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THEORETICAL STUDY FROM GASEOUS EMISSIONS GENERATED BY FISH SCALES UNDER STOICHIOMETRIC COMBUSTION PROCESSES

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Abstract. *Fish production in the State of Maranhão is one of the most representative economic activities from the Brazilian coast. During the fishes processing, the amount of residues generated can reach 50% by mass of the total production. Fish scales are considered useless and impractical, being discarded in a disordered or unplanned manner in inappropriate places, such as in dumps or even thrown into the sea. The present study proposed to address this material in a sustainable and eco-friendly reuse, as a possible alternative energy source under thermochemical conversion processes for the bioenergy generation. The samples were prepared with an average granulometry (363 µm), and their chemical composition determined by ultimate analysis. The theoretical analysis performed under stoichiometric conditions allowed to estimate the main pollutants (CO_2, NO and SO_2), and the air mass necessary for the complete combustion of the fish scales. It was possible evaluating the influence of elements, e.g., nitrogen and sulfur from the sample composition in the generation of these gases, and the importance of carbon content in carbon dioxide emissions and air mass. Therefore, the information presented in this preliminary study can contribute as a basis for further experimental analysis in real thermal systems or on laboratory scale.*

Keywords: *bioenergy, pollutant emissions, solid wastes*

1. INTRODUCTION

Maintaining global security of energy supply has become a huge challenge due to the instability of the current scenario of the traditional fossil fuels (coal, oil, and natural gas) (Bharathiraja *et al*., 2018). The growing concern with the protection of the environment, climate change and the health of living beings have led to large investments in alternative means of electricity generation, reducing the demand for these primitive sources, which are responsible for the emissions of greenhouse gases, particulate matters, and other components harmful to the environment (Lela *et al*., 2016; Williams *et al*., 2012; Lopes, 2016). In this context, the use of solid wastes and different biomasses is a promising and attractive alternative to reduce dependence on petroleum-based productsand decrease the pollutant emissions (Fournel *et al*., 2015). Also, these biofuels are readily available worldwide (Demirbas, 2005; Saidur *et al*., 2011).

In this scenario, the global fishing industry generates large amounts of residues, which are rich in nutrients and their disposal is carried out inappropriately, representing serious risks to the environment and human health, whether these are not properly treated, since the generation rate (waste and by-products) is greater than the degradation rate (Fiori *et al*., 2008; Gumisiriza *et al*., 2009; Arumugam and Ponnusami, 2017; Kara *et al.*, 2018; Castañeda *et al*., 2016). In 2016, the world production of fish was around 171 million tons, 88% of which were destined for the human consumption and 12% was used in non-food products, for example, silage, leather, and oils (Lustosa-Neto *et al*., 2018; Ching-Velasquez *et al*., 2020).

According to a survey carried out by the Brazilian Association of Pisciculture (ABP, 2020), the national fishing industry grew 4.9% concerning the previous year (2019), reaching 758,006 tons of fish produced, with tilapia being the main species, with a total of 432.149 tons (57% of all Brazilian fish farming), which consolidated Brazil as the 4th (fourth) largest producer in the world in this category (behind China, Indonesia, and Egypt, in order). Fish, as a whole, exported around US$ 275 million (2019), with fish farming being the second most important segment, representing US$ 12 million (4% total production).

The production of fish in the State of Maranhão is one of the most economically representative activities carried out on the Brazilian coast, not only by quantity (in tons), but also in species diversity (Almeida, 2008). For example, the productivity of yellow hake (*Cynoscion acoupa*), the most appreciated by the local population, was estimated at 3.565 tons year^{-1} (Almeida *et al*., 2011), tilapia (*Oreochromis niloticus*) at 4.019 t year^{-1}, fish natives (*tambaqui* and *curimatã*) in 38,511 tons year^{-1}, and other species (carp, trout, panga etc) in 2,470 tons year^{-1}, being the 6th (sixth) largest production in the country with 45,000 tons (ABP, 2020). Fish farming in Maranhão grew 15.2% compared to 2018, with emphasis on native fish, whose state is the 3rd (third) largest Brazilian producer.

During fish processing, the amount of organic matter discarded in the form of solid waste (heads, tails, fins, viscera, skins, and scales) can reach 50% w/w of the total production (Bermúdez-Penabad *et al*., 2017; Gumisiriza *et al*., 2009; Leite *et al*., 2016). Depending on the fish type and treatment level, these tailings can reach 80% of the initial feedstock (Bhagwat and Dandge, 2016; Pickler and Filho, 2017; Sockalingam and Abdullah, 2015).

Around 76 million tons of fish waste was generated in 2016 (FAO, 2018). These tailings contain approximately from 40% to 65% lipids and fats and can be converted into biodiesel, where the transesterification process is the most common and used for the bio-oil extraction (Yahyaee *et al*., 2013; Martins *et al*., 2015; Iastiaque *et al*., 2015; Wisniewski *et al*., 2010). The scales correspond to about 4% of the dry residues (Ghaly *et al*., 2013). Generally, these remains of material are considered useless and impractical, being discarded in a disorderly and/or unplanned manner in inappropriate places, such as in dumps or even thrown into the sea (Ghaly *et al*., 2013; Holá *et al*., 2011).

Therefore, the effective use of this biomass based on fish scales (in general) can reduce socio-environmental vulnerabilities and maximize the economic activities of this productive sector. The current study proposes the sustainable and ecofriendly reuse of this material, as an alternative energy source and using it under thermochemical conversion processes (direct combustion) for the bioenergy generation. The main atmospheric pollutants (CO_2, NO, and SO_2), resulting from this thermal process were also estimated through of a theoretical analysis under stoichiometric conditions.

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 fish was used for a more generalized analysis.

In the preparation stage (Fig. 1): (1) *in natura* fish scale samples went through (2) washing in running water to remove impurities, (3) oven drying with residence time and temperature controlled (60 ºC for 48 h) to eliminate the excess moisture, (4) grinding in a knife mill to reduce in particle sizes, and (5) sieving for separation into (6) average particle sizes (≈ 363 μm), according to ASTM D2013 (1972).

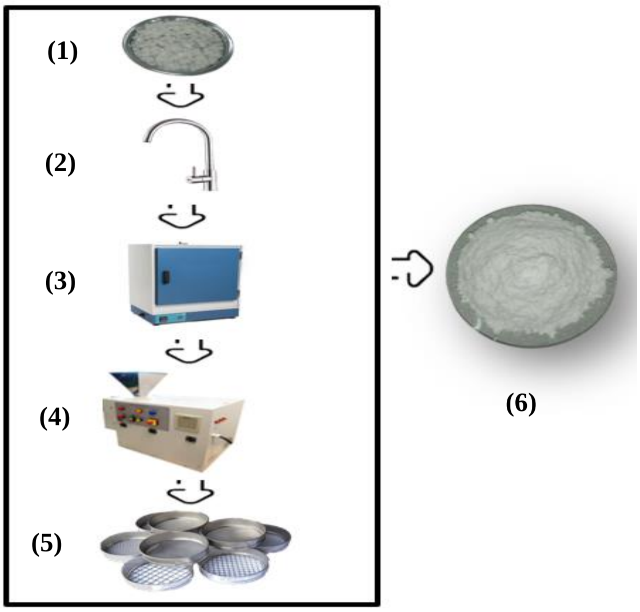

Figure 1. Samples preparation step: (1) *in natura* fish scales, (2) washing, (3) drying, (4) milling, (5) sieving, and (6) average granulometry.

2.2 Ultimate analysis

The contents of carbon (C), hydrogen (H), nitrogen (N), and sulfur (S) present in the material were quantified by an elemental analyzer, Perkin Elmer brand - 2400 CHNS-O model, sample mass of 2.0 ± 0.5 mg. The oxygen content was determined by difference in 100%, considering other elements provided by this analysis (Ghetti *et al*., 1996).

2.3 Stoichiometry of the main gaseous emissions (CO_2, NO, and SO_2)

Equation (1) presents a global combustion reaction for biomass and solid waste, based on the ultimate analysis and formation of the main atmospheric pollutants (CO_2, NO, and SO_2) generated during these thermal processes (Cruz, 2015):

$$C_a + H_b + N_c + O_d + S_e + x(O_2 + 3.76N_2) \rightarrow ACO_2 + BH_2O + CNO + ESO_2 + yN_2 \quad (1)$$

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):

$$\begin{cases} x = \frac{1}{2}\left(2a + \frac{b}{2} + c + 2e - d\right) \\ y = 3{,}76\,x \end{cases} \quad (2)$$

With the x-value duly determined and using part of the first member of Equation (1), x (O_2 + 3.76 N_2), 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 CO_2, NO, and SO_2 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**), was possible to estimate the values of the air pollutants (CO_2, NO, and SO_2) 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} m^3 and denoting by $[A]_{db}$, the concentration (ppm) of the specific pollutant on a dry basis as shown in Equation (6):

$$[A]_{db\,(mg\,Nm^{-3})} = \frac{[A]_{db}}{22{,}4\,.\,10^{-3}}(M_A\,.\,10^3) = 4{,}464\,.\,10^4 M_A [A]_{db} \quad (6)$$

Where M_A is the molecular mass of the component emitted (g $gmol^{-1}$).

3. RESULTS AND DISCUSSION

Table 1 presents the percentages of carbon, hydrogen, nitrogen, sulfur, and oxygen obtained by ultimate analysis for the fish scale samples, in addition the percentage values of other biomasses and a coal type, in order to compare the respective emissions according to the stoichiometry of the combustion process. Through this analysis, it was possible to observe that the compositions of the solid fuels differ considerably, mainly among lignocellulosic biomasses. This variety of properties makes it difficult to establish perfect reference values (Obernberger *et al*., 2006; Vassilev *et al*., 2010). The compositions ranged from ≈ 20 to 55% for carbon; 0.6 to 6.0% for hydrogen; 0.2 to 6.0% for nitrogen, and 0.1 to 1.5% for sulfur. However, the oxygen content ranged from 39 to 69%.

Table1. Ultimate analysis of fish scales, biomasses, and coal type.

Fuels	C (%)	H (%)	N (%)	S (%)	O (%)	Chemical formula
Fish scales	20. 32	3.52	6.32	0.79	69.06	$C_{1.69}H_{3.52}O_{4.32}N_{0.45}S_{0.02}$
Tomato plant waste	36.63	0.68	1.19	1.48	60.01	$C_{3.05}H_{0.68}O_{3.75}N_{0.08}S_{0.05}$
Sugarcane bagasse	45.05	5.57	0.25	0.07	49.06	$C_{3.75}H_{5.57}O_{3.07}N_{0.02}S_{0.00}$
Coffee husk	43.13	5.93	1.55	0.67	51.28	$C_{3.59}H_{5.93}O_{3.20}N_{0.11}S_{0.02}$
Coal CE4500	54.96	3.36	1.07	1.18	39.43	$C_{4.58}H_{3.36}O_{2.46}N_{0.08}S_{0.04}$

(Silva *et al.*, 2019; Cruz, 2015; Garcia *et al.*, 2012; Mortari *et al.*, 2020)

The fish scales are close to the residues from the tomato harvest and showed the lowest carbon percentages (20.32 and 36.63%, respectively), and the highest oxygen (69.06 and 60.01%), while coal showed the highest and lowest C (54.96%) and O (39.43%), respectively. Tomato harvest residues also exhibited the lowest hydrogen content (0.68%). Sugarcane bagasse and coffee husk showed similar levels of carbon (45.05 and 43.13%), hydrogen (5.57 and 5.93%), and oxygen (49.06 and 51.28%). The amounts of sulfur were close to 1.0% for all samples, with the exception for the sugarcane bagasse (0.07%), which also indicated the lowest nitrogen value (0.25%).

Table 2 shows the stoichiometric coefficients obtained through Equations (1) and (2) for the fish scales and different solid fuels. Among the oxygen (x) and nitrogen (y) balances, fish scales had the lowest values (0.66 and 2.50), and coal the highest (4.26 and 16.03). These parameters directly influence the air mass, air-fuel ratio, and the amount of the main pollutants (CO_2, NO, and SO_2) emitted by solid fuels under stoichiometric conditions.

Table 2. Stoichiometric coefficients, air mass, and air-fuel ratio obtained for the different fuel types.

Fuels	a	b	c	d	e	x	y	Air mass* (kg)	Air-fuel ratio
Fish scales	1.69	3.52	0.45	4.32	0.02	0.66	2.50	91.32	0.91:1
Tomato plant waste	3.05	0.68	0.08	3.75	0.05	1.44	5.40	197.12	1.97:1
Sugarcane bagasse	3.75	5.57	0.02	3.07	0.00	3.62	13.63	497.59	4.98:1
Coffee husk	3.59	5.93	0.11	3.20	0.02	3.55	13.35	487.41	4.87:1
Coal CE4500	4.58	3.36	0.08	2.46	0.04	4.26	16.03	585.21	5.85:1

*Air mass required for 100 kg of fuel

(Silva *et al.*, 2019; Cruz, 2015; Garcia *et al.*, 2012; Mortari *et al.*, 2020)

The amount of air necessary for promoting the complete combustion of a certain substance or fuel, is of paramount importance, since whether a sufficient amount of oxygen is not guaranteed, the burning will be incomplete. On the other hand, whether an amount of air is introduced in excess, heat part will be absorbed and the combustion temperature may be decreased, increasing the volume of the combustion gases and resulting mainly from CO_2 and water vapor (Strobel *et al*., 2018; Barreto, 2008). Different fuels commonly present unlike air-fuel ratios (Barreto, 2008). Figure 2 shows the air mass required for the complete burning of the different fuels.

It was observed that the largest amount of air mass required occurred for the coal sample (585.21 kg), while the smallest was attributed to fish scales (91.32 kg). Among the other biomasses, the tomato harvest residues showed a lower value 187.12 kg compared to 487.41 kg for coffee husks, and 497.59 kg for sugarcane bagasse. It was also possible to note that this parameter is directly proportional to the carbon composition presented by the samples, *i.e*., the higher the content of this element, the greater the air-fuel ratio necessary for the complete burning of the fuel.

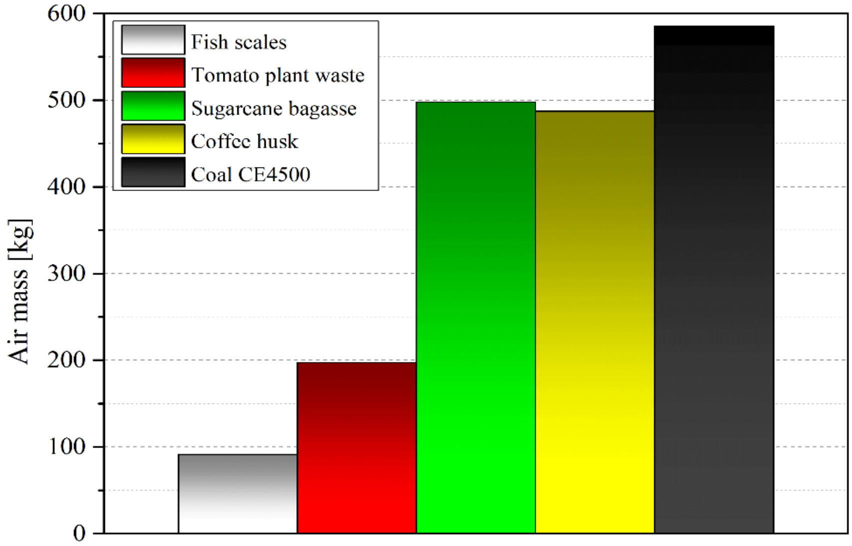

Figure 2. Air mass required for the complete combustion of the different biomass biofuels and coal type.

In real thermal processes, it is unusual to obtain an adequate burning, using only stoichiometric air, commonly, excess air is used to increase the complete combustion or control the chamber temperature (Ionita, 1990; Coelho and Costa, 2007). However, the greater the excess air supplied to the system, greater the CO_2 concentration and lower thermal efficiency of the boiler, making it necessary to seek other resources or tools, for example, adjust of the flame temperature to avoid incomplete combustion (Coelho and Costa, 2007; Silva Segundo, 2014; Salum, 2011).

Figure 3 shows the levels of carbon dioxide (CO_2) emissions under stoichiometric conditions. Among the three main theoretical air pollutants analyzed, CO_2 presented the highest values. The formation of this gas is a strong indicative of a complete combustion (Krumal *et al*., 2020; Cruz, 2015).

For the different fuels evaluated, fish scales and tomato plant residues showed the highest relative CO_2 emission values (710.679 and 697.074 mg Nm^{-3}, respectively) due to the low air dilution demanded for the complete combustion. However, sugarcane bagasse, coffee husks, and coal pointed out lower values, presenting similar compositions for this pollutant (between 412,000 and 433,000 mg Nm^{-3}).

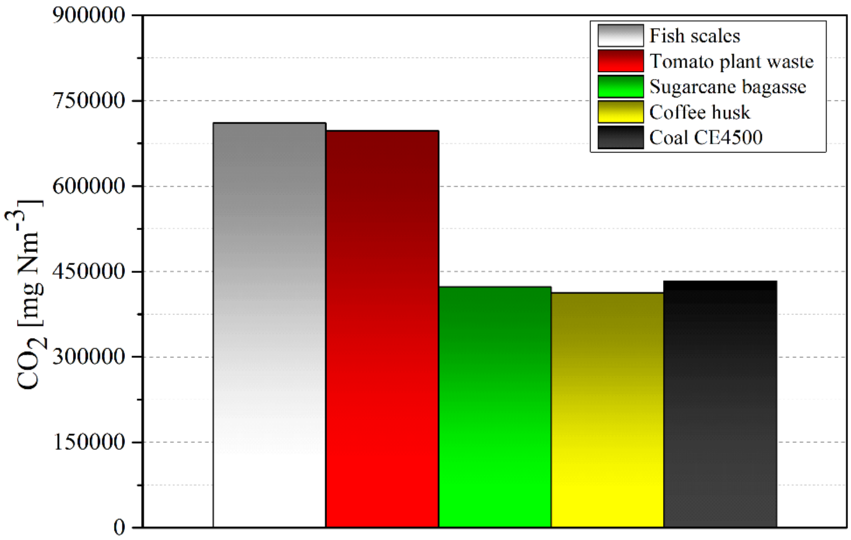

Figure 3. Carbon dioxide (CO_2) emissions calculated for the different solid fuels.

It is understood that the estimated values of air pollutants under stoichiometric conditions is inversely proportional to the sum of the total coefficients (A + C + E + y), as shown in Equations (3) to (5) and reflects the dilution of the exhaust gases. As noted in Fig. 4, coal needs more air to convert all its carbon content into CO_2, causing the balances of oxygen (x) and nitrogen (y) to be greater concerning fish scales (around six times). Thus, the carbon dioxide parcels emitted are about 36% for the fish scales and residues from the tomato harvest, and 22% for sugarcane bagasse, coffee husks, and coal type (CE4500).

Combustion is a thermal process that inherently emits CO_2. One of the currently most promising techniques for reducing carbon dioxide emissions generated in the conventional combustion process (79% N_2/21% O_2) is the oxy-fuel combustion, in which the percentage of N_2 in the air is replaced by a mixture of pure oxygen and flue gases recirculated and/or captured, for example, CO_2 and water vapor (Baskar and Senthilkumar, 2016; Zhang *et al*., 2019; Jang *et al*., 2016; Andersen *et al*., 2017; Engin and Atakul, 2018).

According to Fernández *et al*. (2012), the biomass combustion can be considered neutral for CO_2 emissions. The carbon dioxide released into the atmosphere during the burning of these biofuels can be absorbed by plants during the photosynthesis process, contributing to the maintenance of the concentration of this pollutant (Foletto *et al*., 2005).

The levels of nitrogen monoxide (NO) emissions are shown in Figure 4. For this pollutant, the theoretical values ranged from 2.103 to 8197.847 mg Nm^{-3}, with sugarcane bagasse and fish scales presenting the lower and higher limits, respectively. The other contents measured were 15.411 mg Nm^{-3} for the residues of tomato plants, 13.291 mg Nm^{-3} for coffee husk, and 7.561 mg Nm^{-3} for coal CE4500.

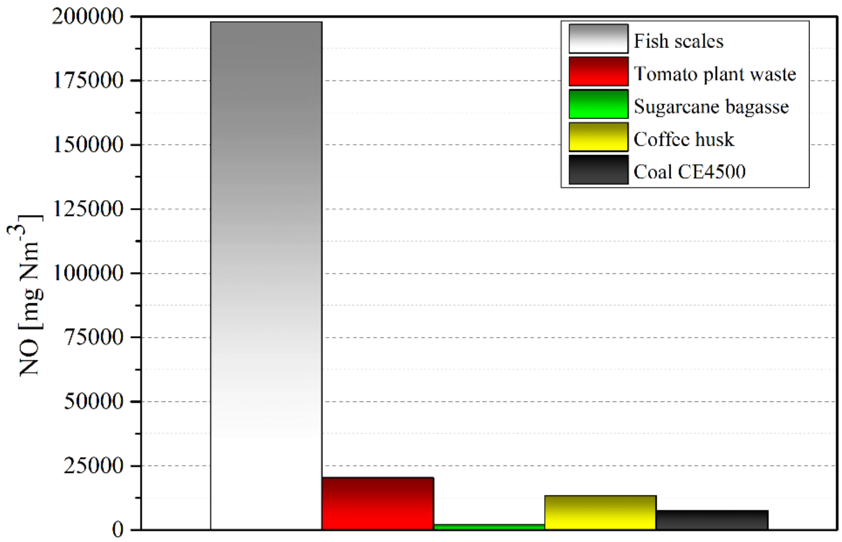

Figure 4. Nitrogen monoxide (NO) emissions estimated for the different solid fuels.

Emissions of nitric oxides (NO_x) are mainly due to the nitrogen content (NO-fuel) of the sample, *i.e*., a higher elemental nitrogen content results in high amounts of NO (Yang *et al*., 2018; Zhao *et al*., 2016; Liu *et al*., 2002). Fish scales presented the highest compositional value (6.32%) and sugarcane bagasse the lowest (0.25%), concerning the other fuels analyzed, which corroborates with the literature. Kazanc *et al*. (2011) also showed NO values below 2000 mg Nm^{-3} for woody biomass and coal under combustion atmosphere. The authors also observed that, in some cases, NO_x emissions do not reflect the nitrogen content of the sample, which was consumed in the combustion process.

In real thermal processes, the formation of NO can also be affected by the combustion chamber temperature and atmospheric O_2 concentration (Duan *et al*., 2015; Ahn *et al*., 2011). According to Kazanc *et al*. (2011), in combustion processes, the portion of nitrogen present in the fuel is distributed between the devolatilization and oxidation of the solid carbon, and in both phases depends exclusively on the material type, reactor temperature, and residence time of the burning reaction inside the combustion chamber.

According to Martins and Ferreira (2010), the NO emitted during combustion also depends on the fixed carbon content of the material. Consequently, agricultural residues can emit high concentrations of NO_x concerning coal, even containing similar amounts of nitrogen, this is due to the low fixed carbon content in these biomasses, making the catalytic effects of coal in reducing this pollutant negligible (Werhther *et al*., 2000).

The estimated sulfur dioxide (SO_2) emission rates are shown in Figure 5. The higher and lower limits of this pollutant were observed, respectively, for the residues from the tomato harvest (15.411 mg Nm^{-3}) and sugarcane bagasse (359 mg Nm^{-3}). However, knowing that the sulfur content (S) of the fuel is the main factor influencing this element for the emissions of this gas (Yang *et al*., 2018; Zhao *et al*., 2016). It was expected that coal presented a value close to the maximum amount, since the elemental composition of this fuel (1.18% sulfur) was the highest compared to the other materials studied (fish scales = 0.79%, coffee husks = 0.67%, and sugarcane bagasse = 0.07%). Fish scales showed SO_2 emissions around 15,118 mg Nm^{-3}. This behavior confirms the influence of the other elements (C, H, N, and O) on emissions under stoichiometric conditions, according to what was discussed for CO_2.

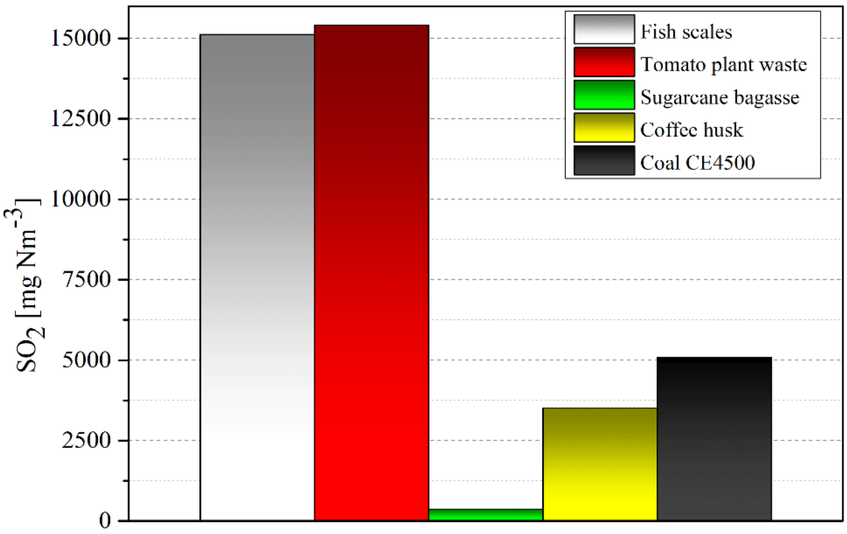

Figure 5. Sulfur dioxide (SO_2) emissions for the different solid fuels.

In real combustion processes, SO_2 emissions increase with the sulfur content of the fuels and decrease with the increase in the concentration of calcium (Ca), also, the amount of S of the fuel that is converted into sulfur dioxide is highly dependent of sample composition and oxygen concentration (Kazanc *et al*., 2011). According to Sartor *et al*. (2014) and Williams *et al*. (2012), higher SO_2 emissions in thermal systems are indicative of the complete or partially complete combustion, as this gas participates directly in the overall combustion reaction, being released entirely as volatile gases and/or in the combustion of biochar or biocoal.

One of the most efficient ways to reduce SO_x emissions in the flue gases is to use a mixture of limestone ($CaCO_3$) together with the fuel. However, as a consequence, there is a proportional increase in sulfur (S) in the ashes generated after the process burning (Werther *et al*., 2000; Roy *et al*., 2014).

From the results obtained in this study, it was possible to observe how the main elements present in the composition of the samples (*e.g.*, carbon, nitrogen, and sulfur) influence the emissions of the air pollutants analyzed, in addition to assessing how the fish scales behave concerning other solid fuels, mainly lignocellulosic biomasses.

4. CONCLUSIONS

This study proposed a sustainable and ecofriendly reuse of fish scales as a possible alternative energy source, using them in thermochemical conversion (combustion) processes for the bioenergy generation. The study was carried out under stoichiometric combustion conditions, which allowed evaluating the thermal behavior of this biomass in relation to different solid fuels, some already applied commercially. In addition to estimating the emissions of the main atmospheric pollutants (CO_2, NO, and SO_2), it was also possible to assess the influence of some elements (nitrogen and sulfur) present in the composition of the samples in the generation of these gases. The air mass required for complete combustion was also determined, which is of paramount importance for this purpose and is directly proportional to the carbon composition present in each analyzed feedstock. Therefore, the information presented in this preliminary study may contribute as a basis for further experimental analysis in real thermal systems or laboratory scale.

5. ACKNOWLEDGEMENTS

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