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Recovering Wood Waste to Produce Briquettes Enriched with Commercial Kraft Lignin

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Abstract

Aiming to use lignocellulosic biomass as energy source, one of the process that may aggregate values is the densification process, which allows the production of bioenergy using solid fuels, mainly for reducing transportation costs. In this research, solid fuel from co-briquetting of wood residues from sawmill using commercial kraft lignin as binder was investigated. The effects of compression pressure (900, 1200 and 1500 PSI) and briquette formulation (varying wood and kraft lignin proportion) on the quality and characteristics of briquettes were evaluated. The main findings were that briquetting of wood residues with kraft lignin resulted in an improvement of bulk density, strength rupture modulus, low heating value (LHV) and high heating value (HHV). The briquettes using 4% and 6% of kraft lignin, and submitted to 1200 to 1500 PSI, presented higher bulk density and strength resistance, respectively. On the other hand, the heating values showed the highest results with the addition of 2% lignin at 900 PSI, being the legal range for additives in briquettes for many countries such as in European Union.

Keywords

Hardwood Biomass Densification, Heating Values, Solid Fuel

1. Introduction

Lignocellulosic biomass is considered a renewable and alternative material as a potential energy source for the society [1] [2] [3] [4] [5]. The renewable supplies rank fourth in the world total energy demand after oil, coal, and natural gas [6]

[7], being biomass the most significant type of renewable supply and due to its advantages of being widely sourced, sustainable, and environmentally friendly, it has attracted more and more interest of academic and industrial sectors [8]. Considering the concerns on the rational utilization of the lignocellulosic biomass by our society, wood processed in the sawmills, which is a common activity in many countries, is another neglected raw material, since it is available in a large volume. In these activities, the wood is poorly used, with around 40% - 60% of waste being generated from the log wood processing [9], which in general is burned in the sawmills to generate energy or discarded into landfills [10].

A challenge for using waste lignocellulosic biomass is that it must be transported from where they are generated, often in milling sites and open fields, to the storage facilities, which may affect the energy balance and costs [11] [12] [13] [14] [15]. One effective strategy to address these concerns is through mechanical densification of the biomass. Mechanical densification is the compaction of biomass to definite sizes, which improves the bulk densities and minimizes the irregularities in shape of biomass, facilitating the transportation [16]. In lignocellulosic biomass processing for the production of solid fuel, pelletizers and briquette press are the most commonly used equipment for mechanical densification. These solid fuels are frequently used as substitute fuels in boilers to produce steam that may be used for electricity production, for example.

Kaliyan & Morey [16] and Ajiboye *et al.* [17] support that as a result of the shape and size, among other variables, can determine the compressibility and the products of the densification. Briquettes with higher density values, produced from agriculture and forest residue, may substitute or compliment solid fuels such as charcoal, firewood and coal. The briquettes are high energy density materials, require lower transportation and storage costs, present uniform quality such as constant humidity content, and high mass fluency [18] [19].

Several studies have experimentally investigated the characteristics of alternative biomass briquettes, as wood residues, under different conditions of pressure. The results show that the sawdust, most abundant waste or residue in wood-based industries [20], may generate briquette with better characteristics than other materials [21].

Aiming to improve the mechanical and physico-chemical properties of the solid fuel, many organic and inorganic binders may be used for the densification process [22] [23] such as starch, protein, fiber, fat/oil, and other additives. The addition of binders in the densified solid fuel production using lignocellulosic biomass as raw materials might have a positive outcome on the strength, in a similar way to the resins used in the production of wood boards [24] [25].

Considering the available materials from the based forest industry that could be used for bindering purpose, the kraft lignin appears as a promissory material, since it is a hydrophobic compound and it also presents a higher heating value when compared to the whole lignocellulosic biomasses [26] [27] [28] [29] [30]. The worldwide lignin production is approximately 50 million tons [31] and this by-product have distinct adhesive characteristics and may be used as a densification binder [29]. The use of lignin as binder is relatively new and studies of this application as a binder have been done recently [28] [29] [32]-[38]. The resulting briquettes would cost less to transport, easier to handle and storage [39].

Regarding the available lignocellulosic biomasses for being used as raw materials, the residual material of the industrial processes seems to be an alternative for improving the rational utilization of the natural resources. Concerning the specie of wood, *Astronium concinnum* (Engl.) Schott, a hardwood species, belongs to the family of Anacardiaceae, commonly known as gonçalo-alves, aroeira-rajada, guarubu-violeta and mucuri, which is a raw material widely used commercially due to its wood quality and availability [40]. Its wood is more used in exteriors, buildings, floors and furniture [41] [42] [43] and its basic density value is around 0.64 g/cm^3 [44].

The main goal of this research was to evaluate the application of common woody industrial waste from sawmills (sawdust of *Astronium Concinnum* (Engl.) Schott) enriched with eucalypt kraft lignin as an additive, aiming to produce briquettes.

2. Materials and Methods

2.1. Raw Materials

In this work residues from the wood sawmill processing *Astronium concinnum* (Engl.) Schott were used. The wood samples evaluated were 37 years old. Three trees samples were collected from an experimental station in the Vale Natural Reserve, which is inside the Atlantic Forest biome, located in Sooretama, Espírito Santo State, Brazil. The residues were obtained from a machining process commonly used on a sawmill. The residues (sawdust) were collected and air dried to a moisture content of about 15% and stored in plastic bags. The commercial kraft lignin used as an additive in the briquetting process was obtained from a Brazilian kraft pulp mill, Suzano Pulp and Paper, which uses *Eucalyptus* spp. as feedstock.

2.2. Methods

The wood biomass, commercial kraft lignin and briquettes were characterized according to the methods described in **Table 1**.

The briquetting process was conducted using a laboratory briquette machine with a piston press (LB-32, Lippel, Brazil). The briquetting conditions were determined experimentally through preliminary tests of pressure application and the time required for pressing and cooling. The temperature chosen (120˚C) was determined as a function of the lignin plasticization, which is a compound responsible for the bond among the wood particles during the application of pressure [59]. Three compression pressure conditions were used: 6.20 MPa (900 PSI); 8.27 MPa (1200 PSI); and 10.30 MPa (1500 PSI), with pressing and cooling times of 6 minutes.

Table 1. Methods used to characterize wood biomass, kraft lignin and briquettes.

Analysis	Standard
Sawdust fractionation	TAPPI T257 cm-02 [45]
Extractives	TAPPI T264 cm-97 [46]
Ashes	TAPPI T15 os-58 [47]; T211 [48]
Carbohydrates	WALLIS *et al.* [49]; SCAN-CM 71:09 [50]
Soluble lignin	TAPPI UM 250 [51]
Insoluble lignin	TAPPI T222 om-97 [52]
Acetyl groups	SOLAR *et al.* [53]
Uronic acids	SCOTT [54]
Elemental analysis (CHNSO)	DIN EN 15104 [55] which was measured by using a TruSpec Micro-Leco Instruments 628 Series C/H/N elemental analyzer with oxygen and sulfur module
Low and High Heating Values	DIN EN 14918 [56]
Briquettes bulk density	VITAL [57]
Rupture Modulus of Briquettes	NBR ISO 11093-9 [58]

To determine the briquetting conditions, preliminary tests relating to compaction time, cooling time and temperature were carried out. The conditions chosen were those where briquettes were obtained without cracks and with less deformation. The working moisture was 8%, obtained by using a laboratory greenhouse, which was within the ideal range proposed by Kaliyan and Morey [16]. The use of moisture higher than 8% would cause the briquettes to rupture. When the raw material moisture content is very dry or above the indicated value, it can impair the packaging of the material or produce an unstable briquette, which may disintegrate when stored or transported [60], resulting in lower durability and therefore becomes more susceptible to damage [61].

The proportion of Kraft lignin mixed with the wood fines was 0%, 2%, 4%, and 6%, and the mass of each briquette was fixed to 20 g, amounting 12 treatments, with 6 repetitions of each one. When an excess of the kraft lignin is used, it loses its efficiency as a binding agent and it negatively affects the briquette density [38]. Preliminary tests were performed and no more than 6% of lignin was used to avoid problems occurring with the equipment, relating to the lignin plasticization, and considering that just 2% of additives may be used in briquettes in many countries, for instance in European Union, the values above this range were used just for evaluating the potential performance delivered by the additive. In order to evaluate the quality of the briquettes and the briquetting process, visual analyzes were first carried out to observe the presence of cracks and deformations on their sides, as well as analyzes of the variations of the dimensions (height and diameter) and loss of mass in the briquetting. These visual observations were performed after the cooling time during measurements of height and diameter of briquettes.

Unfortunately, there was not found literature data which report on the raw material investigated in this study. Therefore, several hardwood species were used as a reference for comparing the results from this work. This also indicates that this work collaborates for disseminating some unpublished scientific information on the raw material used.

Aiming to analyze results obtained in this work, the Shapiro & Wilk test was used to verify the normality of the briquettes data [62]. The data were also submitted to analyses of variance using the Cochran test [63] to evaluate differences among treatments. The Tukey's t-test was applied at a 95% significance level, when significant differences among the results were found.

3. Results

The results on the wood residues and eucalypt kraft lignin composition are described in Table 2, taking into account the chemical composition importance of the materials for energy conversion. As also previously described, there was not found literature data which report on the raw material investigated in this study, being the findings on the wood composition an important piece of information for many other works.

Briquetting process

Table 3 shows results relating to the briquetting processes on the bulk density, modulus of rupture (MOR) and heating values (LHV and HHV) of the studied

Table 2. Chemical characterization of the evaluated biomass and kraft lignin.

	Analysis	*Astronium concinnum* biomass	Kraft lignin
Elemental analysis, %	C	48.40	56.00
	N	0.17	0.14
	H	6.30	4.70
	O	44.50	20.70
	S	0.03	4.20
Soluble extractives, %		6.2	-
Soluble lignin, %		3.8	4.5
Insoluble lignin, %		19.8	80.9
Total lignin, %		23.6	85.4
Carbohydrates, %	Glucan	47.0	0.1
	Xylan	11.1	0.1
	Mannan	1.7	0.4
	Arabinan	0.2	0.1
	Galactan	0.7	0.1
Ash, %		0.7	14.2
High heating value, MJ/kg		19.0	21.7
Low heating value, MJ/kg		17.7	20.8

Table 3. Mean values of briquettes bulk density (ρ_{ap}), modulus of rupture (MOR) and heating values (LHV and HHV) per treatment.

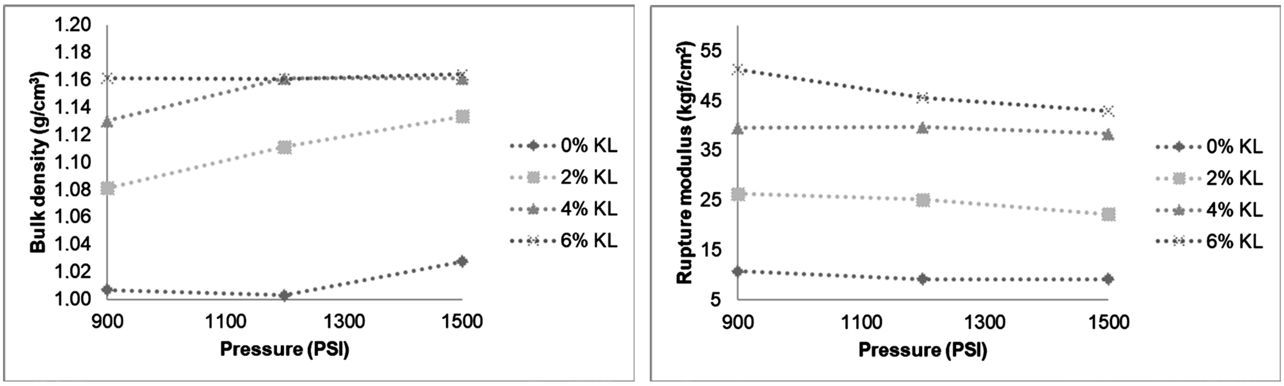
Treatments	$\bar{\rho}_{ap}$ (g/cm^3)	MOR (kgf/cm^2)	LHV (MJ/kg)	HHV (MJ/kg)
T1	1.01 ± 0.01^{f}	10.7 ± 13.6^{cd}	16.3 ± 0.00^{e}	18.2 ± 0.00^{e}
T2	1.00 ± 0.04^{fg}	9.2 ± 14^{cd}	16.3 ± 0.00^{e}	18.2 ± 0.00^{e}
T3	1.03 ± 0.03^{e}	9.1 ± 12.7^{cd}	16.2 ± 0.00^{f}	18.1 ± 0.00^{f}
A1	1.08 ± 0.01^{d}	26.3 ± 10.3^{c}	16.9 ± 0.00^{a}	18.8 ± 0.00^{a}
A2	1.11 ± 0.01^{c}	25.1 ± 10.3^{c}	16.6 ± 0.01^{cd}	18.4 ± 0.05^{d}
A3	1.13 ± 0.01^{b}	22.2 ± 9.9^{cd}	16.7 ± 0.00^{c}	18.5 ± 0.05^{bc}
B1	1.13 ± 0.03^{ab}	30.6 ± 7.9^{c}	16.8 ± 0.00^{b}	18.7 ± 0.00^{b}
B2	1.16 ± 0.02^{a}	39.7 ± 5.7^{b}	16.7 ± 0.00^{c}	18.6 ± 0.00^{cd}
B3	1.16 ± 0.01^{a}	37.3 ± 5.8^{b}	16.7 ± 0.00^{c}	18.6 ± 0.00^{cd}
C1	1.16 ± 0.01^{a}	51.4 ± 4.6^{a}	16.9 ± 0.01^{ab}	18.7 ± 0.05^{ab}
C2	1.16 ± 0.02^{a}	45.6 ± 3.5^{ab}	16.3 ± 0.01^{ef}	18.5 ± 0.05^{c}
C3	1.16 ± 0.01^{a}	43.0 ± 1.4^{b}	16.3 ± 0.00^{e}	18.1 ± 0.05^{ef}

Note: (T1) pressure 900 PSI; (T2) pressure 1200 PSI; (T3) pressure 1500 PSI; (A1) 2% Kraft lignin sample with 900 PSI; (A2) 2% Kraft lignin sample with 1200 PSI; (A3) 2% Kraft lignin sample with 1500 PSI; (B1) 4% Kraft lignin sample with 900 PSI; (B2) 4% Kraft lignin sample with 1200 PSI; (B3) 4% Kraft lignin sample with 1500 PSI; (C1) 6% Kraft lignin sample with 900 PSI; (C2) 6% Kraft lignin sample with 1200 PSI; (C3) 6% Kraft lignin sample with 1500 PSI. Equal letters in the same column indicate equality between the values of the averages at a significance level of 95%.

briquettes. These results are considered important parameters for the quality of the briquettes.

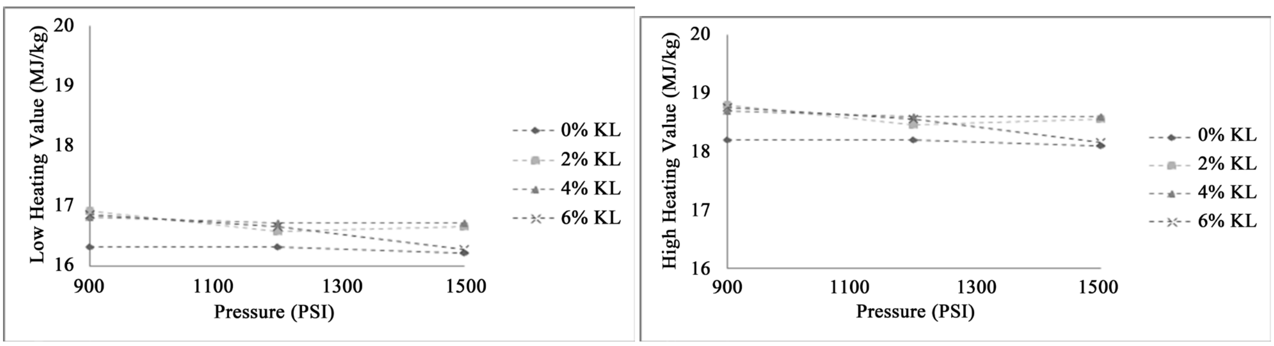
The modulus of rupture result was determined using software coupled with the universal test equipment called "Contenco-Pavitest". The analysis procedure was in accordance with the Brazilian standard NBR ISO 11093-9 [58] with adaptations. The equipment applies a perpendicular force on the upper side of the briquette through a piston until it ruptures. The force was determined by a preliminary test speed (3.5 mm·min^{-1}). The briquette was tested in the vertical position, and the forces were applied parallelly, according to methodology adapted from the ABNT NBR ISO 11093-9 standard [58].

As expected, the bulk density increased, due to the agglutination and plasticization of lignin. The produced briquettes with 4% and 6%, respectively treatments B and C, of incorporated Kraft lignin did not show difference related to the bulk density. However, these treatments showed difference in treatments T (0% KL) and A (2% KL). In general, the use of additives in briquettes are limited to two percent, but this study showed that the inclusion of 4% and 6% of KL contributed to the increase in the bulk density, also 4% and 6% (at 900 PSI) presented significant increase in the rupture modulus of the briquettes (**Figure 1**). On the other hand, the decrease of the rupture modulus in treatments C2 and C3 was observed, and it can be explained by cohesion strength among the particles with higher pressure due to the chemical composition of *wood* biomass



(a) (b)

Figure 1. Properties of the briquettes, being: (a) bulk density; (b) modulus of rupture.



(a) (b)

Figure 2. *Astronium concinnum* briquettes heating values using different concentrations of kraft lignin (KL) as additive, being (a) low heating value; (b) high heating value.

and the kraft lignin used, which possess lower content of lignin and a significant percentage of ashes on its composition, which may affect the binder process.

Analyzing the values shown in Table 3 and Figure 2 is possible to affirm that the briquettes produced in 900 PSI showed higher heating values. The lowest heating value (LHV) was obtained when using 0% KL at higher pressure values (1500 PSI). The briquettes produced with 2% KL presented the highest HHV, followed by the 4% KL and 6% KL, respectively.

4. Discussion

The elemental analysis is a factor that depends on comparison age due to anatomical and structural changes that happen inside the biomass. It was possible to observe that wood biomass evaluated (*Astronium concinnum)* in this research presented similar percentage of carbon (46.0% - 49.95%), hydrogen (4.8% - 6.2%), nitrogen (0.1% - 2.4%), oxygen (43.1% - 46.7%), and sulfur (0.01% - 0.05%) when compared to the other commercial woods such as *Eucalyptus* spp. [64]-[71]. In this study the elemental analysis was completely measured, as previously explained, being the sum of CHNSO almost 100%. In general, the oxygen is calculated by difference, which always generates values of 100% for the sum of these elements [64]-[71].

For the biomass, the observed extractives value was higher than that reported in the literature for *Eucalyptus* spp. According to research by Gomide *et al.* [72], Gomes *et al.* [73], Boschetti *et al.* [28] [29] hardwood species (*i.e. Eucalyptus* clones) present extractives up to 5%. Extractives are correlated to the volatile compounds, which collaborate with the heating value [74].

Regarding the total lignin content, the evaluated wood presents 23.6%, a lower value when compared to eucalypt (26.7% - 31.7%) [29] [67] [69] [72]. The carbohydrates content (60.7%) is also lower when compared to eucalypt carbohydrates content (64.5% - 70.2%) analyzed by Gomide *et al.* [72]. The lignin content is correlated with the fixed carbon collaborating to the biomass heating value [74]-[79], being high values of this parameter desired for energy conversion.

The ash content, another important parameter to heating values, was similar to eucalypt wood (0.1% - 0.4%) according to Pereira *et al.* [35], Borges *et al.* [67], Morgan *et al.* [68], Veiga *et al.* [69] and Silveira [70]. The ash content is not desirable for energy application collaborating for decreasing heating value [74] [79] [80]. The ash content is also undesirable for the operation of boilers, where the biomass can be burned for converting it into energy [15] [81] [82], since ash is responsible for generating incrustations, corrosion, unscheduled stops for maintenance, for example.

The high heating and low heating values of the evaluated wood biomass were quite similar to those values described in the literature [28] [83] [84] for lignocellulosic biomasses.

Regarding to the kraft lignin composition, it is possible to verify that the studied lignin in this research has similar percentage of carbon (49.8% - 61.8%), hydrogen (5.0% - 6.5%), nitrogen (0.1% - 1.3%), but lower oxygen content (29.2% - 37.9%) and significantly higher content of sulfur (0.8% - 2.5%) [29] [85] [86] [87]. This information and the higher ash content directly influence in the lignin purity and energy generation. A higher ash content is disadvantageous because it decreases the heat transfer in the fuel and the biomass heating value [88] [89] [90] [91] [92], as well as increasing the corrosion of the equipment used in the process [93].

The purity is also related to the acid insoluble (Klason lignin) and soluble lignin presented in the binder material. Compared to Zhou and Lu [86] and Boschetti *et al.* [29] the studied kraft lignin used in this research is more impure, presenting lower contents of total lignin on its composition. However, the carbohydrates content presented is lower than other studies [85] [86] [87].

Briquetting process

This study pointed that when using more than 2% of additives, for instance, the use of 6% of kraft lignin showed the highest and the same values of bulk density not depending on the pressure used in the briquetting process. The studies of Boschetti *et al.* [28] [29] also showed that the incorporation of 6% of Kraft lignin has the best results for bulk density. This is important, because high-quality fuels should present high density and strength, with higher energy content [94] to burn for a longer time [8].

The rupture modulus increased after a rise in pressure and a larger amount of lignin was incorporated (6% KL). When the lowest pressure value was used (900 PSI), it was observed the optimum point of the briquetting, regarding the durability and maximum rupture.

Boschetti *et al.* [29] studying *Hymenolobium petraeum* Ducke, *Eucalyptus* spp., and *Pinus* spp. briquettes also found similar results, proving that 6% of impregnated Kraft lignin gives better bulk density and rupture modulus properties. Pereira *et al.* [35] studying pellets confirms that the use of at least 2% of Kraft lignin can change properties of hardwood compressed materials.

The heating values indicate that the best cohesion strength of the sawdust and lignin occurred in the lower value of pressure aiming to deliver heating value. It is important to observe that the heating value is directly related to the fixed carbon content and is also associated with volatile and ash content [74]. Besides the studied lignin presents a high carbon content, which is desirable, it is also presented a high ash content, which reduces the heating value of the briquettes, mainly when the proportion of lignin addition in the briquette increases. This result indicates that the commercial kraft lignin needs to be designed for being applied to the energy application, since in the literature it is possible to observe the commercial eucalypt kraft lignin with 0.01% - 1.4% of ash [95], 13% less than the commercial kraft lignin used in this study, which may collaborate for improving also the briquette heating value beside other strength properties.

5. Conclusion

In order to produce briquettes with higher heating values, it was concluded that evaluated biomass added with lower concentrations of Kraft lignin at 900 PSI generate the best results. The addition of 2% of lignin showed the best performance, however using 4% and 6% of Kraft lignin combined with higher pressure values better briquettes properties can be obtained. The use of the higher concentrations may enter in disaccordance with most standards for using additives in briquettes.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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