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Combustibility behavior of PCI coals, green petroleum coke and charcoal fines used as fuel for injection into blast furnace tuyeres

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Abstract

Pulverized coal injection (PCI) into blast furnace tuyeres is widely used by integrated steel mills worldwide to reduce the consumption of coke and costs. High injection rates are desirable. The big challenge is to achieve them with cheaper and lower quality raw materials, without losing the quality of the hot metal and the productivity of the blast furnace. An increase in the injection rate leads to improved fuel selection. Thus, it is important to have knowledge of the injected fuel quality variables that affect the burning efficiency and the replacement rate of the coke into the furnace, as well as the quality of the hot metal and the stability of the furnace. In this context, the present study is based on the chemical characterization and combustibility behavior of four fuels: Australian coal (CMA), North-American coal (CMN), charcoal fines (MCV) and green petroleum coke (CVP) and fuel blends. Results of chemical analysis show that the CMA, CMN, MVC and CVP are within the ranges of acceptable values in the PCI process for the ash and sulfur contents. The order of combustibility by thermogravimetric analysis was MCV>CMN>CMA>CVP. However, the combustion rate obtained by the simulator test, performed under extreme conditions of short residence time and high temperature, presented a different order of combustion rate of MCV>CVP>CMA>CMN, which may be related to the mineral on char. The blend that presented the best burning efficiency was obtained for the composition containing 20%MCV+80%CVP, followed by blends containing 80 and 90% of CMA, respectively.

Keywords: blast furnace, PCI, fuel, combustibility behavior.

1. Introduction

Pulverized coal injection (PCI) is an effective technology for reducing coke consumption in the blast furnace (BF) (Bennett and Fukushima, 2003; Lion *et al*, 2017; Zou *et al*, 2017), leading to a reduction in BF operational costs, which is explained by the fact that the price of coal is slightly lower than that of coke. Thus, it is increasingly desired to operate the furnace with higher injection rates, resulting, also, in an increased demand for coal.

The development of PCI technology for the blast furnace tends to address energy and environmental issues. PCI stimulates innovations to BFs, through the use of recycled materials, such as waste plastics (Carpenter, 2010), recycled ores and biomass (Assis *et al*, 2014), and others material, such as tires (Assis and Assis, 2009) and petroleum coke (Silva *et al*, 2010), usually by injecting mixtures with coal. On the other hand, as injection rates increase, more complex characteristics, such as combustibility, char reactivity, furnace permeability and flow properties, influence the selection of the fuels (Carpenter, 2010) are involved.

With the fuel particles being injected directly into the blast furnace tuyeres, because of the high heating rate associated with short residence time and a non-efficient dispersion of the particles, the generation of unburned char turns out to be inevitable for the operation at high injection rates (Du *et al*, 2010). At low injection levels, most of the unburnt char is consumed, but the efficient consumption of unburnt char for high PCI rates have worse performance (Gupta *et al*, 2006).

Incomplete conversion of injected carbonaceous materials can cause reduction in the gas permeability, dirtying of the dead man (Babich *et al*, 2010), if the excess of char generated exceeds its consumption, influencing the distribution of gas flow through the lower section of the furnace (Bennett, 2000), and finally decreasing the furnace productivity and increasing the coke rate (Babich *et al*, 2010). Thus, especially when applying high injection rates, it is necessary to evaluate the combustion efficiency of the injected pulverized fuels in order to have a stable operation of the BF.

Many authors have studied the combustion behavior of pulverized fuels injected into the furnace through the tuyeres (Du *et al*, 2010; Nozawa *et al*, 2011; Zou *et al*, 2017) in order to increase the performance and enhance the productivity of the blast furnace. Replacing coke by using alternatives fuels has additional benefits, such as, extended coke oven life, flexibility in the BF operation, and improved consistency in the quality of the hot metal (Carpenter, 2006). An alternative to the use of coal is the charcoal fines, generated due to its breakage during production, transportation and handling. The fines have similar advantages of charcoal, with the benefit of having a market price even lower than the charcoal. All the carbon matter in charcoal is generated from the CO_2 present in the atmosphere and only part of it is released again as CO_2 in the BF, resulting in a favorable CO_2 balance when using charcoal or its fines. According to a study, considering charcoal as a CO_2- neutral component, CO_2 input decreases up to 45% when replaced by 100% coal (Babich *et al*, 2010). Besides the environmental advantages, it is also possible to consider among the advantages of the use of charcoal for injection, the low sulfur and phosphorus content, high combustibility and relatively low ash content.

Another possibility is green petroleum coke or pit coke, a byproduct of petroleum refining. Its high carbon content and calorific power, low ash and sulfur content, relative low cost and abundance have made it an attractive product for industrial use, being suitable for consumption in blast furnace replacing metallurgical coke for its use in blends for PCI purpose (Silva, 2010). Furthermore, the effect of coal injection is well investigated and few studies reporting the effect of green petroleum coke injection into blast furnace tuyeres have been shown in literature until now.

This article presents experimental results to evaluate the combustibility of solid fuels and their possible effect of PCI in the blast furnace. Therefore, this study aims to investigate coal, charcoal and green petroleum coke combustibility and to identify the characteristics that justify such behavior.

2. Materials e methods

Four fuels were studied herein, which included two PCI coals, named CMA (Australian) and CMN (North-American); green petroleum coke, CVP (Brazilian); and charcoal fines, MCV (Brazilian). Fuel sampling and preparation were done according to ASTM D2234 and ASTM D 2013-03, respectively. Samples were ground and sieved to 90µm and then, some blends were prepared for the injection simulator test to determine the combustion rate. Proximate analysis was performed by ASTM D7582-15 in order to determine volatile materials, ash and fixed carbon content. The ultimate analysis was done by using the Elemental Analyzer, Vario EL III to determine carbon (C), hydrogen (H), nitrogen (N), and sulfur (S) contents. The oxygen (O) content was calculated by the difference.

X-ray fluorescence (FRX) analysis was performed according to ASTM D4326. It was possible to determine the contents of the major elements present in the fuel ashes, such as silicon (Si), aluminum (Al), iron (Fe), Calcium (Ca), Magnesium (Mg), Sodium (Na), Potassium (K), Titanium (Ti), and Phosphorus (P), all expressed as oxides, SiO_2, Al_2O_3, Fe_2O_3, CaO, MgO, Na_2O, K_2O, TiO_2 and P_2O_5.

In order to investigate the combustion behavior of selected fuels for PCI application, a relatively rapid and effective technique was used (Ronstam-Abadi *et al*, 1990). The combustibility performance of the pulverized fuels was estimated using the Thermogravimetric Analysis (TGA), which evaluates the loss and/or mass gain of the sample as a function of temperature in an environment with controlled temperature and atmosphere. Smith *et al* (1981) suggest to use of the first thermogravimetric derivative of the weight loss curve (DTG) to follow the combustion profiles of the fuels. The heat releasing state was determined by the difference in the thermal analyses (DTA). Therefore, combustibility tests by TGA/DTG/DTA test were carried out on a thermogravimetric analyzer Du Pont 2960 SDT V3. The temperature was raised from room temperature to 1000° C, and under synthetic air and flow rate of 50ml/min and heating rate of 25 °C/min (Osório, 2008). Combustibility of coals can be determined by the maximum reactivity point, the peak temperature (Tp), when maximum reaction rate can be observed, and as a better combustibility performance is observed for lower Tp (Smith *et al*, 1981; Cumming and Mc Laughlin, 1982; Babieri, 2016).

The main combustibility test was carried out using a laboratory injection simulator equipment with a high thermal gradient. The equipment seeks to reproduce the tuyeres and raceway environments. Therefore, there is no need for running experiments on industrial BF, which may be more expensive and have more risks.

The sections of injection simulator, used to determine combustion rate, can be observed in Figure 1. This simulator was developed in the Laboratory of steel industry studies of the School of Mines, Federal University of Ouro Preto (UFOP) to be able to simulate the combustion of pulverized materials injected into the raceway in terms of high heating rate and short residence time of the particles in the raceway (ASSIS *et al*, 2009). For a better understanding, the equipment was divided into the following sections: section 1: injection lance; section 2: preheating region; section 3: point of injection in the low pressure zone; section 4: tuyeres and section 5: raceway.

The equipment has basically two zones: A high pressure zone, where the pulverized fuel is injected, which contains a pressure regulator P1, an electromagnetic valve V1 and a cooler R1. The low pressure region refers to the rest of the pipes, which consists of the preheating zone and the raceway.

The preheating furnace (FP) heats the inlet gas to about 1000 °C in order to simulate the blast furnace blowing temperature. The combustion furnace (F1) simulates the firing characteristics of the blast furnace combustion zone and operates up to 1500 °C. The injection lance is simulated by a connected pipe, with a 30 ° inclination, between the preheating furnace and the combustion furnace. The injection lance consists of the following components: a pressure regulator P1, a point of injection or inlet (S); electromagnetic valve V1 and cooler R1. At the exit of the furnace F1, there is a flange with filter collector (F) that allows the retention of the oxidized particles and ashes. An electromagnetic valve (V2), which generates, together with the electromagnetic valve (V1), a pressure wave to entrain the sample, and finally the (R3) cooler protects the filter.

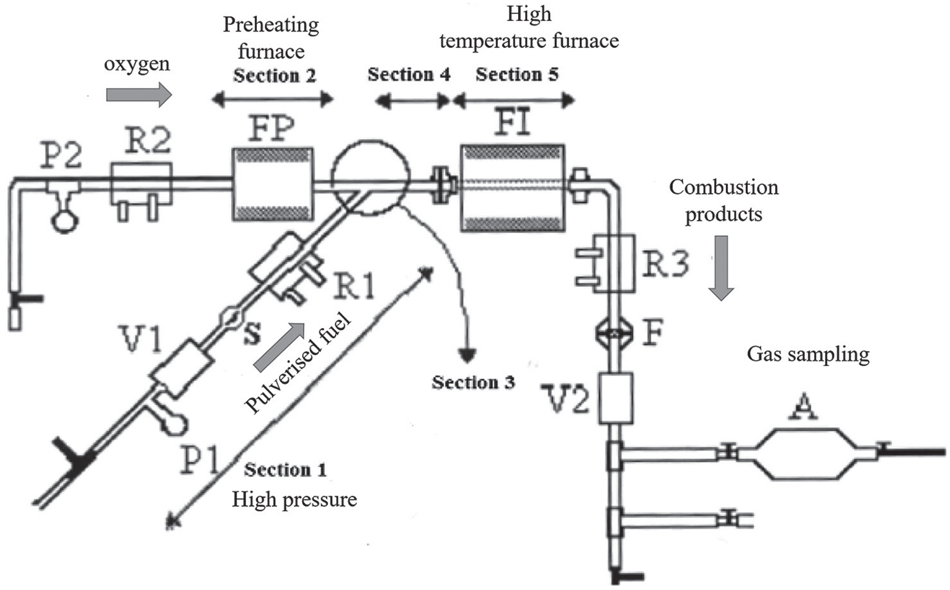

Figure 1
Schematic of the laboratory injection simulator equipment and its sections.
Fonte: Adapted Assis, 2004

The simulation tests were performed for the single fuels and their blends. In order to calculate the combustion rate, the gases generated in the combustion were collected and then analyzed in an Orsat apparatus. The following Equation 1 (Assis, 2014) was used to estimate the combustion rate:

$$CR = \frac{k * (\%CO + CO_2) * n}{(m_a * \%C_f / 1200000) - (\%CH_4 * n_g / 100)} \quad \text{Equation 1}$$

Where CR is the fuel combustion rate (%), K is a constant; %CO, carbon monoxide; $\%CO_2$, carbon dioxide; and $\%CH_4$, methane are the percentages in volume of the gases produced during combustion of fuel; n_g is the number of moles of gas, $\%C_f$ is the fixed carbon; and m_a is the sample mass injected in mg.

3. Results and discussions

3.1 Chemical analysis of fuels and their ashes

Some quality parameters are considered important for the selection of fuel injected into the BF. Results from proximate and ultimate analyses of the single fuels and their ash composition are presented in Table 1. All fuels are applicable for injection into the BF. CMA, CMN and CVP showed low volatile matter content and MCV high volatile content. Studies (Assis *et al*, 2004; Barbieri *et al*, 2016) show that the increase in volatility produces a higher combustion efficiency. The combustion of pulverized fuel follows the steps of preheating, volatile matter combustion, gaseous combustion and char oxidation/burning (Liao *et al*, 2017), so at high temperature, the volatile matter gasifies quickly, resulting in faster combustion (Assis *et al*, 2014). However, the high volatile coals have the lowest coke replacement ratio (Bennet, 2000) showing the dependence on the carbon content that is directly related to the calorific value and coal rank. Furthermore, ultimate analysis shows that CVP was the fuel that presented the highest carbon content followed by CMA, CMN and MCV.

Considering the injection of pulverized fuels, the material can become a relevant source of ash and variations in its quality can affect the stability of the furnace. Therefore, it can be observed that CMA and MCV have similar amounts for ash content and both are close to the limit normally accepted (10%) for BF injection (Rizzo, 2009). The chemical analysis of the ashes by FRX showed different results for the composition of the four fuels. CMA and CMN have the highest content of Al_2O_3 and SiO_2 indicating the presence of minerals rich in these elements. Charcoal, slightly over the limit, originally contains a relatively small amount of ash, but contamination suffered during its process can lead to great variability. CVP has the lowest ash content, being ideal for injection, considering this parameter.

Sulfur is undesirable for the production of the steel, leading to hot brittleness and raising the cost of production due to additional operations for desulfurization (Assis, 2009). Hence, the fuels are below the maximum limit used for sulfur content 0.8 % (Carpenter, 2006), with the exception of CMN, which is slightly over this limit. Phosphorus content is also another element that has been limited because it is capable of generating a negative effect on various forms of embrittlement, which can reduce the toughness and ductility. Low phosphorus content is desired. The phosphorus content was determined as P2O5, in the fuels ranging from 0.001 to 0.16%. Low alkalis content (K_2O and Na_2O) are desirable, since these are harmful to the blast furnace, as among its effects are increases the risk of scaffold formation and coke consumption, and the attack the refractory (Olofsson, 2017). As the presence of Na_2O was not observed, the alkalis content was considered as the K_2O content only, ranging from 1.21 to 1.73% in the samples.

Analysis	Component (wt %)	Sample			
		CMA	CMN	CVP	MCV
Proximate analysis	Ash	9.10	5.08	0.47	10.62
	Volatile Matter	18.03	19.92	14.90	34.59
	Fixed Carbon	64.87	67.00	79.44	48.80
Ultimate analysis	Carbon	79.22	75.62	89.51	67.26
	Hydrogen	3.53	3.57	3.63	2.44
	Nitrogen	1.66	0.86	2.28	0.37
	Sulfur	0.43	0.82	0.61	0.11
	Oxygen	3.56	1.60	1.89	10.90
Ash composition	Al_2O_3	32.05	25.73	9.30	22.54
	CaO	5.88	11.24	10.92	10.17
	Fe_2O_3	9.95	14.06	18.12	9.17
	K_2O	1.21	1.56	0.00	1.73
	MgO	1.64	3.48	1.22	2.23
	MnO	0.12	0.14	0.73	0.49
	P_2O_5	1.74	0.1	0.28	0.73
	SiO_2	42.20	40.06	28.39	43.23
	TiO_2	1.66	1.28	0.51	1.62
	ZnO	0.01	0.05	2.26	0.01

Table 1
Experimental results of proximate analysis and ultimate analysis and ash composition of PCI coals, charcoal and green petroleum coke (Dry basis).

3.2 Combustibility tests

The results of TGA/DTG/DTA analysis are presented in Figure 2 a-d, The TG/DTG/DTA curves were determined for the single fuel samples versus temperature. According to Wang et al 2008, it is possible to identify four combustion stages through the analysis of the TGA/TDG curves: The dewatering period, volatilization and burning, char burning, and burnout. However, as the 4 samples were analyzed on a dry basis, only the last 3 stages can be observed for coal and petroleum coke samples: A, volatilization and burning; B, char burning and C, burnout. MCV presented a single peak referring to volatilization and char burning in one single stage, having in total of 2 stages: A, volatilization and char burning and B, burnout.

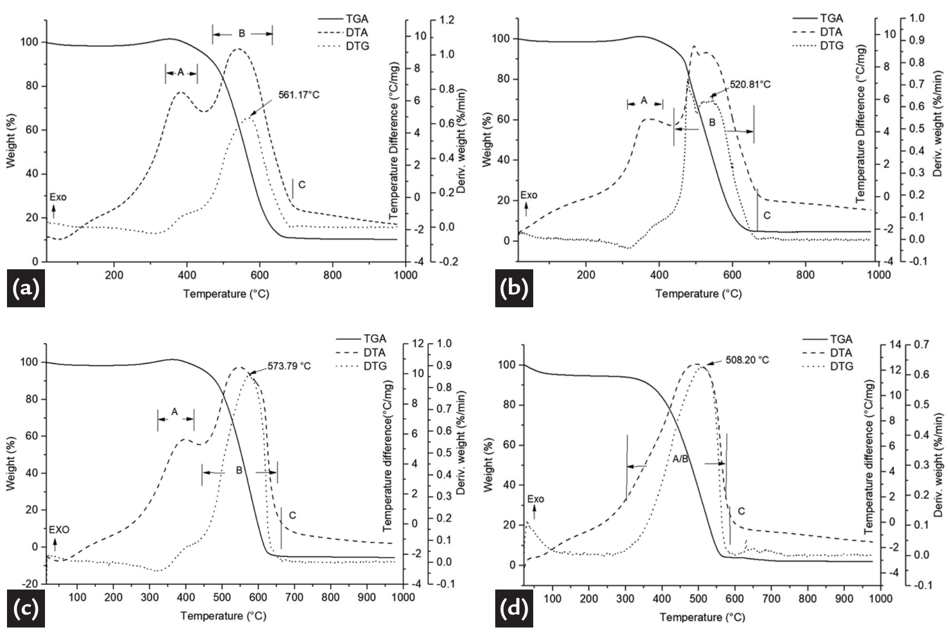

Figure 2
TGA/DGT/DTA curves showing the combustion profiles of single fuels: a) CMA, b) CMN, c) CVP and d) MCV.

As can be seen in Figures 2 a, b and c, in the temperature range 270-310°C, coals and petroleum coke presented a negative deflection in the burning rate curve. According to Cumming and McLaughlin, 1982, this phenomenon is due to the weight gain caused by the solid-state oxidation of the organic matter. It can also be described as an oxygen chemisorption in the carbon structure (Barbieri *et al*, 2016), and with the increase in temperature, the burning of volatile matter leads to weight loss. However, the same gain in weight cannot be observed for charcoal which may be due to the higher oxygen content in charcoal. Through the DTA curve, it is possible to observe for CMA, CMN and CVP, two thermopositive peaks related to volatile and char burning, respectively. According to Wang *et al*, 2008, for volatile matter burning, the thermopositive peak is short and lower, thus a lesser amount of heat is released. While for char burning, the thermopositive peak is higher, resulting in a large amount of heat.

Figure 3 presents the Tps for the 4 single fuels. MCV has the lowest Tp (508.20 °C) compared to the other samples, and is the sample that shows the best combustibility performance among the fuels, in agreement with literature (Barbieri *et al*, 2016), followed by CMN (520.81°C), CMA (561.17 °C) and CVP (573.79°C) which shows inferior combustibility performance, also in agreement with literature, considering TGA/DTG analysis (Osório *et al*, 2008). Additionally, according to studies carried out by Rostam-Abadi *et al*, 1990, fuels with higher burnout temperatures showed lower combustion efficiencies, in agreement with the results obtained.

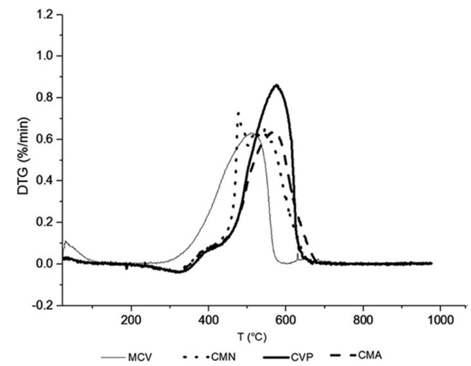

Figure 3
DTG curves comparing combustion profiles of single fuels.

The understanding of the combustibility behavior by calculating the combustion rate under the extreme conditions of short residence time is very important for selecting a fuel or producing blends, especially when considering higher injection rates. The test performed in the simulator equipment is capable of generating results comparable to the results obtained on an industrial scale. Table 2 shows the combustion rates as well as the values of Tp for purposes of comparison of the two techniques of analysis.

Table 2
Peak temperatures by TGA/DTG analysis and combustion rate for single fuels.

Sample	Tp (°C)	Combustion rate (%)
CMA	561,17	79,55
CMN	520,81	77,05
CVP	573,79	85,05
MCV	508,20	90,65

The combustion rate obtained by the simulator test has an order of MCV> CVP> CMA> CMN. The results obtained for charcoal fines and for coals are in agreement with values found by Assis *et al*, 2014. However, the results obtained for CVP and CMN are in disagreement with the results obtained by TGA/DTG analysis.

The second highest combustibility rate was determined for the green petroleum coke, which showed the lowest volatile matter content among the samples. Although this behavior was not expected (Silva *et al*, 2010), when compared to CMA and CMN coals, under the extreme conditions of the simulation test, CVP has a very low ash content and high fixed carbon. It is believed that these characteristics have improved combustibility under extreme conditions. According to Gupta *et al*, 2006, on some occasions, low-volatile fuels have a better combustion performance that could also be explained by the impact of mineral association with carbon and others minerals on char reactivity and hence on the combustion behavior of the fuel. Furthermore, the authors concluded that minerals rich in Si and Al can contribute to slow down the combustion kinetics of coals. On the other hand, minerals rich in Fe, Ca, Mg, Na and K, are capable of improving the combustion reactivity.

Figure 4 a-b show how the combustion rate is affected by the injection rate of the single fuels and mixtures.

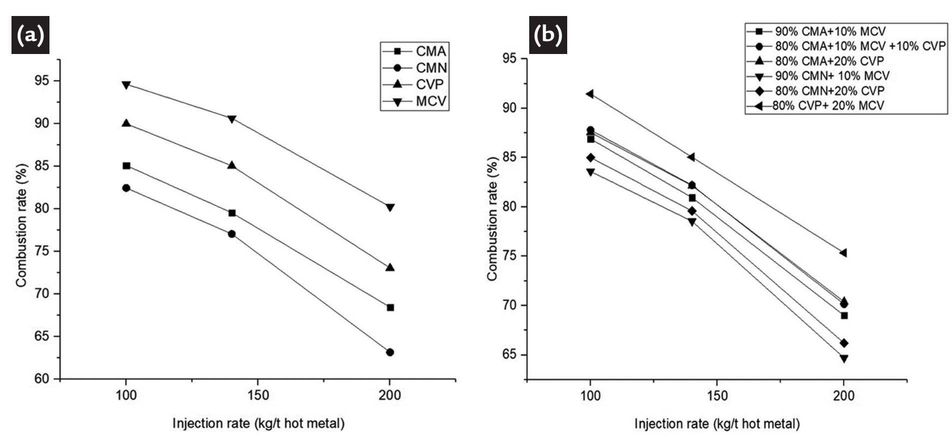

Figure 4
Injection rate versus combustion rate by injecting: a) single fuels and b) mixtures

The fuels are combusted and gasified immediately at high temperature and short residence time, but its combustion rate decrease with the increase of injection rate due to insufficient stoichiometric conditions, which is related to the decrease of oxygen and carbon ratio (O_2/C) (Assis, 2014).

The blends with better combustibility performance were found for the mixtures composed of CVP and MCV followed by mixtures containing the CMA coal. Through the combustion rate results, it is observed that the addition of charcoal fines (high volatile) in the coal mixture contributed to a better combustibility. The volatiles released from charcoal fines would enhance the surrounding coal (Barbieri *et al*, 2016) and petroleum coke devolatilization. The combustion behavior of fuels also depends on the fuel ratio and the nature of the coal (Du *et al*, 2010).

4. Conclusions

Considering the parameters that influence the consumption of fuels in the blast furnace, the chemical characterization and combustibility tests were carried out in order to elucidate the behavior of the Australian and North American coals, charcoal fines and green petroleum coke. As conclusions of the present study, we can cite that:

• MCV (MV: 34.59%) high volatile content, a desirable feature when considering a better combustibility performance in the raceway. CMA (MV: 18.03%), CMN (MV: 19.62%), and CVP (MV: 14.09%) presented low volatile content. In relation to the ashes, the CVP (Cz: 0.47%) had a lower ash content, which was already expected.

• The chemical analysis of the ashes by FRX showed different results for the composition of the three fuels, so that the CMA and CMN have the highest content of Al2O3 and SiO2 indicating the potential effect of these elements on slowing the combustion kinetics of coals.

• The results of the combustibility performance obtained by TGA/DTG analysis were distinct from those determined by test in the simulator. Through the combustion profile obtained by TGA/DTG, using Tp, it is noted that the combustion order was MCV> CMN> CMA> CVP, with CVP presenting the lowest level of burning, which was already expected, as it is the fuel with lower content of volatile materials. On the other hand, the combustibility performance obtained by the simulator test resulted in the following order of combustion rate (%): MCV> CVP> CMA> CMN, suggesting the possible impact of minerals on char on fuel burnout under extreme conditions.

• The blend that presented the best burning efficiency was obtained for the composition containing 20% MCV + 80% CVP, followed by mixtures containing 80 and 90% of CMA, respectively, which would be considered attractive for injection in the blast furnace.

Acknowledgments

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