A MODEL FOR THE DIRECT REDUCTION PROCESS IN GAS-SOLID REACTORS [1]

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ABSTRACT

The Direct Reduction process accounts for about 90% of the DRI world production. This work presents some aspects of an uni - dimensional mathematical simulation of the Direct Reduction in gas-solid reactors, utilizing a model that takes into account the energy balance, iron oxide in lump and pellet forms, reduction kinetics and also the mass, heat and momentum transfer phenomena. The model was developed applying project operational data and laboratory reduction tests, aiming at a representation of the real operation parameters. For the gases it calculates the flowrate, temperature, pressure and composition from the injection to the exhaustion top. The solid phase compositional variations during the different stages of the reduction are also modeled.
In the future this model is expected to be validated in industrial furnaces like MIDREX and HyL.

Key words: Mathematical Modeling.
               Ironmaking.
               Direct Reduction.

■ INTRODUCTION

The sponge iron production suffered significant production acceleration in the last years. This has been so not only due to the technological and economic development experienced by the principal processes of DRI/HBI production but also because of the problems generated with the increasing presence of contaminants in the iron scraps. From 1990 to 2000, the production of DRI grew more than 100%, jumping from 17.68 to 43.2 million metric tons. In the year 2000 the data for the distribution by process of the sponge iron world production was the following [1]

|		
MIDREX	67%	
HyL (I / III)	23%	
SL/RN	3%	
Other	7%	(coal : 5% / gas : 2%)

Today the shaft furnace direct reduction process account for circa 90% of the world's sponge iron production. The demand for this iron product has been fundamental for the continuous optimization of its production operational conditions, mainly aiming at the attainment of higher production and metallization indexes. To reach such goals much has been done by the close collaboration between the mining companies and sponge iron production companies, and further, it is in this context that this work finds its objective. It attempts to simulate the iron oxide reduction with a phenomenological approach and hosts a technical cooperation between the authors.

Important initiatives in turning the modelling of the process direct reduction processes more pragmatic are not new. Yanagiya et alii [2], assuming moving beds, used the reducing kinetic rate parameters in their uni-dimensional model. Also adopting a unidimensional mathematical formalism Takahashi et alii [3] simulated conditions of moving beds in pressurized ambient bringing a more realistic insight for these processes. Further, Yagi et alii [4], conferring porosity to the moving beds broadened the scope of the reduction phenomena analysis to the pressurized reactors.

Looking for the aggregation of a wider range of laboratory information to this scene this work has the mathematical simulation of the direct reduction zone of the shaft furnace as its principal aim. The model utilizes not only the classical differential and global mass and energy balances combined to the usual iron oxides - in the form of pellets and lumps - reduction kinetics linked with the mass, momentum e heat exchange but also technical data from standard essays on the iron ores. Beyond that, in the search of more pragmatic evaluation of the calculated results, the model has a software architecture capable of managing the wide range of calibrations and validations occurring in practice.

■ THE MODEL

The model macro-structure is pictured in figure 1. It is a classical counter-current system constituted by a particle bed and a reactive gas, simulating the reduction zone of a shaft reactor.
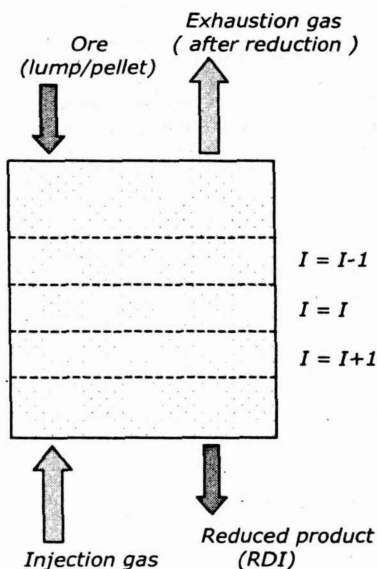
Figure 1.: Reduction Zone

As in this illustration, the reduction zone is criteriously segmented in layers by the model. Further the reactor parameters - like geometry and type - and incoming material flow characteristics - like the temperatures and composition for the injection and exhaustion gases, solids chemical composition and laboratory tests data - together with boundary conditions are considered. Then sequentially, each of the layers, where all the reducing phenomena that are suppose to take place for the pellets and/or lumps, are solved by the model.

As stressed, since the phenomena occurring in the reactor has to obey boundary conditions and is characteristically counter-current, an iterative algorithm are not seldom necessary.

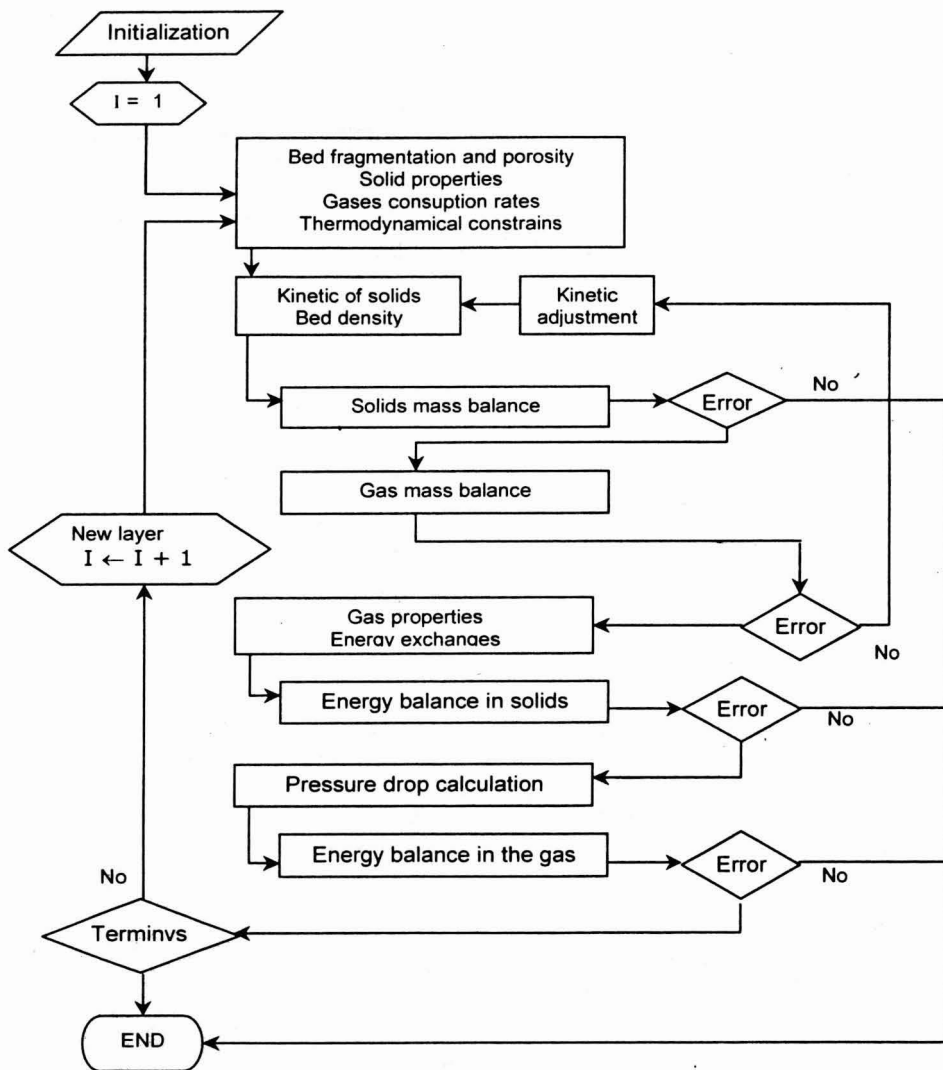The basic calculation procedure is diagrammatically represented in figure 2.

Figure 2. : The model calculation flux

It must be said that the usual balance and fluidynamic equations utilized in proposed model are extended to take into account important factors like ...

— Solid fluxes;
— Residence time;
— Particle porosity;
— Thermodynamic restrictions;
— Reducibility tests of metallic raw materials under various conditions (pellets and lumps);
— Degradation essays (fines generation) according to HyL or MIDREX standards;
— Bed experimental densities for pellets and lumps mixtures and
— Multi-component bed porosity.

In the sake of completeness some highlights of the mathematics beneath the model are given.

For the solids mass balance the key equation was

$$\{M\}_{(l/p),i-1} = \frac{\rho_{L,(l/p),i-1}}{100}\{S\}_{(l/p),i-1} \mid \{S\}_{(l/p),i-1} \equiv \left(S^h, S^m, S^w, S^f, S^g\right)_{(l/p),i-1}$$

Where $\{M\}_{(l/p),i-1}$ is the mass per volume vector (in kg / m^3 of bed), $\rho_{L,(l/p),i-1}$ the bed density and $\{S\}_{(l/p),i-1}$, the weigh percentages for the oxides that enter in layer (i). To evaluate the total variation of mass in each component of the layer the following linear system for $\Delta^T M_{(l/b),i}$ is to be solved ...

$$\Delta^T M_{(l/p),i} - \left\langle \{\Delta M\}_{(l/p),i}, 1 \right\rangle = 0 \mid \Delta M_{(l/p),i} = \begin{bmatrix} \Delta^h M \\ \Delta^m M \\ \Delta^w M \\ \Delta^f M \\ \Delta^g M \end{bmatrix}_{(l/p),i}$$

$$\Delta^T M_{(l/p),i} = C_O^0 \times \left(f_{R,(l/p),i} - f_{R,(l/p),i-1}\right) \times \left(\frac{M_{O_2}}{2}\right)$$

where $\Delta^T M_{(l/b),i}$ is the total mass variation, in terms of kg of $1/2 O_2$ / m^3 of bed, $f_{R,(l/p),i}$ is the rate of reduction of the solids (for the lump as well as for the pellets) in layer "i" , M_{O_2} is the O_2 molecular mass and C_O^0 the initial oxygen concentration in the bed (mol-kg of $1/2 O_2$ / m^3 of bed)

It should be noted that these losses are related to the various oxidation states of iron, which are functions of the oxygen losses.

For the gases a similar balance is made. But here there is a remark, the gas flowrate occurs from the bottom of the reduction zone to the top, in counter-current, thus from layer "i" to "i+1" there is a growth in the reducing species content. Calling $\{ \Delta G \}_i$ the percentage variation vector for the reacting gases the following equation regulates the mass balance ...

$$\{\Delta G\}_i \equiv \begin{bmatrix} \Delta^{CO}G \\ \Delta^{H_2}G \\ \Delta^{CO_2}G \\ \Delta^{H_2O}G \\ \Delta^{N_2}G \end{bmatrix}_i = \Psi \times \left[\begin{pmatrix} \Lambda_{CO} \times C_O^0 \times \Delta f_R \\ \Lambda_{H_2} \times C_O^0 \times \Delta f_R \\ 0 \\ 0 \\ 0 \end{pmatrix}_l + \begin{pmatrix} \Lambda_{CO} \times C_O^0 \times \Delta f_R \\ \Lambda_{H_2} \times C_O^0 \times \Delta f_R \\ 0 \\ 0 \\ 0 \end{pmatrix}_p \right]_i$$

where $\Delta f_{R,(l/p),i} \equiv f_{R,(l/p),i} - f_{R,(l/p),i-1}$ and Λ_{CO} e Λ_{H_2} are the total relative consumption of CO and H_2.

The Ψ factor is : $\Psi = \dfrac{100 \ V_{Mg} \ V_C}{F_{gN} \ \tau}$

being v_{Mg} the gases molar volume (m^3 / mol-kg), V_C the layer volume, F_{gN} the normal-volumetric flowrate of the gases (Nm^3/h) and τ is the solids total residence time in the reducing zone (h).

In order to determine the temperature $T_{(l/b),i}$ in which the solids leave layer "i" the system solves the energy balance ...

$$S_{(l/p)}(i) = S_{(l/p)}(i-1) + E_{conv,(l/p),i} + E_{reaction,(l/p),i} + E_{exchange,(l/p),i}$$

The terms in this equation E_{conv}, $E_{reaction}$ e $E_{exchange}$, represent the convection, reaction and enthalpic exchange heats (in kcal), respectively. Explicitly, it means ...

$$S_{(l/p)}(i) = \rho_{L,(l/p),i} \ V_C \left[\frac{1}{100} \sum_{j = Fe_2O_3, Fe_3O_4, FeO, Fe^o} \frac{S_{(l/p),i}^j}{M_j} \int_{T_{c,s}}^{T_{(l/p),i}} c_{P,j} \times dT \right.$$

$$\left. + L^{TrF}_{Fe_2O_3, <T_{c,s}, T_{(l/p),i}>} + L^{TrF}_{Fe^o, <T_{c,s}, T_{(l/p),i}>} \right]$$

Note that the unknown is $T_{(l/b),i}$ that represents the temperature in which the solids leave layer "i". $\rho_{L,(l/b),i}$ (kg / m^3 of bed) is the bed density, V_C the layer volume (m^3),

$S^j_{(l/b),i}$ the weight percentages of the charge components (lump/pellet), M_j e $c_{P,j}$ the molecular mass and specific heat of the "j" oxide, $T_{c,s}$ a reference temperature, and $L^{TrF}_{X,<Ti,Tf>}$ the phase transformation heat for component "X" in kcal / kg-mol.

As for the gases the energy balance aims at the determination of the temperature $T_{g,i+1}$ in which the gas enters layer "i" coming from layer "i+1" , obviously using the same $T_{c,s}$ as reference and it reads :

$$G(i+1) = G(i) + \sum_{k=l,p} [S_k(i) - S_k(i-1) + E_{reaction,k,i}] .$$

where $S_k(i)$ is the same function as above and $G(i)$ is :

$$G(i) = m_g(i) \left(\frac{1}{100 \times M_{mix,i}} \sum_{j=CO,CO_2,H_2,H_2O,N_2} G^j_i \int_{T_{c,s}}^{T_{g,i}} c_{P,j}\, dT \right)$$

G^j_i is the weight percentages os the gases in layer "i", $c_{P,j}$ the gas "j" specific heat, $M_{mix,i}$ the molecular weight of gas mixture. More, the $m_g(i)$ function measures the total gas mass that bathes layer "i" (in kg) during the residence time τ and values:

$$m_g(i) = \rho_{g,i} \left(F_{gN}\, \tau \right) \left(\frac{1}{p_i} \right) \left(\frac{T_{g,i}}{273} \right) .$$

where $\rho_{g,i}$ is the gas density (kg / m³), F_{gN} it's flowrate (Nm³ / h) and p_i is it's total pressure(atm).

■ **RESULTS**

The model software furnishes the output printing like the one presented in figure 3, generating relevant technological information about the performance of the simulated processes.

As the software accommodates in its many input menus data directly obtained from laboratory essays, it can play an important role in solving operational problems. It goes without saying that it could also be a valuable instrument in the sensibility analysis for the key parameters linked with the quality of the metallic raw materials.

As an example, figures 4 and 5 present two case studies.

Figure 4 compares to the one in figure 3 but using the injection pressure, increased to 3 atmospheres. As shown in the same figures productivity is increased by circa

2%, coherently the gas specific consumption dropped by the same amount and the pressure drop decreased 27 %.

Top gas temperature	353°C
Injected gas temperature	945
Injection pressure	2.54 atm
Pressure drop (Red.Zone)	1.44
[CO]injected	29.1 %vol
[CO2]	1.9
[H2]	58.8
[H2O]	5.2
H2/CO ratio	2.02 vol/vol
[CO]top	17.5 %vol
[H2]	32.4
H2/CO ratio	1.85 vol/vol
Volume of injected gas	61 000 Nm³/h
Gas specific consumption	1 413 Nm³/t Fe
Charging rate	60.00 t/ h
Residence time	3 h 11 min
Pellet rate (pel/lump)	1.2 %
Metalization	90.7 %
Fines in sponge iron (-6.35 mm)	8.2

Production 39.86 t sponge iron / h
Productivity 11.80 t sponge iron / m³ / day
(50.72 t sponge iron / RZ m² / day)

Figure 3. : Model output

Top gas temperature	363°C
Injected gas temperature	956
Injection pressure	3.05 atm
Pressure drop (Red.Zone)	1.05
[CO]injected	30.8 %vol
[CO2]	0.2
[H2]	56.5
[H2O]	7.5
H2/CO ratio	1.83 vol/vol
[CO]top	17.4 %vol
[H2]	30.9
H2/CO ratio	1.78 vol/vol
Volume of injected gas	61 000 Nm³/h
Gas specific consumption	1 383 Nm³/t Fe
Charging rate	61.35 t/ h
Residence time	3 h 7 min
Pellet rate (pel/lump)	1.2 %
Metalization	91.2 %
Fines in sponge iron (-6.35 mm)	8.2

Production 40.71 t sponge iron / h
Productivity 12.05 t sponge iron / m² / day
(51.81 t sponge iron / RZ m² / day)

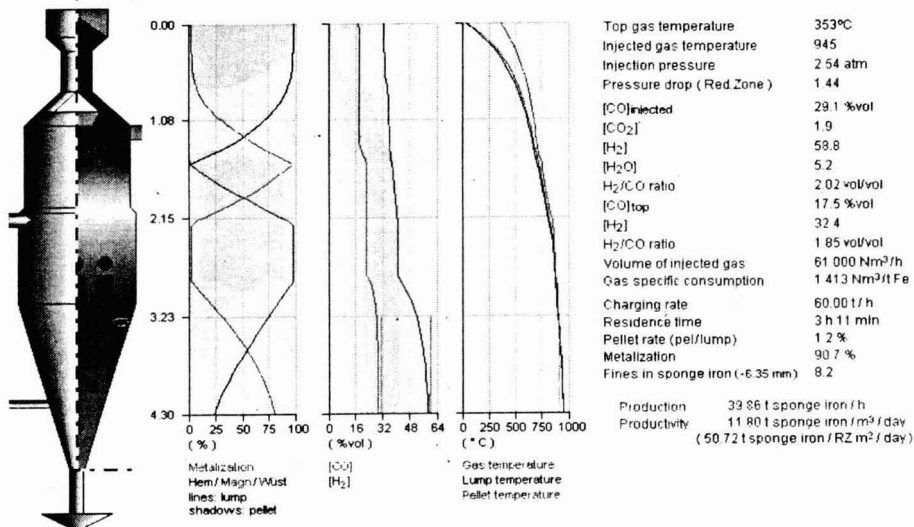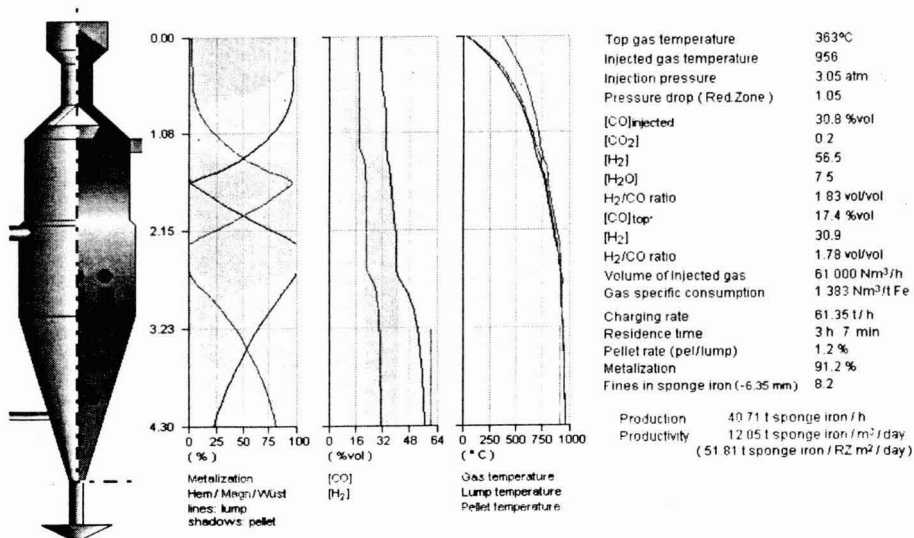**Figure 4. : Simulation for a gas injection pressure of 3 atmospheres**

For figure 5, the conditions are the same as prevailing in the first simulation except for the furnace's charge, that has a much higher pellet rate, namely 4.0 . As expected for the same level of DRI metalization the productivity increased from 11.80 to 13.38 t / m³ / day. The gas specific consumption accordingly decreased 12% approximately.
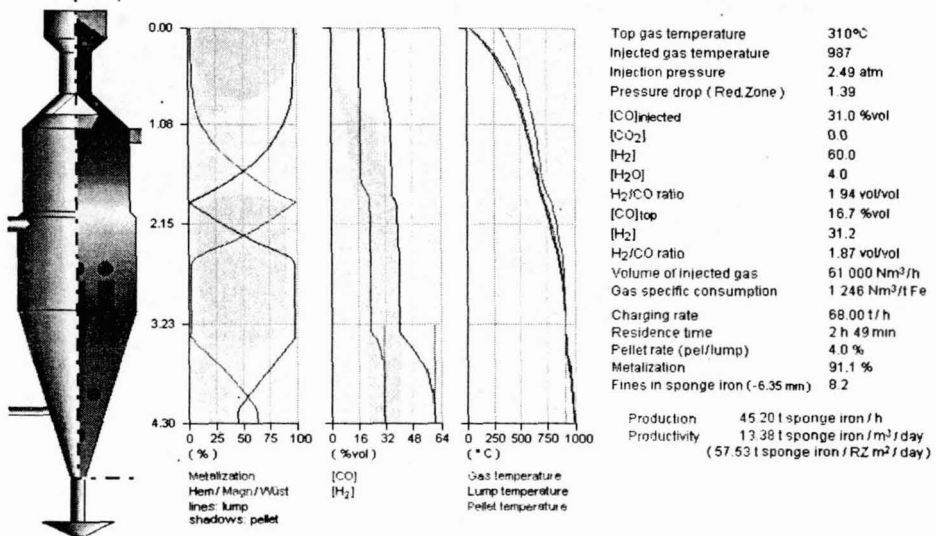

Top gas temperature	310°C
Injected gas temperature	987
Injection pressure	2.49 atm
Pressure drop (Red.Zone)	1.39
[CO]injected	31.0 %vol
[CO2]	0.0
[H2]	60.0
[H2O]	4.0
H2/CO ratio	1.94 vol/vol
[CO]top	16.7 %vol
[H2]	31.2
H2/CO ratio	1.87 vol/vol
Volume of injected gas	61 000 Nm³/h
Gas specific consumption	1 246 Nm³/t Fe
Charging rate	68.00 t / h
Residence time	2 h 49 min
Pellet rate (pel/lump)	4.0 %
Metalization	91.1 %
Fines in sponge iron (-6.35 mm)	8.2
Production	45.20 t sponge iron / h
Productivity	13.38 t sponge iron / m³ / day
	(57.53 t sponge iron / RZ m² / day)

Figure 5. : Simulation for a pellet rate of 4.0 (w/w)

■ **CONCLUSIONS**

The present paper permitted the following general conclusions :

i. The mathematical simulation furnished by the model succeeded in its analysis of comparative performance for different iron burdens (pellets and lumps);

ii. The treatment achieved by the model for key operational parameters, like top pressure and pellet rate, showed that its simulation can be used as a valuable tool for the optimization of important DR processes;

iii. The adopted software architecture showed enough flexibility to insure its utility in further calibrations and validations.

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