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GASIFICATION OF SMUGGLED AND SEIZED CIGARETTES FOR SYNTHESIS GAS AND ENERGY PRODUCTION

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Abstract. *The gasification of solid waste biomass to generate synthesis gas and energy is an environmentally sustainable alternative. Smuggled and seized cigarettes are part of the raw materials that can be used in this process. The characterization of refuse derived fuel (RDF) from cigarette based on elemental, proximate, thermogravimetric and calorimetric analysis contributed to the improvement of the gasification process and synthesis gas generation. The characterization of the cigarette biomass showed 9.6 wt% of moisture, 65.9 wt% of volatiles, 22.0 wt% of fixed carbon, 12.1 wt% of ash and a lower heating value (LHV) of 16.7 MJ kg*$^{-1}$*. In addition, CO,* CO_2*,* H_2*,* CH_4 *are the main elements generated in synthesis gases. The gasification temperature directly affects the synthesis gas formation inside the thermochemical reactor as well as its LHV. Low temperatures (500 °C) favor greater formation of CO (7.70% v/v) and low value of LHV (4.5 MJ kg*$^{-1}$*) and high temperatures (585 °C) provide greater formation of* CH_4 *(9.92% v/v) and* H_2 *(1.4% v/v) and higher LHV (6.8 MJ kg*$^{-1}$*), highlighting the importance of optimizing gasification parameters to improve synthesis gas quality for energy purposes. These results suggest that the gasification process of seized can be a sustainable alternative practice to convert waste to energy (WTE).*

Keywords: *Renewable energy, WTE, seized cigarettes, gasification, gas chromatography.*

1. INTRODUCTION

Although fossil fuels represent the largest source of energy, gasification of municipal solid waste (MSW) for energy generation can be considered a renewable technology, as it adopts a previously unused source of energy, with low carbon emissions and proper control of greenhouse gas emissions (Bioenergy, 2020; Bot et al., 2022).

Gasification allows the generation of synthesis gas, a renewable fuel, which can be used in thermal engines to generate thermal and electrical energy. In this process, synthesis gas or syngas is characterized by presenting carbon monoxide (CO), carbon dioxide (CO_2), hydrogen gas (H_2), methane (CH_4) and other light hydrocarbons with carbon atoms ranging from C_1-C_6 (Havilah et al., 2022; Tezer et al., 2022).

In gasification thermochemical reactors, the partial combustion of carbon-based raw material is treated at temperatures $\geq$ 800 °C in the presence of gasifying agents such as: air, vapors, CO_2 or a mixture of these components (Kumar et al. 2020). The complexity of the MSW composition in the gasification process can make the H_2/CO ratio impaired because the process can be accompanied by the generation of tar and other by-products. In addition, the N and S elements present in MSW are mostly transformed into NOx, SO_2 and tar contaminants (Alnouss et al., 2020; Zhang et al., 2018). The effectiveness of this process is extremely dependent on proper temperature control, as well as of the flow of gasification agent, biomass properties, particle size among other parameters (Tezer et al., 2022).

The yield and chemical composition of the synthesis gas depend on several factors such as: the physicochemical properties of the biomass raw material, humidity, particle size, process temperature, heating rate, type of reactor, among others (Filho et al., 2022; Souza and Alves, 2023).

The literature reports that the optimal gasification conditions are reached when the biomass has a moisture content < 20 % w/v, volatiles between 75 to 90 % w/w (50 to 80 % w/w for biomass from vegetable residues) and ash content between 0.1 to 15% w/w (Marcelino, 2017).

Although efficient energy systems and technologies are relevant for the future, it depends on the availability of biomass as a feedstock. Brazil has the potential to establish itself in biomass gasification technologies from different sources of solid waste for energy generation (Do Prado et al., 2022).

Smuggled and seized cigarettes are part of solid waste. According to the World Health Organization about 9 to 11% of cigarettes consumed in the world are smuggled and the illegal cigarette trade represents 11.6% of the cigarette market. In Brazil, the average number of cigarettes seized reached 276 million packs in 2018. Considering the volume seized, cigarettes can be considered the most harmful product to the Brazilian economy (Domingues et al., 2022; Goncalves et al., 2021). Once seized, cigarettes need to be destroyed and this procedure entails transport, storage and correct final disposal costs.

Currently, incineration has been the practice for destroying seized products, although a costly and environmentally unsustainable alternative due to the release of toxic gases into the atmosphere (Medeiros, 2017).

In this sense, environmentally correct alternatives for disposal smuggled and seized cigarettes are necessary. After apprehension, the seized cargo is crushed and the final product is called refuse-derived fuel (RDF), composed of tobacco, paper, filter, cardboard and plastic. Thus, the characterization of this product for energy generation becomes necessary and proximate, thermogravimetric, elemental and calorific analysis must be carried out. This characterization with the adequate operational adjustment of the gasifier will allow a suitable syngas for energy generation.

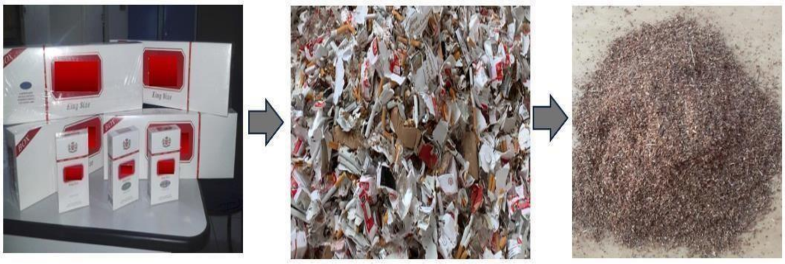
2. METHODOLOGIES

The biomass characterization of seized cigarettes followed the standards described in ABNT NBR 8633/1984, ABNT NBR 8112/1986 and ASTM D5373/2016. Initially, the samples (Figure 1a) are de-characterized with a shredder capable of transforming biomass into refuse derived fuel (RDF) as presented in Figure 1b. Then, for chemical analyzes, the RDF particles size must be standardized using a knife mill (Figure 1c).

The RDF elemental analysis was performed in the laboratory of the Institute of Petroleum and Natural Resources of Pontifical Catholic University of Rio Grande do Sul. It aims to quantify the mass yields of Carbon (C), Hydrogen (H), Nitrogen (N), Oxygen (O) and Sulfur (S) in RDF.

Both proximate and thermogravimetric analyzes were carried out in a thermogravimetric analyzer from Navas Instruments: TGA-2000A. The objective was to quantify moisture, volatile materials, ash and fixed carbon in RDF. In the thermogravimetric analysis, the volatilization or degradation of the biomass is monitored as a function of temperature, and it represents the mass loss of RDF as a function of the temperature.

The high heating value [kJ kg^{-1}] was measured with the calorimetric bomb IKA-C200. Figure 1 shows the preparation of biomass for experimental tests.



(a) (b) (c)

Figure 1. Photos of the seized cigarettes (a) packs; (b) shredding and production of refuse derived fuel (RDF) and (c) particles size standardized using a knife mill.

After characterizing the RDF, the experiments continued with the gasification of the biomass. Figure 2 shows the scheme of the gasification process and synthesis gas collection.

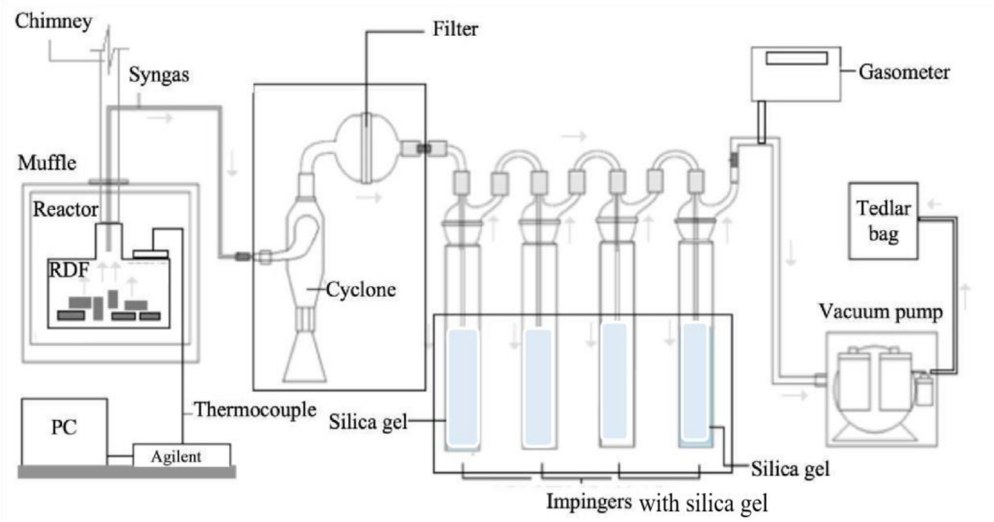


Figure 2. Scheme of the experimental gasification bench.

The RDF of seized cigarettes was introduced into the reactor positioned inside a muffle capable of reaching the temperature of 800 °C (monitored by a temperature controller). During the gasification process, the synthesis gas was collected with the aid of the collection system in different temperature ranges for analysis.

The collection system aims to prevent the passage of particulate matter that may accompany the smoke, as well as residues of pyroligneous liquid and tar that are separated and filtered using the cyclone and filter presented in Figure 2. The silica contained in the impingers reduces the moisture that accompanies the synthesis gas, avoiding the precipitation of condensate in the TedLar bags and inside the gas chromatography equipment.

A gas chromatograph (Shimadzu GC-2014) equipped with a thermal conductivity detector (TCD), flame ionization detector (FID) and a Carboxen 1010 PLOT column was used for the analyses. The data was processed using GCSolution software. Helium gas (He) was used as the mobile phase and the components present in the sample were determined as described by Silva et al. (2021).

As presented in Figure 3, the chromatographic analysis begins with the pressurization of the carrier gas reservoir (helium was used in this research). Then, the sample is placed in the injector where it is vaporized and comes into contact with the mobile phase. The sample is transported by the carrier gas through the interior of the column and heated by the oven where the separation of the sample components occurs. The sample components reach the detectors which determine the peaks of interest and generate signals when passing through substances other than the carrier gas. These signals are received by the signal amplifier and recorded in a chromatogram.

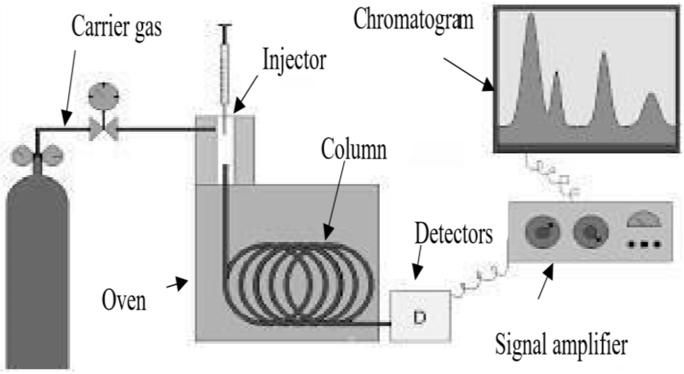


Figure 3. Block diagram of a typical gas chromatograph

At each injection 1000 µL of syngas were injected inside the chromatograph using a micro-syringe.

3. RESULTS

Table 1 presents the proximate analysis of seized cigarettes according to ABNT NBR 8112/1986 and comparisons with other types of local biomass collected and analyzed in TGA-2000A.

Table 1. Proximate analysis and comparisons with other types of biomasses.

Proximate analysis (wt%) wet base	Present work*	Wood chips RDF*	MSW RDF*
Humidity	9.60 ± 0.03	9.23 ± 0.04	7.86 ± 0.20
Volatile materials	65.85 ± 0.12	71.39 ± 0.75	69.76 ± 0.22
Ashes	12.11 ± 0.20	1.35 ± 0.29	4.57 ± 0.49
Fixed carbon	22.04 ± 0.23	27.25 ± 0.74	25.66 ± 0.58

(*) All biomasses were analyzed at the Federal University of Uberlandia using the TGA-2000A

Based on results presented in Table 1 it is possible to verify that the moisture content of cigarettes RDF is very close to that one measured in wood chips RDF. The main difference between these two types of biomasses is verified in the ash content. The heterogeneity of cigarettes RDF, such as tobacco, cardboard, filters, plastics, paper, make the ash content 10 times greater than that measured for the wood chips RDF. Another example of heterogeneous biomass is the MSW RDF. Depending on its composition, the moisture, volatile materials, ash and fixed carbon contents may approach or deviate from those measured for the cigarettes RDF.

Figure 4 shows the cigarette RDF thermogravimetric analysis.

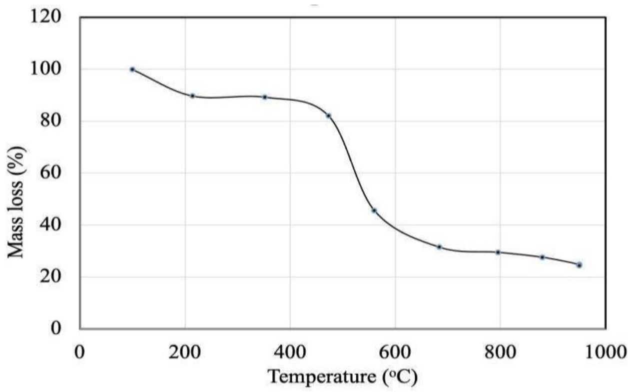


Figure 4. Cigarette RDF thermogravimetric analysis

From thermogravimetric analysis of cigarettes RDF, it is observed the conversion of more than 70% of the initial mass in volatile materials, due to thermodegradation of organic salts (cellulose acetate), inorganic salts (calcium carbonate and others present in coal residues and ash), tar decomposition, among others (Gómez -Siurana et al., 2012; Zhou et al., 2011).

It is reported in the literature that cigarette components exhibit volatilization at temperatures above to 400 °C. As an example, Li et al. (2021) used cigarette butts and observed that at 400 ºC there was volatility and the mass of the sample reduced by 80%. Zhao et al. (2013) and Zhou et al. (2011) observed that volatilization/degradation occurred over 400 ºC, characterized by a mass reduction of 75.4%.

The cigarettes RDF lower heating value determination was carried out in wet and dried bases as presented in Figure 5. In the case of the cigarettes RDF, something unusual was observed, that is, the wet samples had a higher LHV than dry samples.

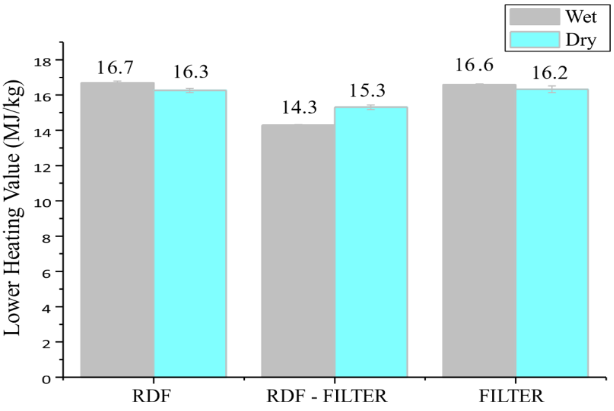


Figure 5. Cigarettes RDF lower heating value (LHV)

As shown in Figure 5, removing the cigarette filter from the RDF and repeating the calorimetric analysis, the wet RDF showed a lower LHV when compared to the dry one. A commonly expected result for all types of biomasses. After repeating the test using only the filter and carrying out a literature research (Silva, 2014), it was found that the filter has in its composition chemical components that contribute to increase the LHV, such as: butanal, 2-methylpentane, 2propanamine, hydroxylamine, heptane, trichloromethane, hexane, 3-methylpentane, ethylamine, 3-methylfuran and other volatiles of lower molar mass. As the filter dries, part of these components volatilizes, which reduces the LHV. Therefore, the filter generates the benefit of increasing the cigarettes RDF LHV and creates a unique condition, in which, wet cigarettes RDF has a higher LHV than the dry one.

Lungulease et al. (2017) analyzed the LHV of briquettes obtained from different parts of crushed tobacco cigarettes: a) cigarette filter and paper; b) tobacco and c) mixture (filter, paper and tobacco) in order to use them as lignocellulosic fuel in stoves and furnaces. After pre-treatment of the samples, the moisture content was around 10% w/w and an LHV and the ash content were determined: in case a) the filter and paper were 16.5 MJ kg^{-1} and 6% w/w, b) tobacco 11.5 MJ kg^{-1} and 19.2% w/w and c) from the mixture 16.5 MJ kg^{-1} and 15% w/w. Table 2 presents the calorimetric analysis of cigarettes RDF and comparisons with the literature.

Table 2. Calorimetric analysis of cigarettes RDF.

Dry base	Cigarette RDF (1)	RDF from agro-industrial activities (2)	Cigarette butts RDF (3)
LHV	16.3	17.0	16.7

(1)Present study; (2) Candine et al. (2016); (3) Marchi et al. (2014)

It is also noted in Table 2 that the results of LHV agree with others published in the literature; RDF from agroindustrial activities (17 MJ kg^{-1}) (Candine et al., 2016) or cigarette butts RDF (16.7 MJ kg^{-1}) (Marchi et al. 2014).

In addition, an elemental analysis was also carried out to measure the CHNS-O content in cigarettes RDF. Table 3 presents the elemental analysis of the samples in wet and dry bases.

Table 3. Elemental analysis of cigarettes RDF.

Elemental analysis (wt%)	Cigarette RDF wet base(1)	Cigarette RDF dry base (1)	Tabacco RDF (2)
Carbon	39.95	40.54	38.0
Hydrogen	5.39	5.46	5.5
Nitrogen	1.24	1.26	2.6
Oxygen	42.90	40.15	
Sulfur	0.13	0.13	

(1) Present study; (2) Senneca (2007)

Based on the results presented in Table 3, as expected the drying process does not generate significant variation in the elemental composition of the biomass under study. Furthermore, the percentages of carbon and hydrogen make this biomass a promising compound for gasification.

Li et al. (2021) analyzed cigarette butts for energy generation and the results found were: C: 48.80%, O_2: 42.68%, H: 6.64%, N: 1.80% and S: 0.25%.

In addition to the previous characterization of the cigarettes RDF, the synthesis gas generated from the gasification of 0.283 kg of cigarettes RDF was also characterized and its chemical composition was determined (Figure 6a and 6b).

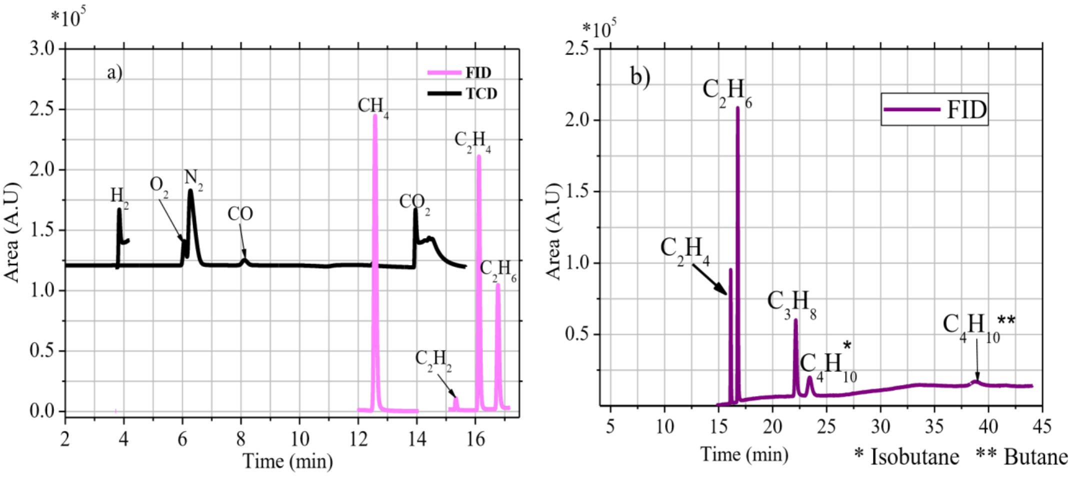


Figure 6. Synthesis gas chromatograms generated from cigarettes RDF: a) FID and TCD analysis. b) FID analysis.

In this analysis 1000 µL of synthesis gas were injected in CG-2014 and the volume of each analyte was calculated based on the ideal gas law formula PV = nRT (Silva et al., 2020). Table 4 shows the chemical composition of the samples collected at different temperatures (500 and 585 ºC) and comparisons with the literature.

In this work the temperatures of 500 and 585 ºC were obtained empirically from the combustion of syngas. A duct was connected to the chimney of the reactor (Figure 2) in order to test whether the syngas was flammable. When a stable flame was identified as presented in Figure 7, the syngas was collected in Tedlar bags and submitted to the chromatographic analysis.

Figure 7. Stable flame produced by syngas at 585 ºC.

As expected, Table 4 shows that at 500 °C more than 85% of syngas composition is non-combustible elements, such as: O_2, CO_2 and N_2. As the temperature increases, at 585 ºC this percentage drops to 80%. In addition, lower temperatures generate greater precipitation of condensable elements in the cyclone and filter shown in Figure 2.

Literature reports some studies about the influence of temperature on the composition of the synthesis gas obtained by gasification.

Table 4. Chemical composition of the synthesis gas at 500 and 585 °C and comparison with literature.

T (ºC)	Components (% v/v)							
	O_2	N_2	H_2	CO	CO_2	CH_4	ΣHydrocarbons (C_2-C_6)CxHy	LHV (MJ/kg)
(1) 500	13.3	74.9	0.0	5.1	2.3	0.14	4.26	4.5
(1) 585	12.9	63.7	1.4	7.7	0.3	9.9	4.1	6.8
(2) 800	1.8	61.5	5.7	9.8	12.7	5.2	3.3	4.5
(3) -	-	50.0	9.0	14.0	20.0	7.0	-	5.4
(4) 930	6.3	58	4.9	14.0	15.3	1.4	-	2.1
(5)1000	-	63.4	6.0	6.6	12.7	6.5	-	7.4

(1)Present work; (2)Ferreira et al. (2021); (3)Friedrich (2017); (4)Fandiño et al. (2022); (5)Santos et al. (2022).

Based on the results presented in Table 4, it was noted that the temperature strongly influences the composition of the synthesis gas, since at 500 ºC there was no generation of H_2 and a very low concentration of CH_4. CO, CH_4 and other light hydrocarbon contents increased significantly at 585 ºC. The increase in temperature promotes the cracking and decomposition of lignin and cellulose, which release volatiles with high levels of H_2, CO, CH_4 and light hydrocarbons (Zhang; Pang, (2019) and Haensel et al., (2009)). In parallel, the rates of exothermic and endothermic gasification reactions in the oxidation Eq. (1) and/or reduction zones (water-gas Eq. (2) and Boudouard reactions Eq. (3)) become more significant, favoring the production of H_2 and CO (Havilah et al., 2022). On the other hand, the oxidation zone reactions and the Boudouard reaction favor the consumption of O_2 Eq.. (1) and CO_2 Eq. (3), respectively. This favoring justifies the reduction in the formation of non-combustible gases (O_2, CO_2 and N_2). About N_2, studies show a reduction of this element in the syngas as the temperature rises (Havilah et al., 2022).

$$2C + O_2 \rightarrow 2CO \qquad \Delta H= -123 \text{ kJ/mol} \tag{1}$$

$$C + H_2O \rightarrow CO + H_2 \qquad \Delta H= 131.4 \text{ kJ/mol} \tag{2}$$

$$C + CO_2 \rightarrow 2CO \qquad \Delta H= 172.6 \text{ kJ/mol} \tag{3}$$

Comparing the results presented in Table 4 with those of Ferreira et al. (2021), who used a continuous feed gasifier of RDFs from municipal solid waste (MSW) at 800 ºC, it was found that the temperature, the type of gasifier (batch or continuous), the characteristics of each type of RDF (cigarettes and MSW) interfere on the characteristics of syngas, changing its lower heating value.

The LHV of the synthesis gas was calculated considering the chemical composition presented in Table 4, the mixture of ideal gases under normal conditions of temperature and pressure (CNTP) (25 ºC and 1 atm) and stoichiometric combustion considering 1 kmol of synthesis gas. Based on the number of moles, the enthalpy of combustion of each element and the mass of synthesis gas at 500 and 585 ºC, the LHVs were calculated (Table 4).

Analyzing the results in Table 4, it is noted that the LHV of the synthesis gas collected at 500 °C has a lower LHV value (4.5 MJ kg^{-1}) compared to that one collected at 585 °C (6.8 MJ kg^{-1}). Such results reveal that in the case of cigarettes RDF the increase in temperature contributes to increase the LHV.

Some studies about the composition and LHV of the synthesis gas generated by the gasification of solid waste are reported in the literature. As examples, in Table 4 were also presented the results obtained by Friedrich (2017) and Fandiño et al. (2022), which analyzed the syngas from the gasification in a fluidized bed of the agroindustrial biomass and Santos et al. (2022), who evaluated the synthesis gas from the gasification of heterogeneous waste.

From such information it is possible to verify that biomasses with a high carbon and hydrogen content have a higher calorific value. Therefore, when subjected to gasification at high temperature, they tend to generate a synthesis gas with a higher LHV.

4. CONCLUSIONS

From this study it was possible to verify that the characterization of biomass (for gasification) from the proximate, gravimetric, elemental and calorific analysis is fundamental. Biomass gasification for energy generation requires the optimization of operating parameters such as temperature, gasification agent and biomass humidity. In this work was observed that at low temperatures (500 °C), the synthesis gas has a higher content of undesirable elements, tar and liquid components, and a lower content of light hydrocarbons, which contributed to the low LHV.

In this preliminary study, the synthesis gas obtained at both 500 ºC and 585 ºC showed a satisfactory LHV for use in thermal engines. However, in order to comply with Brazilian standard CONAMA 316, the minimum gasification temperature required must be 800 ºC. Therefore, batch reactors, such as the one adopted in the present work, end up becoming unfeasible. To feed the reactor with RDF, it is necessary to wait until the equipment reaches room temperature. Furthermore, during heating syngas is produced, mainly at temperatures above 500 ºC, as presented in the present work. When the reactor reaches 800 ºC, practically all that remains inside is ash. In this sense, continuous feed reactors operating at 800 ºC become more interesting, as it becomes easier to control and standardize the production of syngas for generation of thermal and electrical energy purposes.

Finally, the use of smuggled and seized cigarettes in the gasification process demonstrated feasibility, however, the next step is to carry out tests in a semi-industrial scale pilot plant that operates in accordance with CONAMA 316 requirements.

5. ACKNOWLEDGEMENTS

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