NATURAL GAS AND COAL INJECTION TECHNOLOGY FOR BRAZILIAN BLAST FURNACES[1]

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Resumo
Recently discovered major natural gas (NG) deposits in the Santos Basin, Southeaster of Brazil caused discussion about its possible use as a reducing agent in the blast furnace (BF) ironmaking to reduce coke consumption and to improve environmental protection. In this paper, firstly the fundamentals of natural gas combustion are briefly reviewed and then advantageous and constraints of its use in the BF are discussed. Theoretical analysis and industrial experience of NG use in the BF are presented. Its effect on flame temperature, gas volume, direct reduction rate, productivity and further BF operation parameters is discussed based on the calculations using a mathematical model. Furthermore simultaneous injection of NG and pulverised coal (PC) via the tuyere may provide a flexible and cost-effective technology for Brazilian BFs. Factors affecting the coke/(NG+PC) replacement ratio are discussed based on the modelling and industrial results. Tuyere apparatus designs for co-injection of NG and PC are discussed as well considering mixing and combustion of injectants in the tuyere and raceway. Effect of products of NG combustion on PC conversion behaviour in the tuyere and raceway has been examined using an STA (Simultaneous Thermal Analysis) experimental set. Finally injection of the reducing gas generated by steam conversion or by partial oxidation of NG with oxygen is discussed.
Key words: Natural gas; Blast furnace; Co-injection; Tuyere; Reducing gas.

TECNOLOGIA DE INJECÃO DE GÁS NATURAL E CARVÃO PARA ALTOS-FORNOS BRASILEIROS

Resumo
Recentes descobertas de grandes depósitos de gás natural (GN), na região sudeste do Brasil, na bacia de campos, causam discussão sobre a possibilidade de utilização deste combustível como agente redutor no alto-forno (AF) visando a redução do consumo de coque e melhorias na área ambiental. Neste trabalho, primeiramente serão revisados os fundamentos da combustão do gás natural assim como vantagens e limitações da sua utilização no AF. Serão discutidos aspectos referentes a teoria e uso industrial do GN no AF. Um modelo matemático será utilizado para discutir o efeito do GN na temperatura de chama, volume de gases, taxa de redução direta, produtividade e outros parâmetros de operação do AF. A injeção simultânea de GN e carvão pulverizado (CP) nas ventaneiras dos AF se coloca como uma tecnologia flexível e atraente para os AFs brasileiros. Fatores que afetam a taxa de substituição do coque/(GN+CP) serão abordados baseados em modelamento e resultados industriais. Aspectos relacionados ao designs das ventaneiras para co-injeção serão discutidos bem como a mistura e co-combustão destes combustíveis nas ventaneiras e zona de combustão. Efeito dos produtos da combustão do GN no comportamento de conversão do CP nas ventaneiras e zona de combustão será examinado através de ensaios em termobalança. Finalmente a injeção de gases redutores gerados pela conversão ou oxidação parcial do GN será discutida.
Palavras-chaves: Gás natural; Alto-forno; Co-injecão; Ventaneiras; Gás redutor.

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1 INTRODUCTION

Blast furnace (BF) which exists since 700 years still remains the cheapest source of iron; it demonstrates flexibility and adaptability to changing conditions again and again. To keep its competitiveness, main challenges for BF ironmaking are energy saving and reducing the CO_2 emissions. The last goal can be basically achieved by lower consumption of reducing agents, top gas recycling and replacement of carbon-based fossil reducing agents (coke, coal) with hydrogen-based or renewable energy sources (further options like plasma, electrical and nuclear energy are under development and are not mature yet).(1)

Brazil has significant advantages for cost-effective and environmentally friendly ironmaking with tremendous reserves of iron ore and eucalyptus plantations. About one third of total hot metal production (32.45 Mt in 2006) is made in small charcoal blast furnaces.(2) To reduce the coke rate in large blast furnaces, PC is injected (120 - 150 kg/tHM).(3) Newly-discovered in the Santos Basin, Southeaster of Brazil reserves of natural gas open new perspectives for its use in steel industry, in particularly, by injection into blast furnaces alone or with PC or probably with charcoal. Proven natural gas reserves in Brazil are 364.9 billion cubic meters and this number is increasing. Petrobras intends to increase the natural gas offer from 45 million m^3/day in 2008 to 73 million m^3/day in 2011 and to surpass 100 million m^3/day in 2015.(4)

Blast furnace technology with NG injection is widely used in Russia, Ukraine, USA and some other countries.(5) E.g. in Russia and Ukraine about 90% of blast furnaces use NG*;* injection rate is in the range 50 to 150 m^3/tHM. In Ukraine: mining and steel industry consumed in 2005 7.6 billion m^3 of natural gas including 40.1 % for hot metal production plus 2.88 % for sintering;(6) average NG injection rate was 98.3 m^3/Thm.(7) In North America, NG injection has increased substantially since 1990s and has become dominant for furnaces situated in the area where NG is readily available at a low price. In 2005 12 blast furnaces of total 36 inject NG and further 21 furnaces inject NG together with other reducing agents (coal, oil, tar).(8) The typical NG injection rates in North America are in the range 30 - 40 kg/tHM, the highest rate of 155 kg/tHM was achieved (coke rate was 310 kg/tHM).(8,9) In EU-15, NG is virtually not in use; only two blast furnaces at HKM in Germany operate with co-injection of NG (up to 50 kg/tHM) and oil. Some companies in South America like Cosipa in Brazil in 1994-98 and Siderar in Argentina have also injected NG into blast furnaces tuyeres.(10,11)

Co-injection technology, i.e. simultaneous injection of two or three injectants, for example NG and PC, may provide not only flexible BF operation depending on availability and cost of injected substances but also optimal operation parameters by change of the ratio of NG and PC. On the other hand, mutual effect of both components, e.g. influence of NG combustion products on PC conversion and their combined effect on the oxidising potential in the raceway have to be considered. In this context tuyere apparatus design plays an important role. NG can be injected into a blast furnace either as raw material or in form of hot reducing gas generated by the NG conversion outside the blast furnace.

Above mentioned options and problems are discussed here based on theoretical analysis, industrial experience, mathematical modelling and lab experiments.

2 FUNDAMENTALS AND EXPERIENCE OF NG USE IN BLAST FURNACE

Table 1 shows the chemical analysis and physical properties of Brazilian NG.[12]

Table 1 - Composition and characteristics of Brazilian natural gases

Field	Composition (% vol.)						Density (kg/m^3)	Calorific value (MJ/Nm^3)
	CH_4	C_2H_6	C_3H_8	C_4H_{10} and higher	CO_2	N_2		
Rio de Janeiro	89.44	6.7	2.26	0.46	0.34	0.8	0.623	40.22
Bahia	88.56	9.17	0.42	-	0.65	1.2	0.615	39.25
Alagoas	76.9	10.1	5.8	1.67	1.15	2.02	-	47.7
Rio Grande Do Norte	83.48	11	0.41	-	1.95	3.16	0.644	38.54
Espírito Santo	84.8	8.9	3.0	0.9	0.3	1.58	0.664	45.4
Ceará	76.05	8.0	7.0	4.3	1.08	1.53	-	52.4

The pyrolysis and combustion of hydrocarbons are referred to a series of chain reactions, a theory of which was developed by academician N. Semyonov.[13] The scheme of the thermal destruction of methane is presented in (14). Final reactions of carbon and methane conversion in the raceway can be written in the simplified form as follows (NG for the sake of simplicity is considered as methane) :[5]

$$C + 0.5O_2 + 0.5\beta \cdot N_2 \rightarrow CO + 0.5\beta \cdot N_2 + 116.83\ MJ \quad (1)$$
$$CH_4 + 0.5O_2 + 0.5\beta \cdot N_2 \rightarrow CO + 2H_2 + 0.5\beta \cdot N_2 + 35.6\ MJ \quad (2)$$

Comparison of coke or coal and NG combustion (reactions 1 and 2) shows that e.g. for atmospheric air ($\beta = N_2/O_2 = 3.76$) 1 atom of carbon in coke produces 2.88 moles of bosh gas but 1 atom of carbon in methane produces 4.88 moles of bosh gas, i.e. gas volume increases by 70%. Besides, the amount of heat released in the raceway by conversion of methane is 3 times less than that for coke combustion. Both mentioned factors (increase of bosh gas volume and decrease of heat release) as well as absence of sensible heat of NG (t_{NG} = 0 - 20°C) result in a drop of the flame temperature by 320 - 420°C for every 100 m^3 injected NG. The effect of methane on the composition of the bosh gas is shown in Table 2.

Table 2 - Effect of methane injection on the composition of the bosh gas, %.

CH_4, % vol. in blast	CO	N_2	H_2	$CO+H_2$
0	34.7	65.3	-	34.7
1	34.2	64.2	1.6	35.8
2	33.7	63.4	2.9	36.6
5	32.1	60.5	7.4	39.5
10	29.9	56.3	13.8	43.7

When injecting other auxiliary reducing agents, flame temperature is also reduced since the bosh gas volume rises more strongly than the quantity of heat generated by fuel gasification and carried by the hot blast. The difference in the decrease of the flame temperature for various reducing agents depends on the C/H ratio. Heat of decomposition increases with the drop of the C/H ratio. Therefore the less C/H ratio, the less heat is released by gasification. For anthracite C/H = 33-50, for oil 7.7-9.0 and for methane 3.[15] Therefore the decrease of the flame temperature for oil and PC is less than that for NG: 300-350°C per 100 kg/t HM heavy oil and 80-220°C per 100 kg/t HM PC (80-120°C for low-volatile and 150-220°C for high-volatile coal).[5]

The effect of auxiliary gaseous, liquid and solid reducing agents injected into the hearth through the tuyeres can be compensated by enriching blast with process oxygen, by increase in blast temperature or decrease in humidity of the blast.

When co-injecting NG and PC, the oxidizing potential of the raceway at constant oxygen concentration in blast depends on the rate of both components and can be maintained or changed controlling their ratio, according to the equation:

$$\Delta S_{PC} = - (m/n)\, \Delta S_{NG} , \qquad (3)$$

where: S_{NG} and S_{PC}: injecting rate of NG and PC respectively, m^3/kgHM (kg/kgHM)

m, n: theoretical oxygen consumption for combustion of 1 m^3 NG and 1 kg PC respectively, m^3.

The values m and n are calculated by the equations derived from /14/:

$$m = 2\,(CH_4) + 3.5(C_2H_6) + 5(C_3H_8) + ...$$

$$n = 1.8667\, C^w + 11.2\, H^w,$$

where: CH_4, C_2H_6, C_3H_8...: methane, ethane, propane and further components in NG, %/100; C^w, H^w : content of carbon and hydrogen in PC, %/100.

To keep oxidizing potential e.g. for the case of simultaneous PC and NG injection, a PC consumption change by 10 kg should be accompanied according to eq. (3) by change in NG consumption in the opposite direction by 7-8 m^3. Injection of HRG generated by NG conversion (section 5) virtually does not require additional oxidizing potential of the raceway.

At temperature above 1100°C methane decomposes completely (Table 3). To suppress soot generation in the tuyere cavity and in the furnace, a good mixing of NG with the blast as well as its oxidation by oxygen of blast before NG can be decomposed are required. Good mixing with blast and uniform NG distribution around the furnace periphery are also necessary to avoid the local over-cooling of the hearth. NG burning may also start in the tuyere cavity and the degree of extension of this process depends on the NG and blast mixing.

Table 3 - Composition of gas mixture generated by reaction $CH_4 \leftrightarrow C + 2H_2$, % (vol.).[16]

Temperature, °C	CH_4	H_2
300	96.90	3.10
400	86.16	13.84
500	62.53	37.47
600	31.68	68.32
700	11.07	88.93
800	4.41	95.59
1000	0.50	99.50
1100	0.20	99.80

To ensure suitable mixing, NG stream can be introduced in the tuyere or blowpipe cavity perpendicularly to the blast stream or at a certain angle to it, or in the opposite direction.

Automatic control systems of NG and blast distribution around the furnace circumference are needed to avoid flame temperature deviations in different raceways, soot generation, and worsening of drainage of liquid products that might be caused by uneven NG distribution among the tuyeres.

3 CALCULATIONS OF THE EFFECT OF NG ON BLAST FURNACE OPERATION PARAMETERS

3.1 Mathematical Model

The calculations represented in the following were carried out using a mathematical model of the TU Donetsk, Ukraine. This is a global balance model developed on the base of a method of Prof. A.N. Ramm(15) and described in (17).

The key calculations determine:

- amount of oxygen released during reduction
- injected substance characteristics: total quantity of C, H, O and N; the enthalpy: heat released by burning in the raceway etc.
- volume of bosh gas
- direct reduction rate
- coke and total reducing agent consumption and blast volume
- flux consumption and slag volume
- top gas parameter (volume, temperature, composition, calorific value)
- heat balance (heat generated and absorbed)
- flame temperature
- change in the productivity and intensity of the coke combustion.

3.2 Calculation Conditions

Calculations have been carried out for the operation conditions of a modern blast furnace with PCI and oxygen enrichment of the blast; the hot blast temperature is 1180°C, the produced pig iron contains 0.37% Si and 0.029% S.

The burden consists of 58.2% sinter, 27.6% pellets and 11.9% lump ore. In Tables 4 and 5 the burden, coke, PC and NG parameters are given.

Table 4 - Burden consumption and composition.

Burden	Sinter	Pellets	Lump ore	Limestone
Consumption, kg/tHM	923	437	189	38
Composition, %				
Fe	58.09	65.70	65.50	-
FeO	4.90	1.50	2.00	-
CaO	9.67	0.92	0.90	53.5
SiO_2	5.09	4.70	8.20	1.8
MgO	1.54	0.26	0.90	0.7
Al_2O_3	1.05	0.30	0.50	0.3
MnO	0.22	0.07	0.10	0.05

Table 5 - Composition of NG (vol. %), coke and PC (wt %).

Components	NG	Components	Coke	PC
CH_4	92.0	Ash	8.8	7.5
C_2H_6	4.0	C	87.8	79.95
C_3H_8	1.0	H	0.2	4.2
C_4H_{10}	0	N	1.3	1.6
CO_2	1.0	O	-	4.5
N_2	2.0	S	1.09	0.94

3.3 Calculation Results

The main results of the calculation are presented in Table 6.

Table 6 - Blast furnace operating results.

	Base	1	2	3	4
Blast:					
temperature, °C	1180	1180	1180	1180	1180
volume, m^3/tHM	1000.8	605.6	915.2	834.1	820.2
moisture, g/m^3	12.6	12.6	12.6	12.6	12.6
oxygen, %	24.06	46.41	28.00	31.33	32.00
NG, m^3/tHM	0	150	84	100	130
PC, kg/tHM	160	0	0	0	0
Direct reduction rate, %	52.9	34.6	47.0	43.6	38.0
CO utilisation rate, %	43.7	40.9	41.7	41.8	42.1
H_2 utilisation rate, %	46.1	43.1	44.0	44.1	44.4
Slag volume, kg/tHM	276.9	261.8	268.1	266.3	261.7
Coke rate, kg/tHM	344.7	392.7	435.2	422.8	392.3
Top gas:					
volume, m^3/tHM	1546.9	1232.7	1494.6	1421.8	1419.8
CO_2, %	20.83	24.20	20.41	21.38	20.98
CO, %	26.14	34.22	27.89	29.12	28.23
H_2, %	3.51	14.68	7.20	8.76	11.03
N_2, %	49.51	26.90	44.50	40.74	39.76
calorific value, kJ/m^3	3685.1	5911.9	4303.4	4628.3	4761.3
External heat loss, MJ/tHM	407.8	338.9	387.9	373.9	361.9
Flame temperature, °C	2150	2150	2150	2150	2010
Productivity, %	100.0	128.0	108.9	113.6	114.1

The injection of NG affects flame temperature and gas volume stronger than PC. Therefore to keep flame temperature on the initial level when replacing PC with NG, the amount of injected NG has to be adjusted. Another measure to increase NG consumption is enriching blast with process oxygen. This technology ensures increase of productivity by 9% when replacing about 50% of PC with NG (case 2) and by 28% when completely replacing PC with NG (case 1). The blast and the top gas volumes are lowered; the rate of hydrogen in top gas is increased. The amount of CO_2 in top gas is decreased by 7.4%, 5.3%, 5.3% and 7.6% for cases 1, 2, 3 and 4 respectively in comparison with the basis case. The calorific value of the top gas is raised by 17-60% for cases represented in Table 6. The direct reduction rate decreases by 5.9-18.3% for calculated variants in comparison with the basis case. The coke/NG replacement ratio is about 0.8 kg/m^3.

4 CO-INJECTION OF NG AND PC

Comparative analysis of NG and PC injection into blast furnace which apart from price and availability considerations affects the efficiency of blast furnace operation is shown in Table 7.

Table 7 - Qualitative comparative analysis of NG and PC use in a blast furnace.

Reducing agent	NG	PC
Capital costs	low	High
Preparation work	less	More
Transport	simple	Complex
Effect on flame temperature, bosh gas, direct reduction rate	bigger	smaller
Conversion in raceway	fast	Slow
Partial conversion outside the raceway	probable	hardly possible
Effect on hot metal and slag	no	sulphur, ash
CO_2 emission	low	High

Simultaneous injection of both auxiliary reducing agents can increase benefits compared to their single use. Results of numerical simulations have shown that the furnace productivity can be increased by 30% when co-injecting of PC and NG; at the same time the coke rate and carbon emission as CO and CO_2 are reduced. [18]

A technology for co-injection of NG and PC is successfully used for many years at steel company "Donetskstal" in Ukraine.[14] Generalised for 5 years in the late 1990s operation parameters of BF2 with useful volume 1033 m^3 were following: oxygen content in blast 24.0 - 25.5 %, total consumption of injectants 150 - 180 kg/tHM including NG 80 - 90 m^3/tHM, PC 70 - 100 kg/tHM; coke/coal replacement ratio about 0.9 kg/kg, coke/NG replacement ratio about 0.8 kg/ m^3. The compensation rate of PC by NG, considering the effect of auxiliary reducing agents on the oxidising potential of the hearth as well as on the thermal state and fluid mechanics was in the range 0.2-0.5 m^3/kg depending on composition of the injectants and blast furnace operation parameters. Due to rising prices for natural gas blast furnaces at "Donetskstal" operate last years with increasing rate of PC (Table 8 shows operation results of BF2 for two periods of 2004 and 2005)[19] or only with PC injection.

Table 8 - Operation parameters of BF2 "Donetskstal".

Parameters	Period 1	Period 2
Blast temperature, °C	1094	1085
Oxygen in blast, %	25.71	25.64
PC, kg/tHM	131	138
NG, m^3/tHM	69	65
Coke rate, kg/tHM	392	381
[Si], %	0.77	0.78
[S], %	0.032	0.031
CO utilisation rate, %	45.2	45.4
Direct reduction rate, %	31.6	33.9
Flame temperature, °C	2084	2085

Tuyere apparatus design for co-injection of NG and PC should provide a complete mixture of injectants with oxidizing agent, optimal kinetic energy of streams as well as reliable and simple service. Most of these tuyere constructions use a dual lance system. A tuyere apparatus design with special lances for co-injection of NG and PC introduced into the tuyere body is published in (5,14).

5 GENERATION OF HOT REDUCING GASES (HRG) BY NG CONVERSION AND ITS INJECTION INTO BLAST FURNACE

Reducing gas can be generated from NG by its conversion e.g. with steam, carbon dioxide, steam or oxygen:

$$CH_4+H_2O = CO+3H_2 - 9211\ kJ/m^3\ CH_4$$
$$CH_4+CO_2 = 2CO+2H_2 - 11053\ kJ/m^3\ CH_4$$
$$CH_4+0{,}5O_2 = CO+2H_2 + 1591\ kJ/m^3\ CH_4$$

Above reactions correspond to an ideal process. Under real conditions along with main products CO and H_2 also CO_2, H_2O, CH_4, N_2 and soot are present. Catalysts like compounds of Ni are used to reduce the amount of oxidising components. E.g. HRG generated by NG conversion with carbon dioxide and injected into blast furnace of steelworks Novotulskij, Russia had following parameters: t=1230-1270°C; 23,3-29,8% H_2, 36,4-37,8% CO, up to 1,7% CH_4, 0,9-1,7% CO_2, 3,0-6,6% H_2O, 31,6-36,8% N_2. Results of this and further industrial trials with injection of HRG generated by NG conversion with steam and steam-CO_2 in Ukraine and Russia were reported in (20,21).

Prof. A.N.Ramm calculated blast furnace operating parameters when injecting raw NG and HRG generated from NG by catalytic steam conversion into the hearth /15/. From 1 m^3 NG was generated 3.95 m^3 HRG. The calculations were performed for HRG injection rate up to 900 m^3/tHM. HRG characteristics were as follows: t=1200°C, $\Delta H=c_p t=1788$ kJ/ m^3. The calculation results show that flame temperature decreases by 80°C and 410°C per 100 m^3 HRG and raw NG respectively at 21% O_2 in blast and by 125°C and 450°C per 100 m^3 HRG and raw NG respectively at 40% O_2 in blast.

Recently injection of hot gas into the BF shaft was executed in operational trials at BF1 at ArcelorMittal Eisenhüttenstadt.[22] The reducing gas was generated by partial oxidation of natural gas with oxygen within so called hot gas generators (Figure 1); it is designed for a maximum production of reducing gas of about 2,400 m³/h. The generators were installed at the BF shaft (their number corresponds to the number of shaft tuyeres). Every generator is supplied with gas and oxygen independently. Operational results and obtained experience are discussed in (22).

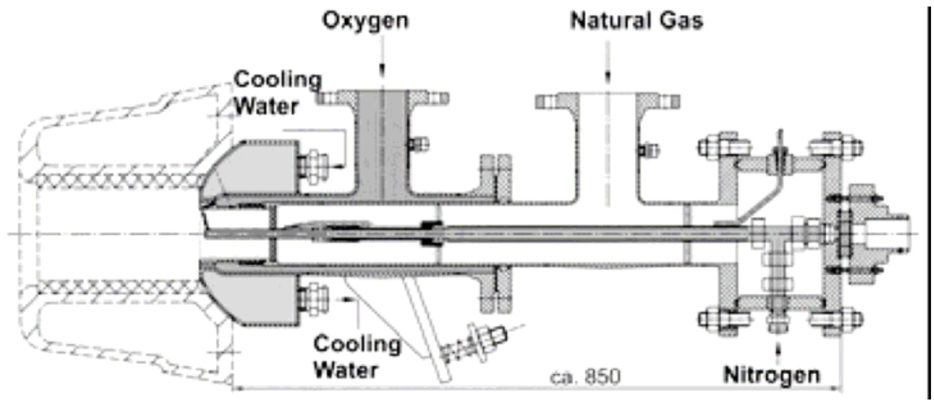

Figure 1: Schematic diagram of the hot gas generator. [22]

6 EFFECT OF NG COMBUSTION PRODUCTS ON PC GASIFICATION

NG ignites and burns quicker than coal and therefore products of its combustion (CO_2 and H_2O that are formed before Boudouard and water gas shift reactions start) may affect coal conversion when both reducing agents are simultaneously injected. In this study the effect of CO_2 on PC reaction rate with air was examined.

The experiments were conducted using the Simultaneous Thermal Analysis equipment (Shimadzu TGA 50-H)[1]. Coal samples were heated up in air and in air+8% CO_2 atmosphere with constant heating rate of 20°C/min up to 1100°C. Results show that the difference in reaction rate in two examined atmospheres makes up 3.4% for (Figure 2).

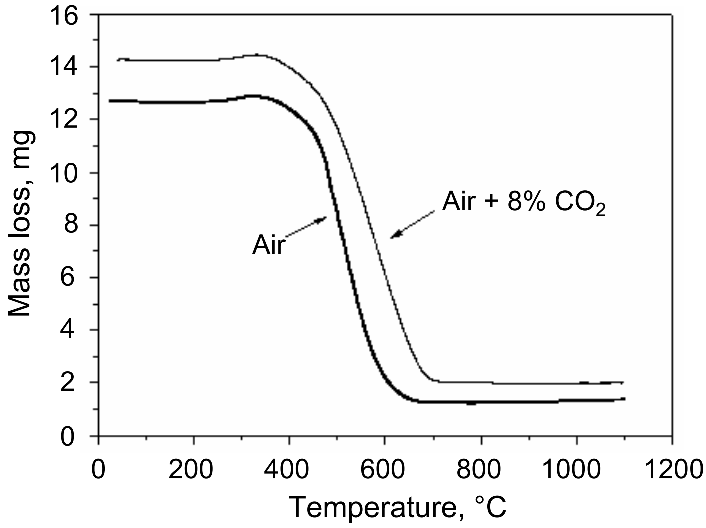

Figure 2: Effect of carbon dioxide on PC reaction rate

7 CONCLUSIONS

- Use of NG in Brazilian BFs can reduce coke consumption and improve environmental protection.
- A good mixing of NG with the blast and its uniform distribution around the furnace periphery are necessary to suppress soot generation and to avoid the local over-cooling of the hearth.
- NG affects BF parameters such as flame temperature, bosh gas and direct reduction rate stronger than coal, charcoal or oil. Calculation results using a mathematical model showed that replacing 160 kg/tHM PC with 84 and 150 m^3/tHM NG decreases the amount of CO_2 in top gas by 5.3% and 7.4% and can increase productivity by 9% and 28% respectively. The coke/NG replacement ratio is about 0.8 kg/m^3
- Simultaneous injection of NG and PC or charcoal can increase benefits compared to their single use. To keep oxidizing potential of the raceway when injecting PC and NG simultaneously, a PC consumption change by 10 kg should be accompanied by change in NG consumption in the opposite direction by 7-8 m^3. Injection of hot reducing gases (HRG) generated by NG conversion virtually does not require additional oxidizing potential.
- Injection of HRG may ensure considerably higher injection rate compared to raw NG and provide higher productivity; "oxygen BF" can be realised as well.
- Presence of CO_2 inhibits the PC reaction rate slightly.

[1] Tests were performed in Centro Nacional de Investigaciones Metalúrgicas (CENIM), Madrid

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