furnace (Fig. 10).

The ores and charcoal charged into the furnace go through the following successive steps during their descent into the shaft:

- Drying of materials
- Dehydration $2\text{FeOOH} \rightarrow \text{Fe}_2\text{O}_3 + \text{H}_2\text{O}$
- Indirect reduction of iron oxides and burden heating
- Devolatilization of charcoal
- Decarbonation of materials $\text{FeCO}_3 \rightarrow \text{FeO} + \text{CO}_2$
 $\text{CaCO}_3 \rightarrow \text{CaO} + \text{CO}_2$
- Indirect Reduction and burden heating
- Direct reduction

And validate them from analyzes of gases and materials taken at various depths of red dots.

The application of the principles and methods of Michard and Rist to the charcoal blast furnace observed by Ebelmen shows all the benefits that can be derived from the application of modern methods of process engineering to the study of reactors or reacting devices that no longer exist.

They allow us to have a very precise description of the operation of the charcoal blast furnace, which operated for at least 3 centuries in Europe.

4 Low shaft furnaces and bloomeries: gas producing reactors

To go back in time again, we have many simulations that are unfortunately too often poorly instrumented. These simulations come from 1 to 2 m high furnaces. In France, in addition to regular demonstrations by blacksmiths, the work of Leroy, Merluzzo Le Carlier with Lorraine ores [20], in Japan those reported in the literature on reduction in shaft furnaces to simulate and better understand the operation of Tataru furnaces for example [17–19,21–24].

The low shaft furnace with a height of between 1 and 2 m approximately behaves like a gasifier [23] where the thermal pinch is located at the bottom of the shaft.

The ratio of coal and ore bed thicknesses is 8 to 16 versus around 0.65 today.

The shaft furnace nevertheless remains a double exchanger of oxygen and heat.

The flame temperature of around 1400°C is close to that of the solids/liquids produced around 1200°C , unlike the modern blast furnace where the thermal pinch rises towards the reserve zone and where the flame temperature can reach 2200°C for liquids at 1500°C . The top gas temperature of around 500°C is very different from that of materials charged at a temperature of 25°C whereas in modern blast furnaces, the top gas temperature is often around 100°C . The overall u parameter of the furnace is of the order of 0.7–0.8, whereas it is today of the order of 1.3.

The energy generated at low flame temperature in low shaft furnaces leads to high gas flow rates while in modern blast furnaces the very high flame temperature makes it possible to reduce specific gas flow rates. The parameter μ

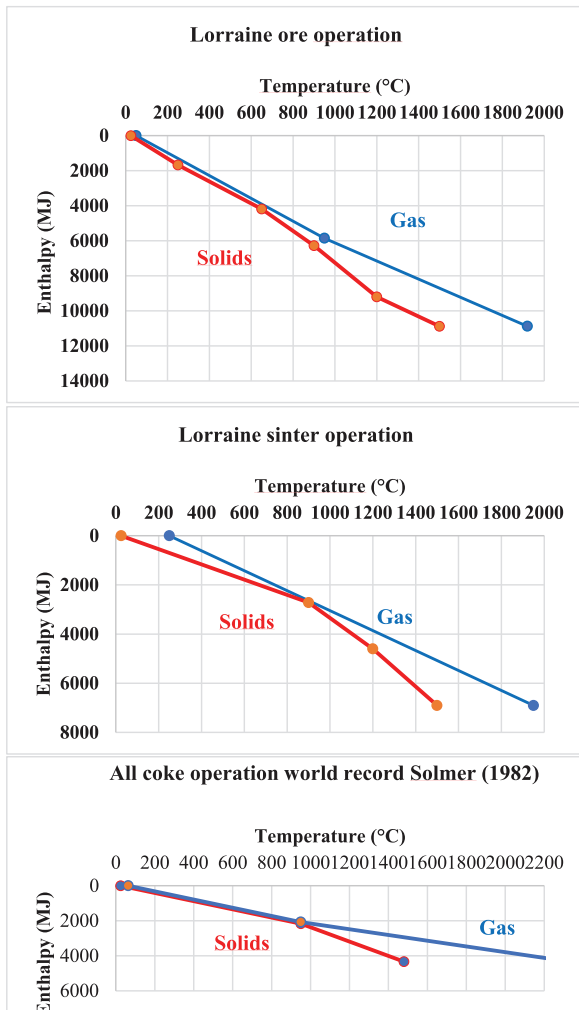


Fig. 5. From the Lorraine ore blast furnace to the all-coke high grade ore blast furnace [4,14].

specific carbon consumption per unit of iron (atom/atom) is of the order of 6–12 while it is close to 2.5 on modern blast furnaces

We will consider two cases encountered in the literature:

- 2 tons of charcoal per ton of ore
- 1 ton of charcoal per ton of ore

The heat balance of these blast furnaces that has been done from the available published information in the quoted papers [19–24] shows the following main items:

- Heating materials in the range of 2500 to 3000 MJ per ton of iron.
- Heat losses of the order of 3000 to 4000 MJ/ton of iron or of the order of 20 MJ/hour and per m² of furnace wall.
- The energy loss through the top as sensible heat is very significant and constitutes between 1/3 and half of the energy balance of the furnace. But the top gas consists mainly of CO and N₂ and its latent heat is, more than 2.7 times that involved in the furnace, which underlines again that these furnaces behave like gas producers.

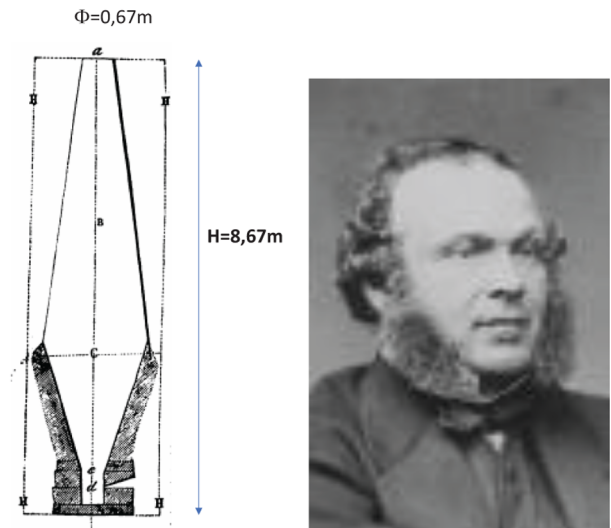


Fig. 6. Clerval blast furnace 1840 J.J. Ebelmen.

- The rest of the available energy is distributed in the competition between solids heating and reduction (essentially direct by carbon) of the iron oxides. Under these conditions, heating and reduction are carried out only very partially.

The temperature enthalpy diagram of the blast furnace highlights:

- The curve of the gases in blue, which cool from the flame temperature of about 1400 °C reached at the tuyeres up to the temperature of 500 °C reached at the top.
- The solids curve made up of 3 segments: two gently sloping segments representing the heating of the solids and a vertical segment representing the enthalpy of the Boudouard reaction $C + CO_2 \rightarrow 2 CO$ (Fig. 11).

The energy available for this reaction at the temperature where it occurs (here 1100 °C) corresponds to the difference, at this temperature, between the curve of the gas and the curve of the solids: therefore, only a fraction of the energy contained in the gas can be used, the remainder being lost in the form of sensible heat at the top. This is the consequence of the thermal pinch located in the vicinity of the tuyeres. However, increasing the coal rate will increase the direct reduction of iron oxides a step further...at the expense of higher energy losses.

Low values of u characterize rapid heating.

The high values of μ characterize gas volumes much greater than those required by reduction reactions.

One of the key factors in the production of iron in the blast furnace is the reduction of iron oxides. Carried out at low temperature by CO gas, its thermal impact is low. Ebelmen's soundings show that a residence time of 2 h at low temperature is necessary for this reduction to take place. Beyond 800 °C (charcoal) or 950 °C (coke), the reduction is achieved in part or entirely directly by carbon at the cost of a very high energy consumption (of the order of 3 to 4.5 GJ/ton iron, a value of the same order of magnitude as the heat losses or of the materials heating energy. Giving the time for indirect reduction implies increasing the

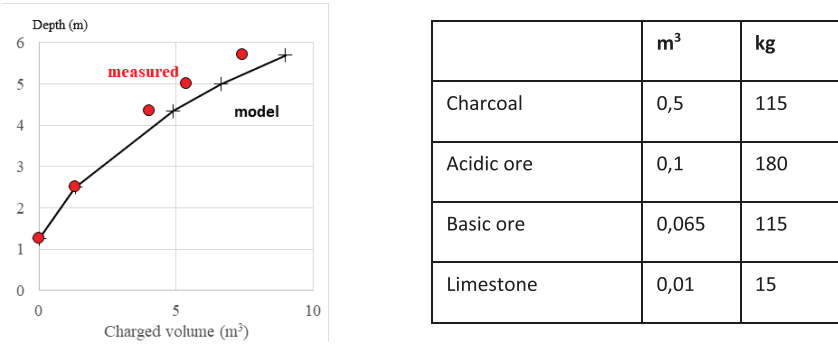


Fig. 7. Materials densities – material plug flow into the furnace.

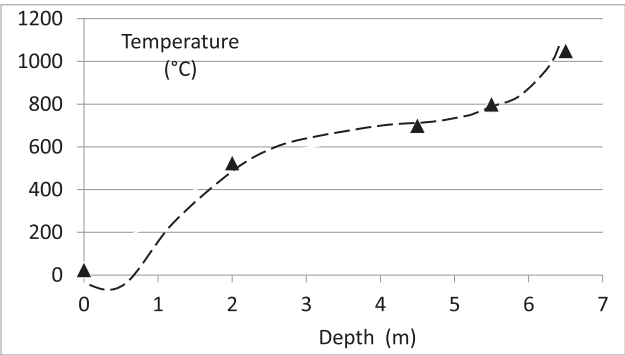


Fig. 8. Temperature vertical profile in the 1840 charcoal blast furnace.

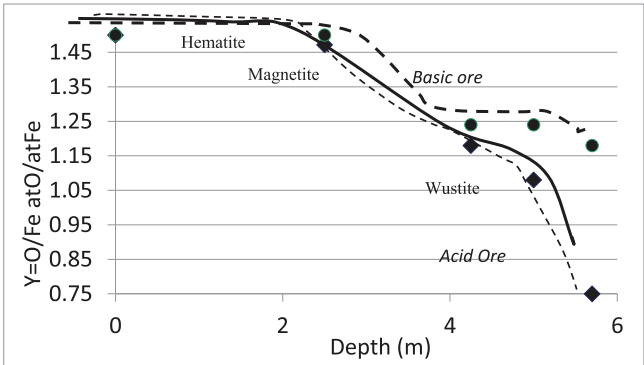


Fig. 9. Iron oxides reduction as a function of the height in the charcoal furnace- acid and basic ores (dotted curves) average reduction degree (bold curve).

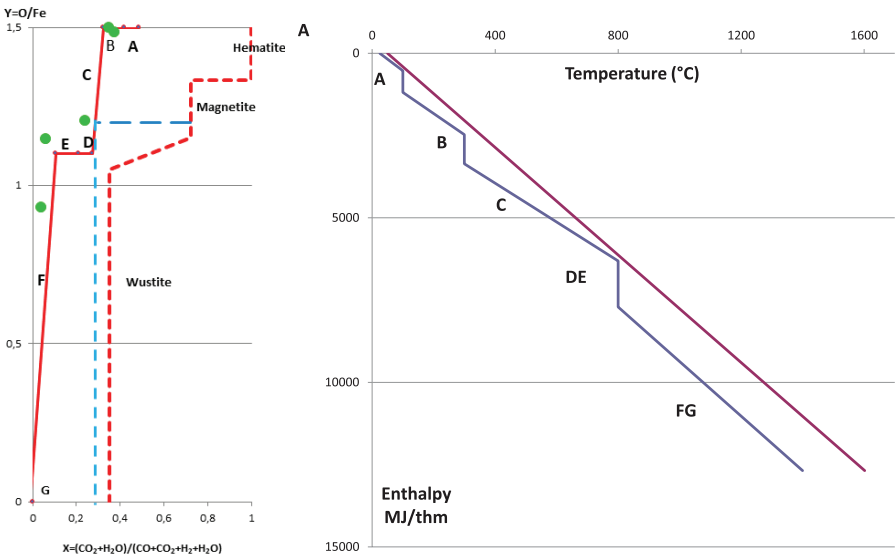


Fig. 10. The 1840 charcoal blast furnace as a dual heat and oxygen exchanger Rist and Reichardt diagrams.

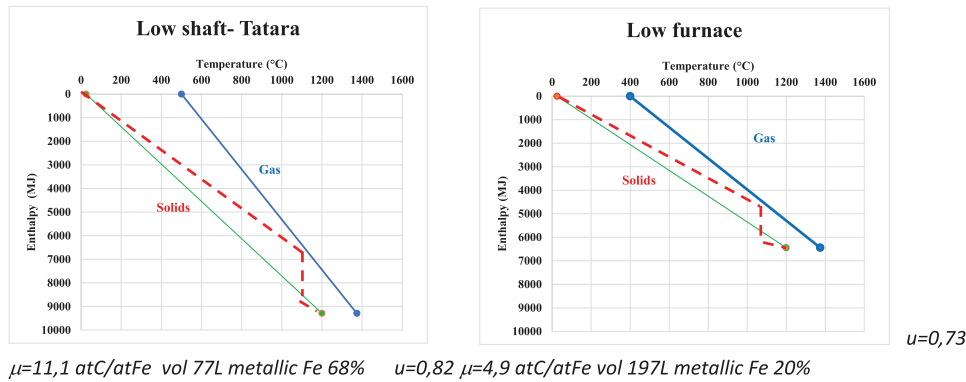


Fig. 11. Low shaft furnaces tentative Reichardt diagrams.

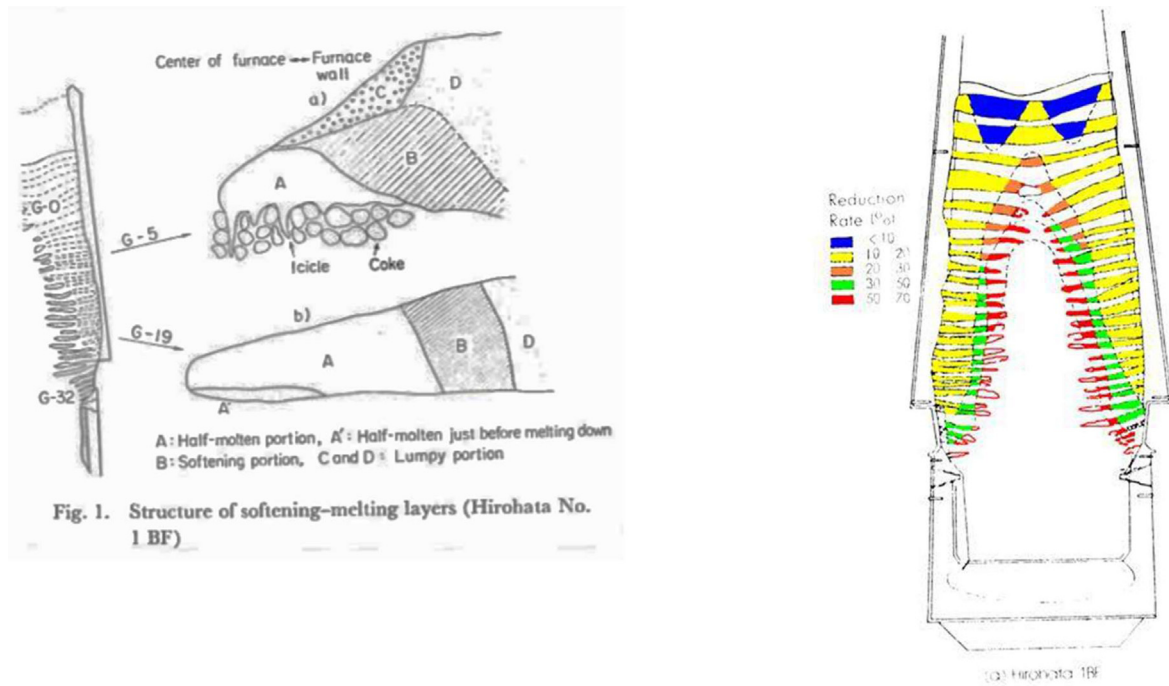


Fig. 12. Hirohata blast furnace short (axis) and long (walls) residence times [25].

residence time of the materials, for example by increasing the height of the shaft or increasing the radius of the belly.

The quenching of modern blast furnaces carried out in Japan in the years 1970–1980 clearly shows us the comparative evolution of reduction phenomena in the axis of the blast furnace where a large flow of gas heats the materials which reach fusion in a time of 1–2 h still with a strongly oxidized state and where the flowing molten liquid FeO is reduced on contact with coke forming stalactites of solid metallic iron.

On the contrary in the wall, where the gas flow rate is low, the high residence time of materials greater than 5 or 6 h allows their total reduction in iron and their partial carburization before melting.

The gradual increase in the height of the blast furnace shaft corresponds, from the point of view of the reduction of ores, to what is observed when moving from the axis towards the wall of this blast furnace at Hirohata [25] (Fig. 12).

To progress, it will be necessary to develop the low temperature reduction of the ore and reduce the hourly heat losses of the order of $20 \text{ MJ/m}^2/\text{h}$. Possible solutions might have been:

- Increase productivity by blowing more ... but it decreases the residence time.
- Increase the diameter of the furnace ... but with an increased number of tuyeres so that blast penetration might be a problem (oval like in China or rectangular furnaces like the Japanese Tataras may be a compromise).
- Increase the height of the furnace ... but the pressure drop might be too high for the blowers.

This is perhaps what the Chinese of the Warring States and the Han Dynasty were able to do, as Joseph Needham and D. Wagner suggest.

5 Conclusions

The physical–chemical model of the blast furnace developed by J. Michard and A. Rist can be fruitfully applied to the iron production tools of the past, in particular by describing them as counter-current heat and oxygen exchangers, between the hot gas produced by coal combustion at the tuyeres and the cold solids charged at the top. It may be applied from the blast furnace of 6000 m³ to the low shaft furnaces of a few tens of liters in volume.

Of course, the phase diagrams used by many archaeologists continue to provide valuable information on the composition of slags in relation with the ores, fuels and fluxes used as well as their melting temperatures.

Still more useful, the material and energy balances, especially as staged balances, make it possible to better understand the parametric space in which these tools have been operated and thus to better understand the constraints which limited the production and the quality of the product.

The most important concept is that of the residence time of materials, that is, the time that is allowed for the exchange of heat and oxygen. The measurements carried out on into a furnace with a 5 m height show a plug flow of materials down to the bosh.

The low shaft furnaces can be regarded as gasifiers with a thermal pinch at the bosh level. They are characterized by gas productions that are much greater than those required by the heat requirements and by the chemical needs of the burden, so that the top gas is very hot (close to 500 °C) and very little oxidized (mostly composed of CO and N₂). As a result of the high volume of gases, materials heat up rapidly to a temperature of around 1100 °C at which they are very partially reduced to iron and essentially melt in an oxidized state. Direct reduction of iron oxides by carbon, a very endothermic reaction but which, due to the small amount of energy available at this high temperature (due to the small difference in temperature between gas and solids at this level), only takes place very partially.

The increase of the furnace (height of the shaft in particular), will allow heating of the coal to a higher temperature at the tuyere level; a slightly longer residence time at low temperature will allow the development of reactions of indirect reduction of iron oxides (with very low thermal impact).

These low temperature reactions will be made easier by the existence of a medium temperature zone which can develop thanks to reactions such as:

- endothermal decomposition of iron hydrates and carbonates ... for example minerals based on goethite, siderite or limestone sedimentary ores;
- endothermal decomposition of limestone, calcium carbonate used as a flux;
- exothermal devolatilization of charcoal as suggested by Ebelmen gas analysis of the top gas containing CH₄ [15] and investigated in detail by Koderá and Kaiho [26];
- and as low temperature reactions develop, the endothermic Boudouard reaction is likely to occur, therefore leading to the modern blast furnace operation.

This study reveals lacks in our knowledge of the operation of these reactors of the past that this physicochemical approach, associated with measurements on today's simulators (temperature soundings, gas analysis, flow and pressure measurements, samples of materials at various levels in the shaft during their transformation, measurement of the rate of burden descent, etc.) would make it possible to complete while confirming the assumptions and data used for the modeling.

Reaction kinetics using TDA or thermobalance would validate the assumptions made on the temperature ranges where these reactions occur and assess their intensities.

Finally, this study shows the tremendous progress made from Antiquity to the present, both in the dimensions of iron production tools and of their performance in terms of productivity and energy consumption.

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