An analysis of pulverized coal injection in blast furnaces by rist diagram

Uma análise sobre a injeção de carvão pulverizado em altos-fornos por meio do diagrama de rist

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ABSTRACT
The reduction process in blast furnace involves the higher cost of a steel company. The operational stability of the reduction process should be accompanied by low fuel rate and a good productivity. Thus, it is extremely important to control the main parameters of this process as the ratio between the direct and indirect reduction, the efficiency of the stack, the temperature of the Thermal Reserve Zone and energy balance of the Elaboration Zone. This analysis was performed using the Rist diagram, to assess the operational changes of operation All Coke in relation to the use of pulverized coal injection in a blast furnace 1 at ArcelorMittal Tubarão. The Rist diagram was effective in to relate the parameters studied with these operational changes.

Keywords: Blast furnace, Ironmaking, Rist diagram, Pulverized coal injection.

RESUMO
O processo de redução em alto-forno envolve o maior custo de uma siderúrgica. A estabilidade operacional do processo de redução deve vir acompanhada de baixo consumo de combustível e boa produtividade. Assim, é extremamente importante controlar os principais parâmetros deste processo como a relação entre a redução direta e indireta, a eficiência da chaminé, a temperatura da Zona de Reserva Térmica e o balanço energético da Zona de Elaboração. Essa análise foi realizada por meio do diagrama de Rist, para avaliar as mudanças operacionais da operação All Coke em relação ao uso da injeção de carvão pulverizado em um alto-forno 1 na...
1 INTRODUCTION

Despite its ancient origin, the production of pig iron in blast furnaces has remained competitive in relation to the alternative processes of primary iron production. Several limitations inherent to the process were gradually supplanted through ingenious modifications like, for example, the agglomeration of iron oxides, injection of ancillary reducers in the tuyeres and use of computers for process analysis.

However, in recent years, due to the appearance of new technologies for the manufacture of pig iron, variations in prices and in the quality of raw materials and restrictions in the emission of pollutants, the use of models for a more precise control of the process has become increasingly important.

The Rist diagram is a model that incorporates the main characteristics of a blast furnace's operation, illustrating solutions to various problems mass and heat balance in stationary state, relating the level of oxidation of gas and of the load. This diagram can be used to understand changes in the operational parameters caused by process modifications. In addition, the Rist diagram can be used through an interaction with an expert, for operation projection [1]–[3].

The main parameters evaluated by the Rist diagram are proportion between direct and indirect reduction, efficiency of the furnace stack, temperature variation of the Thermal Reserve Zone and the energy balance of the Elaboration Zone. These parameters are associated with fuel rate, productivity and blow composition [4].

The Rist diagram has been widely used to determine the variation of operational parameters in blast furnaces when there are variations in the process. An example of this is the work of Ariyama et al. [5] in which the diagram is used to determine the operating conditions of the blast furnace in relation to the modifications proposed for the process to reduce CO₂ emission or the study by Kasai and Matsui [6] that evaluated the influence of the reactivity of coke in reducing the Thermal Reserve Zone Temperature.

With the aim of having more restricted control of the reduction process, a study was developed based on concepts of the Rist diagram to evaluate the main parameters involved in this process. For this, the operational variations of an All Coke process were analyzed in relation to use of Pulverized Coal Injection (PCI), at two different periods of Blast Furnace 1 of ArcelorMittal Tubarão.
ArcelorMittal Tubarão. These variations will be determined through characteristics of the top gas, blow, charge and pig iron composition.

2 MATERIALS AND METHODS

2.1 RIST DIAGRAM

The Rist diagram is based on the thermochemical balance of the main elements involved in the reduction reaction. It has some restrictions, since it does not consider factors like distribution and permeability of the charge, production speed or blow depth.

This diagram is a mathematical model that uses a mass and energy balance to graphically represent the Blast Furnace operation. It makes use mainly of the carbon, oxygen and hydrogen balance involved in the formation of reducer gas, illustrating the conditions in which the oxygen is transferred from the charge to the gas.

In the diagram, the mass balance for the three elements cited above is preferable and must be done as a priority once these are involved in:

- the formation of reducer gas through reactions at high temperatures (combustion reaction, direct reduction of iron oxides and non-ferrous oxides); [1], [7]
- in the use of reducer gas through indirect reduction of iron oxides. [1], [7]

The axes represent the formation of reducer gas according to the sources of oxygen. The x axis describes the fraction of the contribution in the formation one mol of reducer gas, equation 1. This axis can be divided into: combustion reaction in the tuyeres \( (X_b) \), of metalloid reduction \( (X_f) \) and of the solution loss reaction \( (X_{sl}) \) (Figure 1). While the y axis represents these fractions in relation to the formation of one mol of iron, that is, the quantity of reducer gas required to form one mol of metallic iron, \( \mu \), equation 2.

\[
X_b + X_f + X_{sl} = 1 \quad \text{(mol of reducer gas)} [1], [7]
\]

\[
Y_b + Y_f + Y_{sl} = \mu \quad \text{(mol of reducer gas / mol of Fe)} [1], [7]
\]

In relation to the sources of oxygen, we have that the positive part of the y axis represents the level of oxidation of the iron ore, that is, the oxygen from the ore. And the negative part is the quantity of oxygen that comes from the blow and reduction of the non-ferrous oxides.
The origin of the y axis in the diagram is arbitrary. By convenience, a y axis can be chosen such that the oxygen originally combined with the iron (YSi + Yi) appears on the positive side and the other sources of oxygen and hydrogen appear on the negative side.

The x axis can be divided into two parts, that of reducer gas formation and that of indirect reduction, its consumption [1], [7]:

- formation of reducer gas: corresponds to the interval of 0 < x < 1, in which the formation of reducer gas is due to the "solution loss" reaction, reaction of non-ferrous oxides and reaction of the blow combustion; and
- indirect reduction: corresponds to the interval of 1 < x < 2, in which there is reducer gas consumption without its replacement.

Through these divisions, we can form the AE, GW and UV segments (Figure 1). The main segment is called Operational Straight Line, AE. This segment is intercepted by auxiliary lines that represent the level of reduction of the charge on reaching the Elaboration Zone, GR segment, and the energy balance of the Elaboration Zone, UV segment.

The Operational Straight Line illustrated in Figure 1 through segment AE graphically represents the participation of the carbon, oxygen and hydrogen elements, through mass balance, in formation of the reducer gas and in its use through indirect reduction. The BD, DP and PE segments confined in the interval of 0 < X < 1 represent the formation of reducer gas and the AB segment confined in the interval of 1 < X < 2 corresponds to the use of this through indirect reduction of iron oxides [1], [7].
2.2 AE SEGMENT CALCULATION

The operational straight line has two important groups of properties associated with indirect reduction in the furnace stack, point ω, and with thermal balance of the Elaboration Zone, point P.

The Operational Straight Line is determined by points A and E. Point A is determined by oxidation of the outlet gases, coordinate $X_A$, and by the level of oxidation of the metallic charge, coordinate $Y_A$. Coordinate $X_A$ is calculated from the composition of gases in the outlet at the top of the blast furnace, according to equation 3. Its composition is given in percentage.

$$X_A = 1 + \frac{(H_2O + CO_2)}{(CO_2 + CO + H_2 + H_2O)}$$  \hspace{1cm} (3)

Coordinate $Y_A$ is determined by the composition of the metallic charge, in relation to the total Iron content, %TFe (Hematite plus the quantity of Wustite in the metallic charge), and the percentage of FeO. The level of oxidation is determined by the quantity of oxygen contained in the ore in relation to one iron atom, that is, the percentage of iron oxides times its quantity in mol of oxygen in relation to one mol of iron (1.5 for Hematite and 1.05 for Wustite), according to equation 4.
Point E is determined by coordinates $X_E$ and $Y_E$. Coordinate $X_E$ is by convention equal to zero, and coordinate $Y_E$ represents the quantity of reducer gas produced in the combustion reaction in the tuyeres, $Y_b$, plus the quantity of reducer gas due to direct reduction of the metalloid $Y_f$. Thus, we have: \[1\], \[7\]

\[X_E = 0 \quad (5)\]
\[Y_E = -Y_b - Y_f \quad (6)\]

Coordinate $Y_f$ is dependent on the chemical composition of pig iron, that is, the percentage mass of Si, Mn and P in the pig iron. Thus, the quantity of CO formed by the reaction of direct reduction of non-ferrous oxides per mol of iron in one ton of pig iron ($\text{MolFe}$) is represented by:

\[
Y_f = \frac{(%\text{Si} \cdot 10 \cdot \frac{2}{28.09} + %\text{Mn} \cdot 10 \cdot \frac{1}{54.94} + %\text{P} \cdot 10 \cdot \frac{5}{30.97} \cdot \frac{1}{2}) \cdot 1000}{\text{MolFe}} \quad (7)
\]

The term $Y_b$ is determined by the quantity of CO and $\text{H}_2$ formed in the tuyeres in relation to the quantity in mol of iron for one ton of pig iron.

\[
Y_b = \frac{(\text{MolCOVent} + \text{MolH2Vent})}{\text{MolFe}} \quad (8)
\]

With these coordinates, one can outline segment $AE$ and determine some quantities as is being represented in the diagram of Figure 1. With segment $AE$, we can determine other points like the coordinates that represent Indirect Reduction ($X_i, Y_i$) and Direct Reduction, point ($X_{SL}, Y_{SL}$), since we already know that in the interval of $1 < x < 2$ corresponds to the dominion of reducer gas consumption. \[1\], \[7\]:

The slope of straight line $AE$, $\mu$, indicates the number of reducer gas mols (CO and $\text{H}_2$) required to produce one mol of iron \[1\], \[7\].
2.3 GW SEGMENT CALCULATION

The GW segment represents a mass and energy balance of the Preparation Zone. The mass balance is relative to the level of oxidation of the metallic charge when it passes to the Elaboration Zone, Point R, and the energy balance refers to this balance temperature (Figure 1).

This segment is calculated in function of the top gas composition and of the level of oxidation of the metallic charge. It represents, when intercepted with the operational straight line, the level of oxidation of the metallic charge when it passes to the Elaboration Zone, point R of Figure 1. This point is determined by intersection of segment GW with AE.

Point G is determined by the level of oxidation of the metallic charge in charging of the blast furnace, we therefore have that \( Y_G = Y_A \) and since there is no direct reduction in the Preparation Zone \( X_G = 1 \) [1], [7].

Point W is determined by coordinates \( Y_W \) and \( X_W \). By definition, \( Y_W \) is equal to 1.05, molar ratio between oxygen and iron in the Wustite.

\( X_W \) corresponds to the level of oxidation of the gases in the beginning of the Elaboration Zone. The composition is dependent on the coke's reactivity temperature, which according to literature TR=950°C and on the composition of gases in this temperature, equation 9.

\[
X_W = 1 + \left[ \frac{\text{CO} + \text{CO}_2}{1 + K_{10}} \right] + \left[ \frac{\text{H}_2 + \text{H}_2\text{O}}{1 + K_{11}} \right] \tag{9}
\]

Where \( K_{10} \) and \( K_{11} \) are balance constants of reactions 10 and 11 respectively [6]

\[
\text{FeO} + \text{CO} \leftrightarrow \text{Fe} + \text{CO}_2 \hspace{1cm} \Delta G^\circ = -5454.55 + 5.80 \cdot T \tag{10}\text{ (cal/mol)}
\]

\[
\text{FeO} + \text{H}_2 \leftrightarrow \text{Fe} + \text{H}_2\text{O} \hspace{1cm} \Delta G^\circ = 1834.93 - 1.01 \cdot T \tag{11}\text{ (cal/mol)}
\]

Point R is determined by the intersection of the straight line AE with segment GW, and it determines the level of oxidation of the iron ore, as shown in Figure 1.

Through this point, one can determine the Rist deviation, the Omega Factor. This calculation is determined by the difference between the level of oxidation of the ore in the O/Fe axis of point R and of point W, which is that of wustite, 1.05 [1], [7]:

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This segment has direct relation with the Fe-C-O system, that is, the O/C coordinates of the Rist diagram represent the respective ratios $\frac{P_{CO}}{P_{CO} + P_{CO2}}$, of the FeO-Fe balance, associated with points R and W presented in Figure 1.

The efficiency of the furnace stack ($\eta$) represents the quantity of iron oxide that is reduced to wustite up to the Elaboration Zone, that is, indicates the extent of the indirect reduction in the furnace stack. The greater the efficiency of the furnace stack, the lesser the extent of the direct reduction reaction and consequently the lesser the energy consumption of the blast furnace. The furnace stack’s efficiency can be determined by the ratio between segments RG and GW [1], [7]:

$$\eta = \frac{RG}{GW}$$  \hspace{1cm} (13)

The temperature of the start of coke reactivity has direct relation with the composition of the gases, as shown in equation 9. And through Figure 1, we could relate this temperature with the Rist deviation and efficiency of the furnace stack.

For a value of $Y_E$, the greater the Rist deviation, the less the furnace stack efficiency and the greater the specific consumption of coke. This becomes clear because, for a same value of $Y_E$, entry of reducer gas in the tuyeres per mol of iron, there is an increase in the value of $\mu$. This increase is proportional to the direct reduction of the blast furnace in response to the increase in temperature of the Thermal Reserve Zone.

2.4 UV SEGMENT CALCULATION

The properties of the operation line associated with thermal balance of the Elaboration Zone is represented by point P. This point is determined by the intersection of the operational straight line with segment UV.

The UV segment is constructed from a thermal balance of the combustion reactions, solution loss and other thermal needs of the Elaboration Zone like heating and melting of the charges and thermal losses.

The energy balance of the furnace is done as follows [1], [7]:

$$q_b \cdot Y_b = q_{SL} \cdot Y_{SL} + \theta$$  \hspace{1cm} (14)
The quantity $q_b$ represents the heat supplied by the blow and $q_{SL}$ the heat required by the Boudouard reaction. Variable $\theta$ quantifies the heat required by the Elaboration Zone minus the solution loss reaction, and its value is determined indirectly through point P. Thus, this variable represents the other thermal needs of the Elaboration Zone [7].

The variables that represent the quantity of heat, $q_b$ and $q_{SL}$, refer to a mol of reducer gas. While variable $\theta$ does not have this reference.

Point P divides segment UV into two regions in which its length corresponds to the quantity of heat required by the direct reduction in relation to the heat supplied by the blow. And this point divides segment UV in the ratio $PU/PV = q_{SL}/q_b$ [1], [7].

The operational straight line turns around P under influence of factors that affect the quantity of solution loss without altering other thermal requirements of the Elaboration Zone. The coordinates of point P, in relation to one mol of reducer gas are:

\[
X_P = \frac{q_{SL}}{q_{SL} + q_b} \\
Y_P = \frac{\theta}{q_{SL} + q_b}
\]  

(15)  

(16)

It is interesting to note how much the Boudouard reaction is endothermic, this can be compared when, by simple inspection in Figure 1, we compare the UP segment with the PV.

3 RESULTS AND DISCUSSION

Through the data of each period, their mean values and their deviations were determined for each input variable of the model. Table 1 gives the values for the period in which the PCI is not used, called All Coke operation and the data for the period in which the pulverized coal injection is used.

With this data, it was possible to determine the Rist diagram that compares the two forms of operation of Blast Furnace 1 of ArcelorMittal Tubarão. Table 2 contains the points of each segment used in determination of the diagram of Figure 2.

| Input Variables                  | All Coke Operation | Operation with PCI |
|----------------------------------|--------------------|--------------------|
|                                  | Mean       | Mean deviation | Mean       | Mean deviation |
| Blow Volume (Nm$^3$/min)         | 7134.92    | 73.732      | 6308.16    | 62.657        |
| Blow Air Temperature (°C)        | 1142.94    | 7.852       | 1246.79    | 0.923         |

Table 1. Input variables for the All Coke operation with PCI.
Due to the different forms of operation, we have some variables in the values of the input variables. The use of PCI increases pig iron productivity significantly with practically the same quantity of Fuel Rate.

The efficiency of the operation with PCI depends on the parameters that are influential in the yield of its combustion, like availability of oxygen and quantity of heat. For this, the period in which the PCI is used shows an increase in the enriching of oxygen and blow temperature and reduction of the blow humidity.

| Rist Points | All Coke Operation | Operation with PCI |
|-------------|-------------------|-------------------|
| XA          | 1.464             | 1.485             |
| YA          | 1.452             | 1.463             |
| YE          | 1.466             | -1.494            |

Table 2. Rist Points for the All Coke operation and with PCI.
Points X_E, Y_E, X_W, X_U and X_V of Table 2 have mean deviation equal to zero because they are two fixed points in the Rist diagram.

Figure 2. Rist diagram comparing the two forms of operation.

Figure 2 was divided into four regions for better viewing of each part of the segments. The points presented in the diagrams below have indices 1 and 2. Index 1 corresponds to the All Coke operation and 2 to the PCI operation.

Figure 3 presents the quantity of direct reduction for each form of operation, points B_1 and B_2. Figure 4 presents the region of segment GW and points R_1 and R_2, the Figure 9 and Figure 10 compares these points with increase in temperature of the Thermal Reserve Zone, points T_1 and T_2. Figure 11 shows the thermal balance of the Elaboration Zone through points V_1 and V_2.
Direct reduction of the *All Coke* operation, point B₁, is greater than that in the operation with PCI, B₂. This is represented by the intersection of segment AE with the vertical segment that passes through X=1, Figure 3. The Rist deviation, under operating conditions is show in Figure 4 by segment GW.

![Figure 3. Points B1 and B2. Measurement of the extent of direct reduction for each point.](image)

The graph of Figure 5 indicates the percentage daily direct reduction determined by the model. The direct reduction was calculated by dividing coordinate Y_B by coordinate Y_A and expressing in percentage. According to Figure 5, the percentage direct reduction in the first period is greater than in the second.

![Figure 4. Comparison between R1 and R2 for both forms of operation.](image)
Figure 5. Percentage direct reduction calculated daily for each form of operation.

![Direct Reduction Graph](image)

Figure 6. Level of oxidation of the top gases calculated daily for each form of operation.

![Top gas oxidation Graph](image)

Table 1 shows that there is a small variation in the Fuel rate, with a significant increase in the production of pig iron. This is due to the higher direct reduction rate in the All coke process. Direct reduction occurs with greater specific consumption of the reducer, since part of it is used to generate heat due to the Boudouard reaction.

Oxidation of the top gas is an indication of the increase in indirect reduction proportion. Figure 6 shows that the All Coke process has less oxidation of the top gas due to greater amount of direct reduction. Thus, the greater the extent in which the gasification, the greater the gas regeneration and, consequently, the less its top gas oxidation from the top.

The Rist deviation and Efficiency of the furnace stack are calculated in Figure 7 and 8 respectively. In them, it becomes evident that the process in which there is use of PCI has its metallic charge reduced, with greater efficiency in the Preparation Zone.
Figure 7. Rist Deviation determined by the model.

Rist Deviation

Figure 8. Furnace stack Efficiency determined by the model.

Furnace stack efficiency

The furnace stack efficiency reduction of the *All coke* process indicated in Figure 8 is accompanied by increase in temperature of the Blast Furnace's Thermal Reserve Zone. This increase is presented in the graph of Figure 9 and Figure 10 by the points $T_1$ and $T_2$. In response to this increase in temperature, there is a proportional increase in quantity of direct reduction.
Figure 9. Deviation from the ideal and temperature of the Thermal Reserve Zone for both forms of operation.

Figure 10. Thermal Reserve Zone determined by the model.
Figure 11 shows the energy balance of the Elaboration Zone in relation to its thermal needs. The variables studied were point P and point V.

The process with use of PCI needs greater amount of heat from blow to form the reducer gas, since the amount of heat from blow is proportional to the burning efficiency of the PCI, therefore, $V_2$ is greater than $V_1$. This is confirmed by Table 1, since the operation with use of PCI has higher value for the Enriching of oxygen and blow Temperature and a lower Blow humidity.

An increase in the silicon content in pig iron is expected in the operation with use of PCI, since there was an increase in the quantity of heat supplied to the Elaboration Zone, but according to Table 1, there is a reduction in its content. This fact is due to the greater production speed in this period. The productivity is proportional to the charging speed and the greater this frequency, the lesser is the time for incorporation of silicon in the pig iron.

In relation to the thermal balance of both situations, we have that the higher quantity of heat supplied in situation 2 is used by the combustion process of the pulverized coal, $V_2$ is greater than $V_1$. In relation to the chemical balance, the higher quantity of reducer gas formed in the Elaboration Zone due to PCI injection reduces the quantity of direct reduction, since the CO that would be formed by direct reduction is supplied by PCI combustion.

The displacement of P in segment UV is not very pronounced, since there is great variation in the value of $\mu$, that is, there was no variation in the specific Fuel rate, Figure 12. Thus, substituting a coke fraction in the Fuel rate of the All coke process did not bring significant changes in the quantity of reducer gas.
Figure 12. Specific production of reducer gas for each period.

5 CONCLUSION

In stable operating conditions of Blast Furnace 1 of ArcelorMittal Tubarão, the Rist diagram showed in a simple way the variation of the main operating parameters when there was a change in the reduction process. And the variation of these parameters showed good agreement with the actual data from the reduction process.

The diagram made use of parameters that are of great importance to the reduction process, in addition to making correlations with the indices that evaluate operational quality of this process, like specific fuel rate, volume and blow temperature, and productivity.

The parameters used (direct reduction, furnace stack efficiency, temperature of the Thermal Reserve Zone and thermal balance of the Elaboration Zone) by the Rist diagram are effective to evaluate the Blast Furnace reduction process.

According to the Rist diagram, we have that the use of PCI brings benefits like: reduction in temperature of the Thermal Reserve Zone, of the percentage direct reduction, of the Coke rate and promotion of an increase in the furnace stack efficiency. Pulverized coal injection also did not alter the quantity of CO of the reduction process.

The temperature of the blast furnace’s Thermal Reserve Zone is related to the extent of the direct reduction reaction and with the coke rate. The greater the quantity of direct reduction, the higher the temperature of the Thermal Reserve Zone that is proportional to the coke rate. This brings consequences like reduction of the furnace stack efficiency and oxidation of the top gas.

The lower level of oxidation of the top gas is the result of increase in the proportion of direct reduction in response to increase in temperature of the Thermal Reserve Zone. Thus, the greater the proportion of direct reduction in relation to the indirect, the greater the regeneration of gases and the lower the oxidation of the top gas.
The measures to increase combustion efficiency of the PCI generate an increase in the quantity of heat in the Elaboration Zone. However, this increase is compensated by reduction in the quantity of direct reduction. Thus, the CO that would be generated by the direct reduction is formed by the PCI combustion.

The Rist diagram, despite its restrictions, can be used to evaluate the reduction process, as well as to understand and relate the operational parameters, such as: direct reduction, specific coke rate, furnace stack efficiency and study of the thermal profile of the blast furnace. In addition, it can be used through an interaction with an expert, for operation projection.

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