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ASSESSMENT OF THE FISH SCALES AND COAL BLENDS COMBUSTION IN A DROP TUBE FURNACE (DTF)

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Abstract. *One of the main technologies for converting biomasses into thermal energy is the co-firing of these materials with coal. This study proposed the reuse of fish scales as biofuel in co-firing processes with coal in a Drop Tube Furnace (DTF). The samples were prepared with particle sizes < 100 μm in different proportions (B10, B30 and B50). The combustion efficiency was measured by burnout and combustibility index (proximate analysis). The gaseous emissions of the main pollutants (CO, CO₂, SO₂ and NO) were evaluated under stoichiometric conditions and through a gas analyzer coupled to the reactor during experiments. The addition of fish scales in the blends promoted higher burnouts and combustibility rates compared to the burning of 100% coal. The sample B50 presented the highest burnout (≈ 93%), and lower carbon monoxide emissions (about 50% lower than 100% coal). However, a slight increase was observed in the CO₂, SO₂ and NO concentrations, related to the improvement in the combustion process and, due to the increase in the nitrogen quantity in the samples.; This behavior was also observed under stoichiometric conditions. However, further studies are needed to assess its real influence on pollutant emissions, for example, in sulfur dioxide concentrations.*

Keywords: *fish scale, co-firing, coal, blends, burnout, solid wastes, pollutant emissions.*

1. INTRODUCTION

Coal is the most used solid fuel in the production of electric energy in thermoelectric plants (more than 40%), as it has the greatest abundance in terms of reserves in the world and is economically attractive (Wang et al., 2014; Zellaoui et al., 2016). This primary source is also the most polluting, regarding to carbon monoxide and dioxide emissions, nitrogen and sulfur oxides, particulate materials and heavy metals (Coelho and Costa, 2007; Dai et al., 2012).

The use of urban solid waste and different biomasses is a promising and attractive alternative to reduce dependence on petroleum products that generate polluting gas emissions (Fournel et al., 2015). Furthermore, these biofuels are readily available around the world (Demirbas, 2005; Saidur et al., 2011). However, the use of pure biomass in some industrial processes is not economically viable due to the limitations of existing technologies, in addition to its low bulk density, high moisture content, low heating value, and its use (Rasul et al., 2000; Gao et al., 2010; Kazanç et al., 2011; Mortari et al., 2018).

One of the methods to enable the application of biomass in energy generation processes and reduce the use of fossil fuels is the co-combustion technique, *i.e.*, using mixtures of coal and biomass (Liu, 2002; LI et al., 2015). This technology can be immediately implemented in almost all existing coal-fired power plants, without the need for large investments (Basu, 2010; Woytiuk et al., 2017). Co-combustion is a matter of great interest worldwide and represents an alternative technology to reduce emissions of greenhouse gases, including sulfur oxides (SO_x) and nitrogen oxides (NO_x). In addition, this process offers the advantage of increasing the participation of renewable sources in energy generation (Sahu et al., 2014; Tabet and Gökalp, 2015).

Co-combustion of biomass with coal must take into account the different physical properties of the two fuels (Nzihou and Stanmore, 2015). Before introducing a certain biomass into the plant, it is necessary to characterize the new fuel as to its combustion behavior, ash deposition, possible corrosive compounds and flue gas emissions. Such information is relevant to predict the maximum levels of coal substitution, possible impacts on the plant's operation and necessary changes in the production process (Hillig et al., 2020).

Silva et al. (2019) evaluated the potential of the application of fish scales in thermochemical processes and obtained HHV values of 9.08 MJ kg⁻¹ and theoretical LHV of 8.17 MJ kg⁻¹. The sulfur content of the sample (less than 1.0%) suggests low emissions of sulfur oxides (SO_x), since these pollutants are directly related to the amount of this element present in the biofuel. The authors also observed an amorphous behavior of the material (confirmed by X-ray diffractograms and SEM images), whose calculated crystallinity index value was ≈ 11%. In thermal processes, the amorphous product has a higher reactivity than the crystalline product (Xu et al., 2013). This absence of crystalline agglomerates or crystallinity makes the material more susceptible to complex thermal degradation reactions during a combustion process (Martinez et al., 2019). Furthermore, the air mass required for complete combustion of the material was lower than that of other biomasses and coal found in the literature (Silva et al., 2020).

The higher the proportion of biomass, the lower the emission of greenhouse gases. However, co-combustion of coal-biomass mixtures presents the best results for a maximum of 20% by mass of biomass. It is estimated that the use of 10% biomass in co-combustion with coal could reduce CO₂ emissions from 45 to 450 million tons by the year 2035 (Sahu et al., 2014). This study proposed the reuse of fish scales as biofuel in co-firing processes with coal in a Drop Tube Furnace (DTF). The samples were prepared with particle sizes < 100 μm in different proportions (B10: 90% coal + 10% fish scales; B30: 70% coal + 30% fish scales, and B50: 50% coal + 50% fish scales) and some atmospheric pollutants (CO, CO₂, NO and SO₂) were evaluated, resulting from these processes and the efficiency of burning (burnout).

2. MATERIALS AND METHODS

2.1 Samples preparation

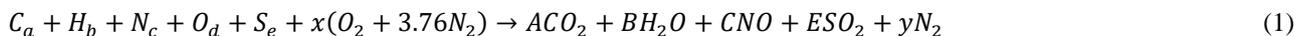
The fish scales used in this research were supplied by a local fishmonger, located in the central region of the capital of Maranhão. A samples mixture from various species of fishes was used for a more generalized analysis.

In the preparation stage: *in natura* fish scale samples went through washing in running water to remove impurities, oven drying with residence time and temperature controlled (60 °C for 48 h) to eliminate the excess moisture, grinding in a knife mill to reduce in particle sizes, and sieving for separation into average particle sizes (< 100 μm), according to ASTM D2013 (1972).

The coal samples were provided by the Combustion Laboratory of the Federal University of Rio Grande do Sul (UFRGS), in an average particle size of 75 μm and with the composition (according to immediate and elemental analyses) as shown in Table 1. The blends of coal and fish scales were prepared in the proportions: 90% coal + 10% fish scales (B10); 70% coal + 30% fish scales (B30); and 50% coal + 50% fish scales (B50).

2.2 Stoichiometry of the main gaseous emissions (CO₂, NO, and SO₂)

Equation (1) presents a global combustion reaction for biomass and solid waste, based on the ultimate analysis and formation of the main atmospheric pollutants (CO₂, NO, and SO₂) generated during these thermal processes (Cruz, 2015):



Where the indexes **a**, **b**, **c**, **d**, and **e** are, respectively, the percentages of carbon, hydrogen, nitrogen, oxygen, and sulfur divided by the respective atomic masses of each element (12, 1, 14, 16, and 32, respectively). The stoichiometric coefficients **x** and **y** are calculated to balance Equation (1), according to the linear system presented in Equation (2):

$$\begin{cases} x = \frac{1}{2} \left(2a + \frac{b}{2} + c + 2e - d \right) \\ y = 3.76x \end{cases} \quad (2)$$

With the x-value duly determined and using part of the first member of Equation (1), x (O₂ + 3.76 N₂), it was possible to calculate the air mass that will be needed for the stoichiometric combustion of the material studied, *i.e.*, the amount of oxidant mass necessary and sufficient for complete burning of all biofuel in an ideal process (Coelho and Costa, 2007; Paraschiv *et al.*, 2020). For obtaining the theoretical values of CO₂, NO, and SO₂ emissions, the second member of Equation (1) is used, *i.e.*, the products of the global combustion reaction, where **A**, **B**, **C** and **E** indexes are referred to the gaseous concentrations of the products generated, and equivalent to:

$$\begin{cases} A = a \\ B = \frac{b}{2} \\ C = c \\ E = e \end{cases}$$

Dividing each of these parameters (**A**, **C**, and **E**) by the sum of the total coefficients and not considering the moisture (**B**) (dry base), was possible to estimate the values of the air pollutants (CO₂, NO, and SO₂) under stoichiometric conditions, according to the Equations (3) to (5):

$$[CO_2]_{\%} = \frac{A}{(A + C + E + y)} \quad (3)$$

$$[NO]_{\%} = \frac{C}{(A + C + E + y)} \quad (4)$$

$$[SO_2]_{\%} = \frac{E}{(A + C + E + y)} \quad (5)$$

The concentrations of the gaseous pollutants are given in terms of mass unit per volume unit (mg Nm⁻³) (Carvalho Júnior and Lacava, 2003). It is interesting to highlight, that 1 gas mole corresponds to 22.4x10⁻³ m³ and denoting by [A]_{db}, the concentration (ppm) of the specific pollutant on a dry basis as shown in Equation (6):

$$[A]_{db} \text{ (mg Nm}^{-3}\text{)} = \frac{[A]_{db}}{22.4 * 10^{-3}} (M_A * 10^3) = 4.464 * 10^4 M_A [A]_{db} \quad (6)$$

Where M_A is the molecular mass of the component emitted (g gmol⁻¹).

2.3 Combustion process in a Drop Tube Furnace (DTF)

The combustion profile of the blends was analyzed in a Drop Tube Furnace (DTF), due to its ability to simulate burning processes in boilers. The used DTF (Figure 1a-b) consists of a cylindrical ceramic combustion chamber, electrically heated by kanthal resistors, with temperature control consisting of three parts, each with three uniformly distributed K-type thermocouples: one for controlling and two for monitoring the wall furnace temperature.

The reactor has a useful length of 1232 mm and an inner diameter of 48 mm. The feed system consists of a concentric tube injector in 310 stainless-steel, water-cooled, coupled to the top of the reactor, supplying the solid fuel to the combustion chamber. The samples are injected into the reactor by a pneumatic system, composed by a brass tube, which moves vertically through a power screw inside of a fixed glass tube. The controlled air flow applied concentrically inside the brass tube drags the biomass particles to the injector and later to the combustion chamber.

The combustion of coal samples and blends (B10, B30 and B50) was evaluated at 1100 °C and residence time of 500 ms. The sample mass flow rate applied was 15 g h⁻¹, with feeder screw rotation of 0.10 rps, air flow of 30 L min⁻¹ and experiment time of 20 min.

The samples were collected along the reactor during experiments by means of a stainless-steel probe, water-cooled. Immediately after enter in the probe, the collected samples received a jet of nitrogen, with a flow rate of 6 L min⁻¹ to stop the particle combustion reaction. Then, the collected ashes were deposited in a quartz filter for subsequent analysis of the burning yield (burnout), determined by the ash tracer method (Cloke et al., 2002; Biswas et al., 2006; Osório et al., 2006), according to Equation 7:

$$\Psi = \left[1 - \left(\frac{\omega_f}{100 - \omega_f} \right) * \left(\frac{100 - \omega_x}{\omega_{w_x}} \right) \right] \quad (7)$$

where ω_f and ω_x are the mass fractions of ash (dry basis) in the original sample of biomass and in the collected residue from combustion, respectively.

After 20 min collecting ash from the experiment, the nitrogen jet in the probe was stopped to enable the analysis of the combustion gases, considering the same experimental conditions used for the burnout analysis. After collection, the gases from the DTF passed through a conditioning system, composed of filters and condensers, and were analyzed by a Siemens gas analyzer (NO, SO₂, O₂ and CO₂) and Kane gas analyzer (CO).

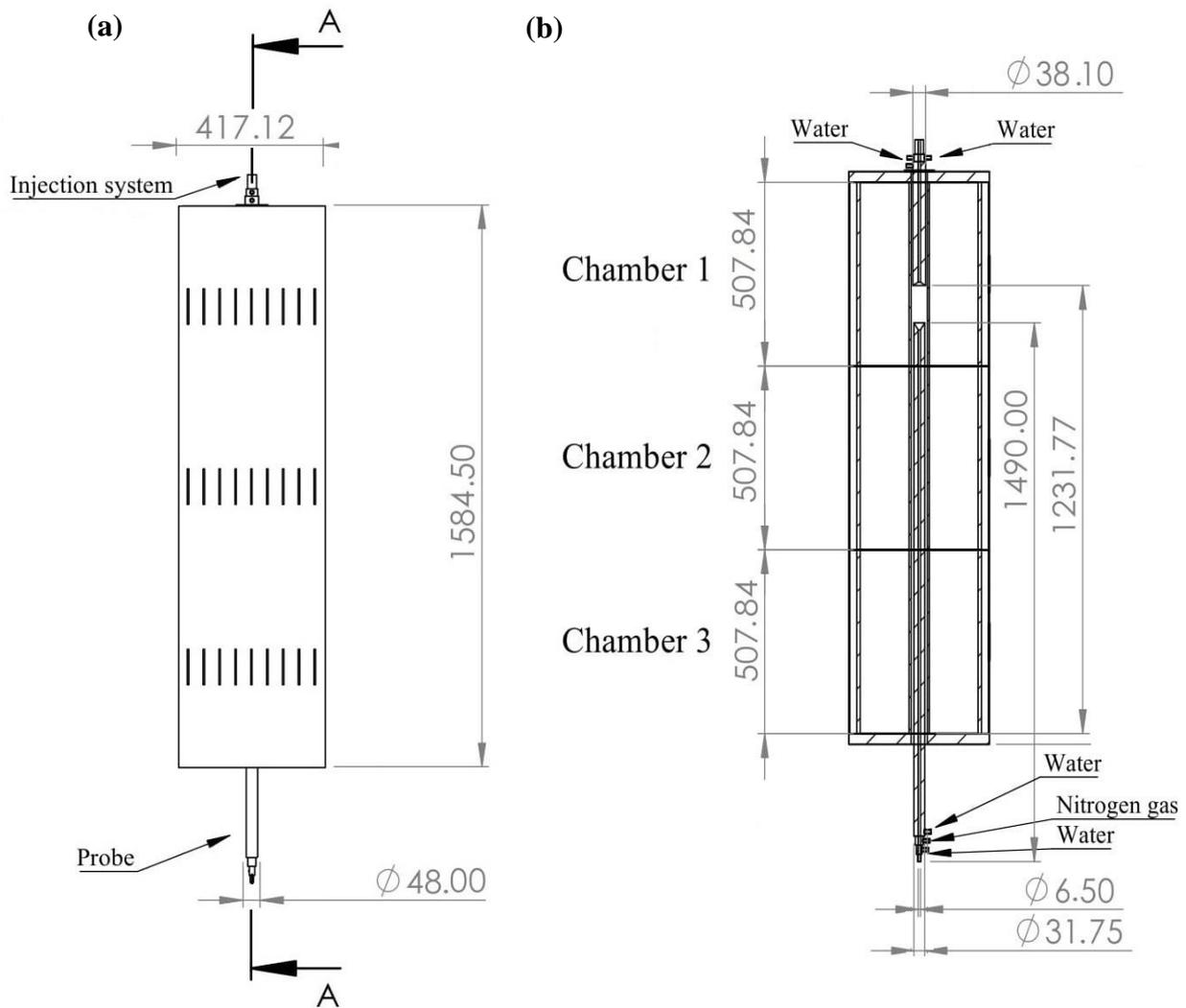


Figure 1. Schematic representation of a Drop Tube Furnace – DTF: (a) side view and (b) section.

3. RESULTS AND DISCUSSION

Table 1 shows the properties of coal samples and blends with fish scales in the proportions of 10, 30 and 50%, which were obtained by proximate and ultimate analysis. The addition of fish scales resulted in an increase in the physicochemical properties of the samples such as moisture content (4.06 to 7.70%), volatile materials (27.66 to 36.33%) and ash (32.06 to 38.98%), and reduction in the amounts of fixed carbon (40.28 to 23.94%). Despite the increase, the resulting moisture from blends remained below 10%, this percentage is ideal for application in burning processes (Garcia et al., 2012; Yao et al., 2005). Furthermore, with the increase in volatile material, the spread of combustible gases also increased during combustion, considerably enriching this process (Garcia et al., 2012).

Regarding the ultimate analysis of the blends, the addition of fish scales resulted in an increase in the amount of oxygen (40.98 to 55.02%) and nitrogen (0.87 to 3.59%), and a reduction in the content of carbon (53.80 to 37.06%). The amounts of hydrogen (3.52%) and sulfur ($\approx 0.80\%$) not changed significantly because of the similar compositions between the coal used and fish scales. It is important to emphasize that high amounts of oxygen reduce the heating value of a fuel due to the high values of the binding enthalpies, requiring greater activation energy to break the bonds formed by its atoms, for example, CO bond (Mckendry, 2002; Atkins and Jones, 2012; Braz and Crnkovic, 2014).

Table 2 presents the stoichiometric coefficients obtained by Equations 1 and 2 for the coal samples and blends. There was a reduction in the oxygen (x) and nitrogen (y) balances with the increase in the amount of added scales. These parameters directly influence the formation of the main atmospheric pollutants (CO_2 , SO_2 and NO) emitted under stoichiometric conditions. In addition, the air mass required to promote complete combustion of the solid fuel also decreased (568.30 kg for 100% coal to 329.81 kg for B50). This reduction is also related to the carbon content in the

sample, since these are directly proportional, *i.e.*, the higher the carbon content of the material, the greater the air-fuel ratio necessary for the total burning of the material (Silva et al., 2020).

Table 3 shows the concentrations of gaseous emissions of CO₂, SO₂ and NO, under stoichiometric conditions, for coal and blends in a dry and volumetric basis, while Figure 2 presents the levels of air pollutants in absolute values. Among the three main theoretical air pollutants analyzed, CO₂ had the highest values.

Table 1. Composition of coal samples and blends with fish scales in proportions of 10, 30 and 50%.

	Coal	B10	B30	B50
Proximate analysis (%)				
Moisture	4.06	4.79	6.24	7.70
Volatile material (VM)	27.66	29.39	32.86	36.33
Fixed carbon (FC)	40.28	37.01	30.47	23.94
Ash	32.06	33.44	36.22	38.98
VM + FC	67.94	66.40	63.33	60.27
Ultimate analysis (%)				
Carbon	53.80	50.45	43.76	37.06
Hydrogen	3.52	3.52	3.52	3.52
Oxygen*	40.98	43.79	49.40	55.02
Nitrogen	0.87	1.41	2.50	3.59
Sulfur	0.83	0.83	0.82	0.81

* O * = 100 – (%C + %H + %N + %S)

(Silva et al., 2019; Hillig et al., 2020)

Table 2. Stoichiometric coefficients, air mass and air-fuel ratio for coal samples and fish scales blends.

Samples	a	b	c	d	e	x	y	Air mass* (kg)	Air-fuel ratio
Coal	4.83	3.52	0.06	2.56	0.02	4.14	15.56	568.30	5.68:1
B10	4.20	3.52	0.10	2.74	0.02	3.79	14.26	520.57	5.20:1
B30	3.65	3.52	0.18	3.09	0.02	3.10	11.65	425.27	4.25:1
B50	3.09	3.52	0.26	3.44	0.02	2.40	9.03	329.81	3.30:1

Table 3. Gaseous emissions (CO₂, SO₂ and NO) from coal and blends under stoichiometric conditions.

Samples	CO ₂ (%)	SO ₂ (ppm)	SO ₂ (mg Nm ⁻³)	NO (ppm)	NO (mg Nm ⁻³)
Coal	22.26	1288.07	3683.87	3086.04	6326.38
B10	22.62	1395.33	3990.36	5418.00	11106.90
B30	23.53	1653.36	4728.62	11521.71	23619.50
B50	24.90	2040.77	5836.60	20674.02	42381.75

It is possible to notice an increase in the concentrations of the three gases analyzed (CO₂, SO₂ and NO) with the addition of fish scales in the blends, with the B50 sample being responsible for the highest emissions, namely: 24.90% for CO₂; 2040.80 ppm for SO₂ and 20674.02 ppm for NO. As observed in Table 2, coal needed more air mass to oxidize the entire carbon content, turning it into CO₂. The increase in the proportions of fish scales reduced the amount of carbon in the blends and, consequently, the air-fuel ratio needed to promote the complete burning of that fuel.

The increase in the concentration of NO emissions was due to the increase in the nitrogen content in the blends caused by the presence of fish scales, which presented a greater amount of this element in the elemental composition when compared to coal: 6.32 and 0.87% for scales and coal, respectively (Liu et al., 2002; Zhao et al., 2016; Yang et al., 2018). Greater formations of carbon dioxide (CO₂) and sulfur (SO₂) are indicative of complete or partially complete combustion (Williams et al., 2012; Sartor et al., 2014; Cruz, 2018; Krupal et al., 2019). Therefore, the analysis of emissions under stoichiometric conditions suggested that burning coal together with fish scales was more efficient compared to burning pure coal.

The burnout in Figure 3 shows that the addition of fish scales to the coal blends influenced the combustion efficiency at 1100 °C and 500 ms. A strong upward trend in burning efficiency was observed with the increase in the amount of fish scales in the blends. The lowest efficiency occurred for burning 100% coal, with 84.40%, and the highest for B50, with 92.94% (about 10% higher than coal). The other values measured were 89.92% for B10 and 91.21% for B30. The best repeatability occurred for B10 and B30, however, all errors remained below 5%.

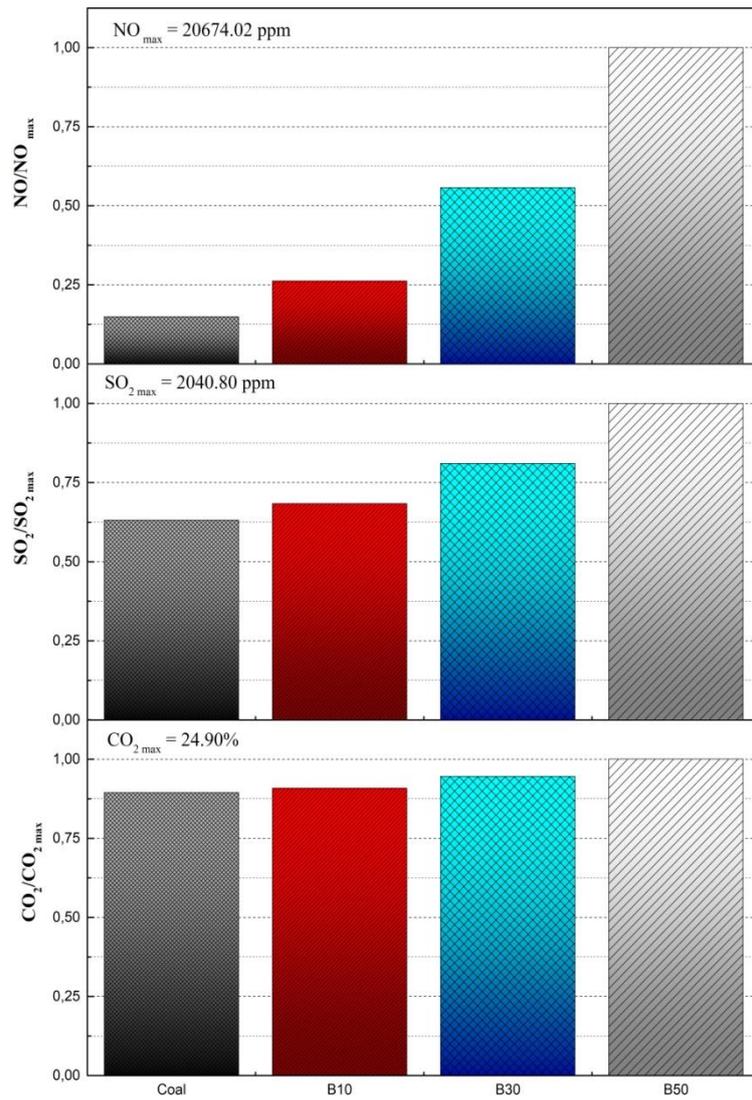


Figure 2. Gaseous emissions under stoichiometric conditions for coal samples and blends with fish scales.

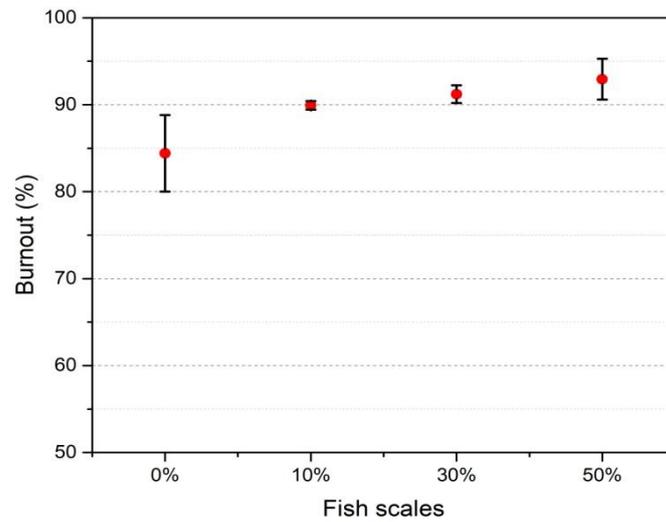


Figure 3. Burnout for coal samples and blends at 1100 °C and 500 ms.

The highest burnouts obtained are related to the higher volatile material (VM) content and lower fixed carbon (FC) content of the blends compared to 100% coal (Mortari et al., 2021). The index (dimensionless) associated with these two parameters is the Combustibility Index (CI), which is defined as the ratio between the content of volatile material and fixed carbon and described by Equation 8:

$$CI = \frac{VM}{FC} \quad (8)$$

Through this parameter, it is possible to compare the energy potential of different materials or substances, so that the higher the combustibility index, the more reactive the sample will be in combustion processes and the better its burning (González and Montes, 2019). The combustibility index for coal and blends is presented in Figure 4 and shows how the addition of fish scale scales helped to improve the burning efficiency for the studied samples.

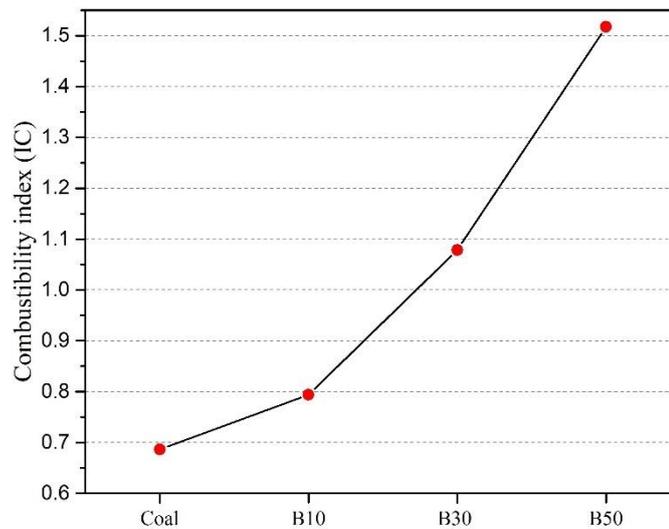
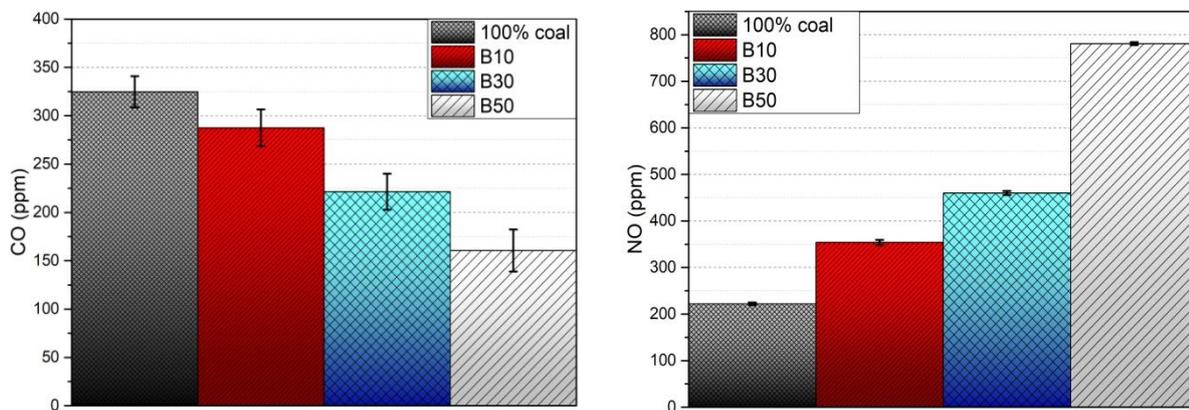


Figure 4. Combustibility index for coal and fish scales blends.

Fish scales exhibit important characteristics in their composition that contribute to improving the efficiency of burning coal, among them some can be highlighted, for example, the presence of elemental calcium and calcium oxides (CaO), which influence the decomposition rate in the devolatilization stage at temperatures below 400 °C (Werther et al., 2000; Yuan et al., 2019). In addition, as the fish scales are a mostly amorphous material (crystallinity index \approx 11%), these showed greater reactivity in thermal processes, making this material more susceptible to complex reactions of thermal degradation during the combustion process (Xu et al., 2013; Martinez et al., 2019; Silva et al., 2019).

Figure 5(a-d) shows the emissions concentrations of carbon dioxide (CO₂) and sulfur (SO₂), carbon monoxide (CO) and nitrogen (NO) for the combustion of coal samples and fish scale blends (B10, B30 and B50) in a DTF (Drop Tube Furnace) at 1100 °C and a residence time around 500 ms. The values shown in Figure 5 were corrected for 10% excess oxygen (O₂).



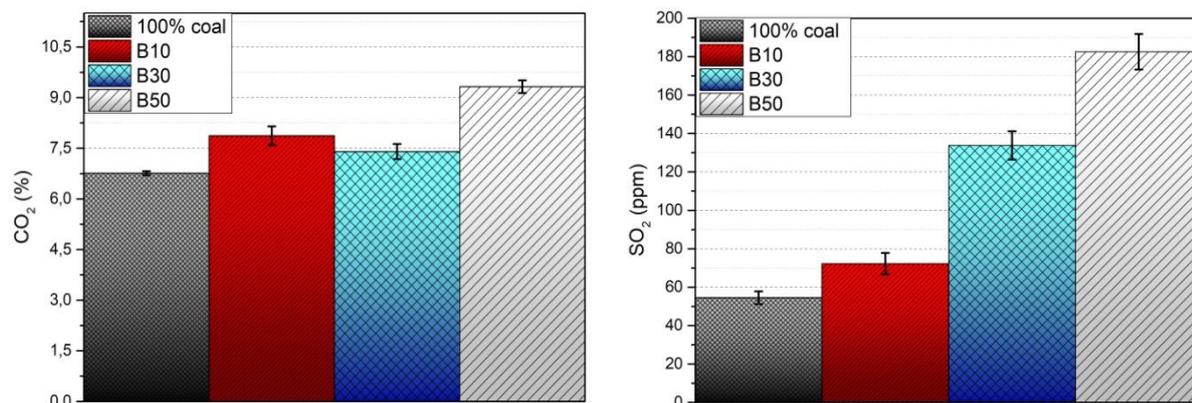


Figure 5. Gaseous species concentrations obtained at 1100 °C and 500 ms, corrected to 10% of O₂: (a) CO₂, (b) SO₂, (c) CO and (d) NO.

As observed in the combustion process under stoichiometric conditions (Figure 2), there was an increase in the concentrations of carbon dioxide and sulfur emissions, and nitrogen monoxide, as the quantities of fish scales were added to the blends. The variations for the gases evaluated were: 6.76 to 9.32% for CO₂; 54.55 to 182.52 ppm for SO₂; and 222.01 to 780.89 ppm for NO.

B10 and B30 samples presented similar emission concentrations of carbon dioxide (7.87 and 7.40%, respectively). Regarding the burning of pure coal and the B50, CO₂ was the one with the lowest growth among the analyzed gases (around 37%). Fish scales present calcium oxides (CaO) in abundance in their composition and, according to the literature, materials rich in CaO, when in burning processes together with fossil fuels, contributes for the capture of CO₂ (Yuan et al., 2019).

Emissions of nitrogen monoxide and sulfur dioxide showed growth above 200% in their respective concentrations, in relation to the burning of 100% coal and B50. As observed under stoichiometric combustion, the increase in NO was related to the amount of nitrogen in the sample composition (0.87% for pure coal and 3.59% for B50). However, the increase in SO₂ can be correlated to the improvement in burning efficiency with the addition of fish scales to the blends, as shown in Figure 5, as higher emissions of this gas are indicative of complete combustion (Williams et al., 2012; Sartor et al., 2014). However, knowing that fish scales are rich in calcium and that SO₂ emissions decrease with increasing concentration of this mineral (Kazanc et al., 2011), further studies are needed on the real influence of fish scales on blends and, consequently, in the generation of this pollutant.

Regarding carbon monoxide, the addition of fish scales in the blends with coal promoted a reduction of approximately 50% in the concentration of emissions of this pollutant. The values obtained for CO ranged from 160.55 to 324.77 ppm, with the B50 and 100% coal presenting the lowest and highest value, respectively. The decrease in the amount of CO emitted in the thermal process confirmed the improvement in burning efficiency observed in Figures 3 and 4, since high emissions of this gas directly reflect on poor/incomplete combustion of the analyzed material (Carvalho Jr and Lacava, 2003; Coelho and Costa, 2007).

It is also interesting to point out that even with better burning efficiency and lower carbon monoxide emissions, the B50 resulted in high concentrations of carbon dioxide and nitrogen monoxide in relation to pure coal and other blends. In addition, B10 and B30 samples were sufficient to ensure burnouts close to 90% and lower concentrations of the main air pollutants mentioned in this study.

4. CONCLUSIONS

This study proposed a sustainable and ecofriendly reuse of fish scales as a possible alternative energy source, using them in thermochemical processes (combustion) in a Drop Tube Furnace for the bioenergy generation. The addition of 10% of fish scales in the blends with coal favored the combustion process, resulting in 90% efficiencies and reduction in carbon monoxide emissions, compared to burning 100% coal. However, the higher proportions of scale (30 and 50%) in the blends resulted in high amounts of nitrogen oxides and sulfur dioxide. Therefore, the DTF results showed that the process of burning fish scales is a viable way to use these residues, which can favor the co-burning of coal and reduce pollutants emissions such as carbon monoxide.

5. ACKNOWLEDGEMENTS

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