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PELLETS PRODUCED WITH HARVEST RESIDUE OF PINUS: NEEDLES, BRANCHES, AND TREETOPS

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Resumo

Pellets produzidos com resíduos da colheita de pinus: acículas, galhos e ponteiras. A produção e o consumo de pellets de madeira têm aumentado nos últimos anos, para uso em aquecimento doméstico, geração de energia termoelétrica e aquecimento de aviários, entre outros. Os resíduos da colheita da madeira de Pinus podem ser fonte de matéria-prima para a produção desses pellets, no entanto, é importante avaliar o efeito de cada compartimento da árvore (ponteira do tronco, galho e acículas) nas propriedades dos pellets. Neste contexto, foram produzidos pellets empregando os componentes individualmente, três misturas binárias e uma mistura ternária, compondo sete tratamentos experimentais. Um delineamento experimental centroide simplex foi adotado para avaliar as propriedades físicas, químicas e energéticas. Foi realizada modelagem estatística e ajustados modelos de regressão linear, quadrática e cúbica para prever as propriedades dos pellets em função das proporções dos componentes nas misturas. Pellets com biomassa de galhos e ponteiras e a mistura desses dois componentes apresentaram as menores densidades unitária e aparente, e a maior quantidade de teor de finos. Os pellets de acículas apresentaram maior durabilidade mecânica e maiores valores de poder calorífico superior e líquido, além de maior teor de cinzas. Pellets de acículas e de misturas dos três componentes apresentaram o melhor potencial para produção comercial e uso final industrial. A mistura dos três componentes é a alternativa mais viável porque os resíduos podem ser aproveitados sem qualquer separação.
Palavras-chave: colheita de madeira; *Pinus taeda*; modelagem estatística; agropellets

Abstract

The production and consumption of wood pellets have increased in recent years for home heating, thermoelectric power generation and aviary heating, among other end uses. Residues from harvesting of *Pinus* wood can be a source of raw material for this pellet production, however, it is important to evaluate the effect of each tree compartment (treetops, branch and needles) on pellet properties. In this context, pellets were produced using each component individually, three binary mixtures, and one ternary mixture, composing seven experimental treatments. A simplex centroid experimental design was adopted to assess the physical, chemical and energy properties. Statistical modeling was performed, and linear, quadratic, and cubic regression models were fitted to predict the properties of the pellets as a function of the proportions of components in the mixtures. Pellets made from branches and treetops and the mixture of these two components presented the lowest unit and bulk densities, and the highest quantity of fine particle content. Pellets from needles had the highest mechanical durability and the highest net and higher calorific values, and also the highest ash content. Pellets produced with needles and ternary mixtures presented the best potential for commercial production and industrial end uses. The mixture of all three components is the most feasible alternative as the residues can be used without any separation.
Keywords: wood harvesting; *Pinus taeda*; statistical modeling; agripellets

INTRODUCTION

Wood pellets have been used worldwide as an alternative biofuel to replace conventional fossil fuels (THRÄN *et al.* 2019), with the advantage of being produced from industrial wood residues residue and agricultural by-products. Wood pellets, for instance, are classified as a carbon-neutral and renewable energy source. According to the same author, pellet consumption in Europe is increasing, mainly to meet the demand of the residential market and thermoelectric sector.

In this latter sector, several coal-powered plants have been converted to operate exclusively with pellets to mitigate greenhouse gas emissions. In 2022, wood pellet production in Europe reached 25.6 million metric tons, accounting for 55% of global production, followed by the Americas with 31% (WBA, 2023). Europe represents 77% of global demand but produces only 50% of the region's consumption, with the difference being largely supplied from the USA and Canada (SMITH *et al.*, 2019). South America has increased its production by 385%

in the last decade, with Brazil and Chile being the contributors to this growth (BIOENERGY EUROPE, 2018). At Brazil 30 active industries were identified distributed across six states, producing near 820 thousand tons in 2020, using pine, eucalyptus, black acacia, sugarcane bagasse, peanut shells and coffee husks (GARCIA *et al.*, 2022).

The advantages of pellets as a solid biofuel include their low cost (since their production can be based on wood residues) and low sulfur, chlorine and ash contents. In addition to these virtues, depending on the chemical composition of the biomass, wood pellets can meet all gaseous emission limits established by environmental regulations (GARCIA *et al.*, 2017).

Planted forests of the *Pinus* genus occupy 1.64 million hectares in Brazil, producing wood for many sectors, such as pulp and paper, furniture, wood panels and chemical industriy, among others. One of the most important planted species is *P. taeda* due to its fast growth in the southern and southeastern regions of Brazil. In the country as a whole, the biomass of 10-year-old *P. taeda* trees can reach 32.88 tons ha^{-1} for branches, and 24.08 tons ha^{-1} for needles, according Brand *et al.* (BRAND *et al.*, 2014). Along with treetops, these two components constitute the residues usually discarded from planted forest harvesting. This kind of biomass represents an alternative for direct burning in industrial furnaces, because of its low cost and large availability.

However, the high intrinsic volume, high moisture content and low energy density limit the use of these harvest residues to local uses, since their transportation is not economically viable. Ash content is also an important limiting factor, since high ash content can limit the use of pellets to industrial applications only (GARCIA at al., 2022). The alternatives most applicable to harness this type of material are chipping, densification and biochar production despite the higher content of dirt and bark compared to the wood from energy plantations. Even so, the harvest residues from planted forests can be appropriately used as a secondary raw material for power generation or the production of biofuels for industry (AMORIM *et al.*, 2021).

A previous study of the production of pellets from harvest residues was conducted by Pinto *et al.* (2015), who used eucalyptus treetops as raw material. The authors reported bulk densities higher than 600 kg m^{-3}, moisture content below 10%, and an average net calorific value of 4,034 kcal kg^{-1}. However, there is a lack of research studies related to use of harvest residue of *Pinus* trees for pellets production. The proportion of each tree component (needles, branches and treetops) can influence the final quality of the pellets, especially because the chemical and elemental composition of these components varies.

Therefore, if the energy properties vary from one component to another, the pellets will exhibit distinct properties depending on their composition. Mixing components is an alternative for utilizing harvest residues while producing pellets with acceptable quality for industrial use. As woody resources are increasingly scarce and expensive, the possibility of harnessing the whole tree to produce pellets gains importance to add value to the forests where usually only is the main interest.

The present study aimed to produce pellets with harvest residues (needles, branches and treetops) from *Pinus taeda*, and to evaluate the influence of each component on the physical-mechanical properties, proximate chemical composition, and calorific value on the final product.

MATERIAL AND METHODS

Tree harvesting and collection of residue biomass

Five *Pinus taeda* trees were harvested from a 12-year-old forest stand (3.0 m x 2.0 m spacing) located in an experimental area designated to pulp and paper production at coordinates 25° 28' 3" S and 50° 39' 4" W, Paraná, Brazil. The trees were felled using a chainsaw. For the collection of the residue biomass, harvesting method routinely used by a pulp and paper company from Paraná State was adopted. After harvesting, the trees were debranched and the portion of the trunk with diameter smaller than 8 cm was considered treetop. Treetops along with branches and needles were collected for further processing.

The collected material was classified into three categories and placed separately on a plastic tarp before further processing. The three fractions were processed in a knife-mill and the particles were oven-dried at 65 °C until reaching an equilibrium moisture of 8%. After drying, the particles were processed using a Wiley mill. The particle size of each material used for pellet machine processing was defined in preliminary tests. Different particle sizes ranging from 3.00 to 1.00 mm were tested, and it was found that the ground material from treetops and branches with granulometry between 1.70 mm and 1.00 mm yielded good results. For the needles acceptable particle sizes ranged from 3.00 mm to 1.00 mm. After sieving, the moisture content and bulk density of the particles were determined.

Production of pellets

The moisture content of the biomass particles was adjusted to 12 + 2% (dry basis). The pellets were produced using an Amandus Kahl model 14-175 pelletizing machine equipped with a 6-mm-diameter flat matrix, power of 3.5 kW, and production capacity of 30 kg h^{-1}. The pelletizing temperature ranged from 80 to 90 °C for branches and treetops, while for needles the temperature range was 50 to 60 °C. These temperatures were established in preliminary tests when the best conditions of pelletizing for the different tree fractions were determined. The roller speed of the pelletizing machine was set to 1,200 rpm, and the material was passed through the machine once.

In this pelletizing machine, pressure is not directly controlled; instead, the height of the passage of the compressor roller over the matrix is controlled, which acts with pressure of up to 1.3 MPa to force the passage of the material through the holes in the matrix. The pressure on the material as it passes through the die holes varies between 70 and 180 MPa (SADEQ *et al.*, 2024).

Assessment of pellet quality

To determine the hygroscopic equilibrium moisture the pellet samples were prepared in accordance with ISO 14780-17 and placed in an environmental chamber at a temperature of 20 °C and 65% relative humidity. The unit density of the pellets was measured from a representative sample of 30 pellets per experimental treatment, obtaining the length (mm) and the diameter (mm) using a digital caliper. The weight of each pellet was determined with an analytical balance (precision of 0.0001 g). The bulk density (D_b) was determined in accordance with ISO 17828-15, using a 500 mL container (glass beaker), adapted to the amount of sample available in the laboratory. The mass of the material required to fill the beaker was measured and the density was obtained by dividing the volume of the recipient by the mass.

Proximate chemical analysis was performed in triplicate for the pellets from each experimental treatment following the procedures recommended by the standard ASTM D-1762-84 to determine the percentages of the volatile matter, fixed carbon and ash content. The higher and net calorific value (kcal kg^{-1}) was determined in two replicates using an Ika Werke 5001 adiabatic calorimeter following the procedures described in ASTM D240-17.

The mechanical durability test was performed to assess pellet strength using a rotary tumbler (300 mm length x 200 mm diameter) at a speed of 50 rpm for 10 min with 500 g of pellets and three replicates per experimental treatment, following the procedures described in ASTM D3402/D3402M-16. The standard recommends a 3.15 mm sieve to separate the fines from the tested sample, but in the present work we use a 3.35 mm sieve. The percentage of fines generated in the assay was calculated as the ratio of sieved fines and the initial mass of pellets. The quality (physical-mechanical properties) of the pellets from each treatment was classified according to the quality classes defined in ISO 17225-2-15 for material for commercial/residential and industrial use.

Experimental design and statistical analysis

Based on a completely randomized design called simplex centroid (HILLIG *et al.*, 2003), a statistical model based on mixtures (weight to weight percentage) of the three fractions was adopted to identify the best combination and to analyze how the different components interacted with each other. The effects of the interactions among components were evaluated using a triangular diagram in which the properties of pellets obtained from a given mixture were related to the proportions of components, which in turn were dependent on each other.

Table 1 d presents the details of the experimental design developed to produce pellets with harvest residues from *Pinus taeda*, encompassing seven treatments: three types of pellets produced with the pure components, three types with different proportions of two components (50% to 50%), and one type with a mixture of the three components (33.3% of each one).

Table 1: Experimental design (simplex centroid) employed for the pellets production with harvest residues from *Pinus taeda*.
Tabela 1: Delineamento experimental (centróide simplex) empregado para produção de pellets com resíduos da colheita de *Pinus taeda*.

Experimental Treatment	Type of Model	Proportions of Mixture (%)		
		Needles	Branches	Treetops
1	Linear	100	0	0
2		0	100	0
3		0	0	100
4	Quadratic	50	50	0
5		50	0	50
6		0	50	50
7	Cubic	33.3	33.3	33.3

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As shown in Table 1, linear models were fitted to describe the effect of each individual tree component (treetops, branches, and needles) on the properties of the pellets, quadratic models considering the effect of binary mixtures, and cubic models to describe the effect of the three components together were fitted and assessed. The statistical analysis was carried out using the SPSS software and the means were compared by the Tukey test at a 95% significance level. As commented previously, linear, quadratic and cubic regression models were fitted by correlating the proportions of components in the mixtures and the properties of the pellets. The best models were selected based on the following decision criteria: correlation coefficient between experimental and estimated values; significance of coefficients of the equations; realism of the model; and root-mean-square error (RMSE).

RESULTS

Properties of the harvest residues of *Pinus taeda* and the pellets

Table 2 presents the mean values of the moisture content and bulk density of the harvest residues used in different proportions of the three components corresponding to the experimental treatments. It also shows the final dimensions (diameter and length) and physical-mechanical properties of the produced pellets with them.

Table 2: Moisture content and bulk density of the chips (biomass) and the means of physical-mechanical properties of the pellets produced with harvest residues of *Pinus taeda*.
Tabela 2: Teor de umidade e densidade a granel dos cavacos (biomassa) e médias das propriedades físico-mecânicas dos pellets produzidos com resíduos da colheita de *Pinus taeda*.

	Biomass		Pellets						
ET	MC (%)	BD (kg m^{-3})	D (mm)	L (mm)	MC (%)	UD (kg m^{-3})	BD (kg m^{-3})	MD (%)	FPC (%)
1	13.09 a (9.00)	128.00 e (6.55)	6.00 a (0.00)	14.98 a (7.61)	7.91 b (10.32)	1,156.93 a (13.95)	649.11 a (2.08)	99.70 a (0.08)	0.30 c (24.85)
2	12.88 a (6.36)	174.06 d (2.15)	6.07 a (1.73)	13.19 b (13.51)	7.36 b (14,39)	980.63 bc (10.88)	556.81 d (1.75)	84.15 c (0,02)	15.85 a (0,12)
3	14.89 a (6.09)	272.73 a (2.90)	6.10 a (4.44)	15.06 a (8.96)	9.27 a (3.18)	1,004.73 bc (12.32)	569.92 d (2.95)	85.43 c (0.53)	14.57 a (3.13)
4	13.24 a (11.15)	168.40 d (2.20)	6.10 a (1.90)	14.88 a (13.47)	7.69 b (6.37)	1,115.18 ab (16.69)	645.92 a (3.23)	99.11 a (0.02)	0.89 c (2.79)
5	13.28 a (8.02)	201.13 c (6.32)	6.00 a (0.00)	15.44 a (8.12)	8.08 b (11.44)	1,037.26 ab (20.28)	610.44 b (2.49)	99.60 a (0.05)	0.40 c (18.43)
6	12.63 a (8.19)	224.60 b (2.43)	6.15 a (3.17)	13.58 b (13.29)	7.41 b (16.04)	946.97 c (18.66)	529.85 e (2.35)	83.50 c (3.40)	16.50 a (17,25)
7	14.73 a (19.33)	219.60 b (0.51)	6.00 a (0.00)	13.26 b (12.16)	7.97 b (18.13)	1,028.20 ab (18.91)	590.15 c (3.20)	89.15 b (2.30)	10.85 b (18.91)
OM	13.53 (9.74)	198.36 (3.30)	6.06 (0.97)	14.34 (11.02)	7.96 (11.41)	1,038.55 (15.95)	593.17 (2.58)	91.52 (0.91)	8.08 (12.11)

ET = Experimental treatment; OM = Overall mean; MC = moisture content; BD = bulk density; D = Diameter; L = Length; UD = unit density; MD = mechanical durability; FPC = fine particle content. Mean followed by same letters in the rows are equal by the Tukey test at 95% significance. Values in parentheses beneath the means are the coefficients of variation (%).

Table 3 presents the mean values obtained for the proximate chemical analysis (volatile matter, fixed carbon and ash contents) and the higher and net calorific value (at equilibrium moisture content) of the pellets.

Table 3: Mean values of proximate chemical analysis and higher and net calorific values (at equilibrium moisture content) of the pellets produced with harvest residues of *Pinus taeda*.
Tabela 3: Médias da análise química imediata e do poder calorífico superior e líquido (ao teor de umidade de equilíbrio) dos pellets produzidos com resíduos da colheita de *Pinus taeda*.

Experimental Treatment	Proportions of Mixture (%)			VM (%)	FC (%)	A (%)	GCV (kcal kg^{-1})	NCV (MJ kg^{-1})
	Needles	Branches	Treetops					
1	100	0	0	72.87 f (0.41)	23.98 a (1.07)	3.15 a (1.39)	4,882 a (2.1)	17.50 a (2.31)
2	0	100	0	77,81 b (0.11)	21.48 c (0.47)	0.71 e (2.94)	4,670 cd (1.48)	16.79 b (1.60)
3	0	0	100	78.85 a (0.73)	20.70 d (2.61)	0.45 f (16.61)	4,530 d (0.83)	15.87 c (0.90)

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Experimental Treatment	Proportions of Mixture (%)			VM (%)	FC (%)	A (%)	GCV (kcal kg-1)	NCV (MJ kg-1)
	Needles	Branches	Treetops					
4	50	50	0	74.31 e (0.54)	23.65 a (1.51)	2.04 b (2.69)	4,815 ab (0.12)	17.29 ab (0.13)
5	50	0	50	75.38 d (0.54)	22.84 b (1.71)	1.78 c (1.61)	4,705 bc (0.72)	16.78 b (0.78)
6	0	50	50	79.32 a (0.04)	20.24 d (0.04)	0.44 f (5.62)	4,672 c (0.03)	16.79 b (0.03)
7	33,33	33,33	33,33	76.42 c (0.39)	22.33 b (1.08)	1.24 d (4.78)	4,721 bc (0.34)	16.87 b (0.37)
Overall Mean				76.41 (0.40)	22.18 (1.21)	1.40 (5.09)	4,714 (0.81)	16.84 (0.87)

VM = volatile matter; FC = fixed carbon; A = ash; GCV = higher calorific value; NCV = net calorific value at equilibrium moisture content. Means followed by same letters in the rows are equal by the Tukey test at 95% significance. Values in parentheses beneath the means are the coefficients of variation (%).

Figure 1 illustrates the pellet properties in triangular/ternary diagrams. Each vertex of the triangles refers to a given harvest residue (needles, branches, and treetops). The edges show the variation between the means obtained for the pellets produced with the pure residues. The interactions between each type of residue are displayed by curved lines and by color graduation from green to red, passing through yellow. In the red regions, the higher values are observed for the property, and in green regions, the lower values are shown, respectively. The ternary diagrams are very useful since they enable clearly visualizing where a particular property is favored or disfavored by varying (increasing or decreasing) the proportion of each the three residues and interaction between them. Figure 2 shows the pellets produced.

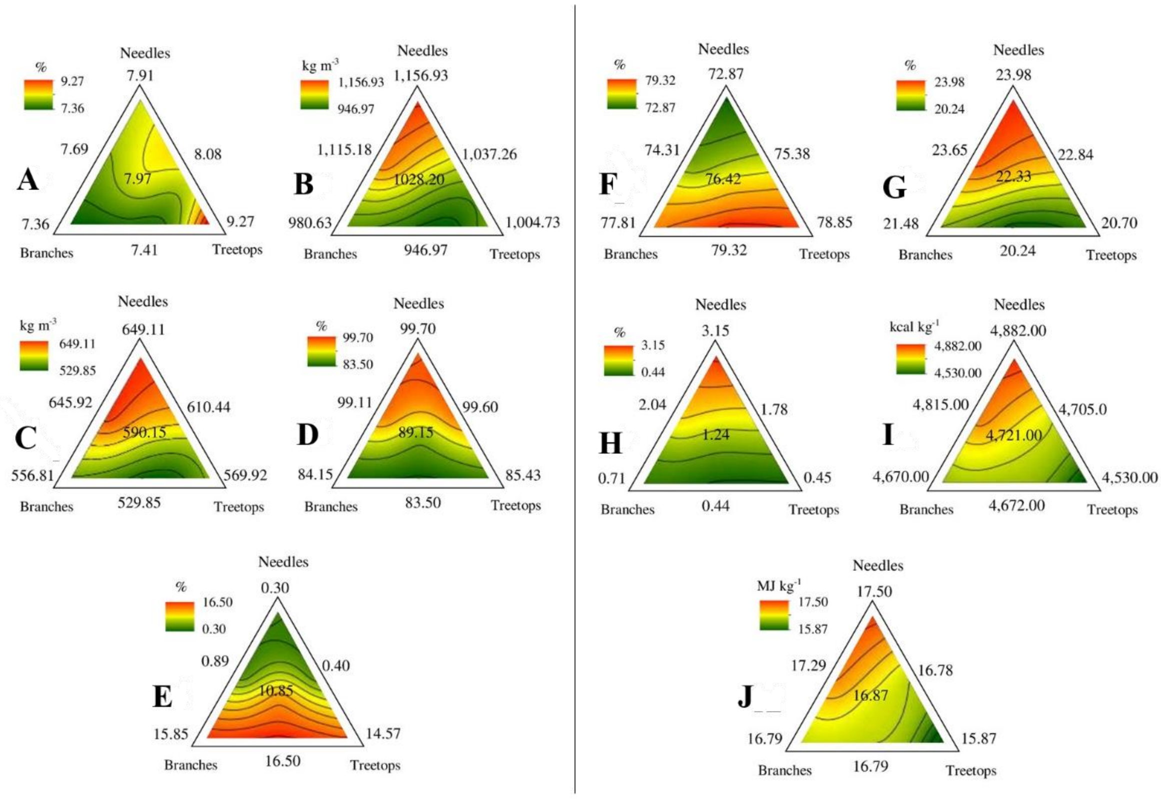

Figure 1: Ternary diagram of the physical-chemical properties of the pellets produced with harvest from *P. taeda* harvest residues. (A) moisture content; (B) unit density; (C) bulk density; (D) mechanical durability; (E) fine particle content; (F) volatile matter; (G) fixed carbon; (H) ash; (I) higher calorific value; (J) net calorific value.

Figura 1: Diagrama ternário das propriedades físico-químicas dos pellets produzidos com resíduos da colheita de *P. taeda*. (A) teor de umidade; (B) densidade unitária; (C) densidade a granel; (D) durabilidade mecânica; (E) teor de finos; (F) matéria volátil; (G) carbono fixo; (H) cinzas; (I) poder calorífico superior; (J) poder calorífico líquido.

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Statistical modeling based on mixtures of harvest residues of *Pinus taeda*

Table 4 presents the regression models fitted to predict the physical and mechanical properties, proximate chemical composition, and higher and net calorific values (at equilibrium moisture content) of the pellets produced from *Pinus taeda* harvest residues. For all quality parameters analyzed, when the coefficients were not significant by the T-test, the model was discarded. Therefore, only the equations that significantly represent the quality parameters are listed in Table 4.

Figure 2: Pellets produced with harvest residues of *Pinus taeda*, needles (A), treetops (B), branches (C), mixture of needles-treetops (D), and mixture of needles-branches (E).

Figura 2: Pellets produzidos com resíduos da colheita de *Pinus taeda*: acículas (A), ponteiras (B), galhos (C), mistura de acículas-ponteiras (D) e mistura de acículas-galhos (E).

Table 4: Statistical models fitted to predict the physical-mechanical properties, proximate chemical composition, and net and higher calorific values of the pellets as a function of the proportions of harvest residues of *Pinus taeda*.

Tabela 4: Modelos estatísticos ajustados para prever as propriedades físico-mecânicas, química imediata e poder calorífico líquido e superior dos pellets em função das proporções de resíduos da colheita de *Pinus taeda*.

Statistical Models	Root-mean-square Error			R^2
	Linear	Quad.	Cubic	
Physical-mechanical Properties				
MC = 0.079*N+0.074*B+0.062*P+2.200e-005*AG-2.174e-005*AGP	0.002	>0.001	>0.001	0.286
UD = 11.569*A+9.806*G+10.047*P+0.19*AG	0.309	0.015	0.001	0.145
BD = 6.491*N+5.568*B+5.699*T+0.017*NB-0.013*BT-1.114e-005*NBT	0.042	0.002	>0.001	0.874
MD = 0.997*N+0.842*B+0.854*T+0.003*NB+0.003*NT-0.001*BT	0.009	>0.001	>0.001	0.982
FPC = 0.003*N+0.158*B+0.146*T-0.003*NB-0.003*NT+0.001*BP	0.009	>0.001	>0.001	0.982
Chemical Composition and Calorific Values				
VM = 0.729*N+0.778*B+0.789*T+2.594e-005*NBT	0.002	>0.001	>0.001	0.983
CF = 0.240*N+0.215*B+0.207*T+6.415e-006*NBT	0.002	>0.001	>0.001	0.962
A = 0.032*N+0.007*B+0.004*T+4.227e-005*NB-6.341e-006*NT-5.096e-005*BT-4.534e-006*NBT	>0.001	>0.001	>0.001	0.998
GCV = 48.820*N+46.700*B+45.305*T	0.365	0.018	0.001	0.891
NCV = 0.175*N+0.170*B+0.160*T	0.002	>0.001	>0.001	0.829

DISCUSSION

Properties of the harvest residues of *Pinus taeda* and the pellets

As shown in Table 2, the average moisture content of the different harvest residues was equal to 13.53%, which aligns with expectations, as the conditioning applied to them aimed to reach a range of 12 to 14% for this parameter. For the production of pellet stability, the moisture content is recommended to be in the range from 5 to 15%, but pelletizing pressure increased with increasing wood moisture (PRADHAN *et al.*, 2018). When this recommendation is not followed, 50 to 60% of the water is vaporized upon contact with the hot pelletizing matrix. So, if the moisture content is higher than 14%, a "shell" is formed over the matrix preventing entry of particles in the pelletizing channels. On the other hand, if the moisture content of the particles is lower than 11%, there is not enough interaction among them and only dust and brittle pellets are formed.

Regarding bulk density, needles were the component with the lowest value (128.00 kg m^{-3}) and the treetops had the highest value (272.73 kg m^{-3}). The mixture of the three types of harvest residues presented bulk density of 219.60 kg m^{-3}. Usually for sawdust and other wood residues, the value for this parameter ranges from 190 to 265 kg m^{-3} (HILLIG *et al.*, 2009), a value close to those found in this work. This property is one of the most limiting for the use of biomass as an energy source because the lower it is, the more costly storage and transportation will be (TUMULURU, 2023). According to the same authors, the presence of fine particles improves mechanical durability, as long as the pressure and temperature rearrange them by molecular diffusion to increase the contact surface. In general, the pellets of all components (Table 2) presented average diameter and length of 6.00 and 14.34 mm, respectively, similar to the parameters established in ISO 17225-2. This property influences the feeding the burners and, in the case of solid biofuels with standardized dimensions, facilitates the automation of feeding systems.

The average moisture content of the pellets was below 10%, and none of the treatments exceeded this value. According to ISO 17225-2, 10% is the maximum permissible value for pellets. When moisture content ranges from 6 to 9% during the pelletizing process, the final product is less likely to be subjected to the action of insects and decaying fungi, thus being considered a material with adequate sanitary conditions for storage and transport (HOLM-NIELSEN and EHIMEN, 2016). Moreover, lower moisture content is associated with higher calorific value, as energy losses due to water evaporation are reduced (ORO *et al.*, 2018).

In general, the pellets produced in the present experiment had unit densities higher than 900 kg m^{-3}, demonstrating that the densification process was effective. Pellets made from pure needles or mixtures containing this component (Table 2) presented the highest unit density. Most likely, the needles provided the best agglutination of the particles, which contributed to the results. The best pellets agglutination obtained with needles was attributed to their high proportion of lignin (43.37%) and extractives (30.01%) in relation to wood (MUÑIZ *et al.*, 2014) as these components favor the pelletization process.

Thus, the addition of needles to pellets produced with wood may serve as an additional improvement for the pelletizing process by increasing the unit density of the final product. Also, the bulk density of the pellets also increased when needles were combined with treetops and branches.

On the other hand, a significant decrease in this property was observed when the components were mixed. Therefore, a decrease in the mechanical durability of the pellets was reduced for this type of mixture. When energy density (energy amount by volume) is considered, it is desirable to have the highest bulk density possible for a given type of solid biofuel, because as mentioned previously, a reduction of transportation costs is favored by higher density (ALBASHABSHEH AND STAMM, 2021). As shown in Table 2, the highest unit densities corresponded to the highest mechanical durability and lowest fine particle content. Pellets from pure branches and treetops and their mixture generally presented the lowest mechanical durability. The bark present in these pellets may have contributed to this low result since, in the pelletizing process, lignin acts as a binding agent and the bark has a lower content of this macromolecule in its composition. Fernández *et. al.* (2023) also found low mechanical durability of pellets produced with *Pinus pinea* bark.

According to the ISO 17225-2 standard, wood pellets for industrial use should have a minimum mechanical durability of 96.5% and fine particle content lower than or equal to 6%. In this study, only the pellets produced with needles, needle-branch mixture, and needle-treetop mixture (treatments 1, 4, and 6) met these requirements. Both parameters are related to the ability of particles to aggregate in the final product. When those properties are in the recommended range, during handling the pellets remain intact or generate a low percentage of fine particles, thereby reducing risk of instantaneous ignition or explosion (ABELHA AND CIEPLIK, 2021).

Since there was little variation in the moisture content of the pellets produced in this experiment, other intrinsic characteristics of the harvest residues may have contributed to the differences in the mechanical performance. Variation in production parameters, such as matrix temperature, pressure, geometry, and those related also moisture content, particle size, and chemical composition should be considered in further research works because they can decisively influence the pelletizing process (TUMULURU, 2023). As commented above, pellets produced in treatments 1, 4, and 6 presented the best mechanical performance, but if the pelletizing conditions are adjusted, other types of mixtures (possibly including additives) may also yield final products in compliance with the standard requirements.

The overall mean volatile matter (VM) content of the pellets was equal to 76.41% (Table 3). Pellets produced from treetops and treetops-branches had the highest value of VM. Volatile matter refers to the fraction of the organic biofuel that is readily volatilized and released to burn in combustion conditions. Therefore, higher VM means easier and faster combustion (OBERNBERGER AND THEK, 2010). The volatilization of these organic fractions releases flammable gases (hydrogen and light hydrocarbons), favoring the combustion reaction. Pellets made only from needles presented the lowest VM and the highest fixed carbon content.

The average fixed carbon values found in this experiment ranged from 20.84 to 23.98%, which are higher than those reported by Spanhol *et al.* (2015). A decrease in VM was observed when needles were mixed with branches and treetops, with a concomitant increase in the fixed carbon content. On the other hand, these mixtures brought about an increase in the ash content of the pellets. When branches and treetops were mixed, the ash content decreased in comparison with the experimental treatments in which they were pelletized alone. Solid biofuels with high contents of fixed carbon tend to burn slowly. This is advantageous for pellets because of the more stable combustion and firing temperature. Ash, in turn, represents the mineral fraction of wood and other solid biofuels.

The ash content found in this study was higher than the value of 1% usually reported for wood pellets. Pellets produced from needles and branches presented the highest ash content. Ash is undesirable when in large quantity, because it is inert and does not act as a fuel (HENNE *et al.*, 2020). Another drawback is the formation of residues that adhere to and corrode the internal walls of burners and fireplaces, resulting in increased cleaning and maintenance costs (HUPA *et al.*, 2017). Pellets with ash content greater than > 0.7%, produced from branches, needles and their mixtures, did not meet the requirements for commercial and residential use class A1, but they can be used swimming pool heaters, air heating machines for animal husbandry (poultry and swine) and generation of thermoelectricity, among other uses that fall within classes B (residential and commercial use) and I3 (industrial use) of the standard ISO 17225-2. The pellets from needles did not fall in any use category, since they presented ash content higher than 3%. However, when mixture with branches and treetops, this parameter decreased.

There was a statistically significant difference in both the higher and net calorific values among the pellets produced. The highest values for these parameters were observed in pellets produced made exclusively from and in the mixture of needles and treetops. Concerning the net calorific value, except for the pellets produced from treetops, all were in compliance with the requirements of ISO 17225-2. For pellets produced with different percentages of bark of two *Pinus* Mediterranean species, Lerma-Arce *et al.* (2017) determined a value of 17.80 MJ kg^{-3} for the average net calorific value. Despite having the lowest calorific value, the pellets made from treetops had the lowest ash content. The mixture of the three components produced pellets with acceptable volatile matter, fixed carbon, and ash content, and also higher calorific value, demonstrating the potential of this material for commercial and industrial uses as biofuel.

As shown in the ternary diagram in Figure 1A, the highest moisture content (9.27%) was found in pellets produced from treetops, while the other experimental treatments resulted in materials closer to the overall mean. The highest unit density values were obtained for needles and mixtures of branches and treetops (Figure 1B). The same pattern was observed for the bulk density (Figure 1C), where pellets with values above 600 kg m^{-3} are considered suitable for industrial use.

Regarding the mechanical durability, the highest values were observed near to the top vertex of the diagram (Figure 1D). For fine particle content, the greyscale was inverted (Figure 1E), since the dense and resistant pellets generate lower values of this parameter, demonstrating a perfect relationship between these two properties.

Figures 1F and 1G, respectively show the contents of volatile matter and fixed carbon. In these diagrams, the pellets with the highest values for fixed carbon were produced with mixtures containing needles. As previously noted, for the ash content (Figure 1H), the pellets produced with branches and treetops presented the lowest value, and when needles were added to the mixtures, that property tended to increase. The diagrams enable visualization of the properties of the pellets from different treatments in terms of the quality required for a particular end uses. Figure 3 presents the appearance and color of the pellets produced in each experimental treatment; however, pellets from treatments 6 and 7 are not shown, as their integrity was insufficient for proper evaluation.

Statistical modeling based on mixtures of harvest residues of *Pinus taeda*

Based on the R^2 values of the fitted models (Table 4), variability of the samples could be predicted with good reliability. Unit density and moisture exhibited the lowest variation among the evaluated properties, which explains the lower R^2 values values for these models. For the other physical-mechanical properties and energy content, the models also presented good adjustment. As a general trend, the pure components had a stronger influence on the final properties of the pellets than the mixtures of harvest residues.

Needles were the component with the most significant influence on the physical and mechanical properties, except for the fine particle content. In the models, a negative interaction of needles-treetops and branches-treetops was indicated by the decrease in the unit density of pellets produced with these mixtures. Needles exerted a greater influence than the other components on unit density, and also bulk density and mechanical durability. Conversely, they were associated with the lowest fine particle content among all treatments.

The parameters used to estimate VM in the binary mixtures, generally did not show positive interaction. However, when all three components were combined, a significant interaction was observed, resulting in an increase in VM. A similar pattern was determined for fixed carbon. The binary mixtures of needles and treetops, branches-treetops, along with the triple mixture of residues, presented negative coefficients for ash, indicating that the addition of treetops was the responsible for the decrease in this characteristic. For both higher and net calorific values, the needles brought the highest positive contribution, followed by branches.

CONCLUSIONS

The results obtained in the present study provided the following conclusions:

- The feasibility of producing pellets from *Pinus taeda* harvest residues was demonstrated. However, further studies are necessary to classify these pellets according to the ISO 17225-2 standard for industrial use, as the current standard permits only this use.
- The inclusion of needles increased both unit and bulk densities, mechanical durability, and both higher and net calorific values of the pellets. In contrast, the lowest unit and bulk density were observed in pellets produced from branches and treetops, which also exhibited the highest fine particle content.
- Pellets produced from branches, treetops, and their mixture showed the lowest ash content (below 1%), whereas those made from pure needles or mixtures containing needles exhibited the highest ash content.
- Pellets made from pure needles and from the mixture of branches, treetops and needles showed potential for commercial and industrial applications, with the minimum requirements established by the standards.
- The statistical models fitted using the component proportions and pellet properties proved effective in predicting the final quality of the pellets.
- The modeling demonstrated that, in mixtures, the individual characteristics of needles predominantly influenced the physical-mechanical properties, and the proximate chemical composition of the pellets, except for the volatile matter content.

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