

ORIGINAL ARTICLE

Physical properties and formaldehyde emission in particleboards of *Eucalyptus* sp. and ligno-cellulosic agro-industrial waste

Propriedades físicas e emissão de formaldeído em painéis aglomerados de Eucalyptus sp. e resíduos lignocelulósicos agroindustriais

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Abstract

The aim of the paper was to evaluate particleboards made from eucalypt wood in combination with waste from agricultural activity (carpels of macadamia nuts, coffee husks, and papaya stalks), using urea-formaldehyde and tannin-urea-formaldehyde adhesives. The apparent density, water absorption (after 2 and 24 hours of immersion), thickness of swelling (after 2 and 24 hours of immersion) and emission of formaldehyde were analyzed. The majority of particleboards was classified as of medium density; the increase in the percentage of wastes on composition and the addition of tannic extract in urea-formaldehyde promoted the reduction of the physical properties; the particleboards attended performance specifications for water absorption and thickness swelling. The increase of tannic extract in the urea-formaldehyde adhesive reduced the free formaldehyde emission by 22.5% in the particleboards. The results indicate a potential of the wastes to be used as raw material to produce particleboards where these technological properties are required.

Keywords: Carpel of macadamia nut; Coffee husk; Papaya stalk; Adhesive.

Resumo

O presente trabalho teve como objetivo caracterizar painéis aglomerados confeccionados com madeira de eucalipto em combinação com resíduos agrícolas (carpelo de noz macadâmia, pergaminho de café e caule de mamoeiro), usando como aglutinantes os adesivos ureia-formaldeído e tanino-ureia-formaldeído. Analisou-se as propriedades físicas densidade aparente, absorção de água e inchamento em espessura, ambos após 2 e 24 horas de imersão; e a emissão de formaldeído. Os painéis aglomerados em sua maioria foram classificados como de média densidade; o aumento do percentual de resíduos na composição dos painéis aglomerados e a adição do tanino ao adesivo ureia-formaldeído promoveram a redução das propriedades físicas; os aglomerados atenderam às especificações de desempenho para absorção de água e inchamento em espessura. A adição de extrato tânico ao adesivo ureia-formaldeído reduziu 22,5% a emissão de formaldeído livre nos painéis aglomerados. Os resultados obtidos indicam o potencial de utilização dos resíduos lignocelulósicos como matéria-prima para fabricação de painéis aglomerados.

Palavras-chave: Carpelo de noz macadâmia; Pergaminho de café; Caule de mamoeiro; Adesivo.

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INTRODUCTION

The main source of raw material for the production of particleboards in Brazil comes from the wood of planted forests, mainly pine and eucalypts (Indústria Brasileira de Árvores, 2015). A study conducted in 2009 by the Ministry of the Environment already pointed out that the use of timber and industrial waste, in the southern and southeastern regions of Brazil, had a potential for power generation as well as in the wood reconstituted particleboard industry (Wiecheteck, 2009).

The importance of the Brazilian agricultural production for the country's economy during the last decade was remarkable (Organização de Cooperação e de Desenvolvimento Econômico, 2015). However, with the agricultural expansion, an increase of waste generation happened. Large amounts of wastes could accumulate in an uncontrolled way that can cause environmental problems (Instituto de Pesquisa Econômica Aplicada, 2012) when burned or even attracting insects and rodents. One way to reuse them is by using them as raw material for energy production (Penoni et al., 2011; Paula et al., 2011; Preilipper et al., 2016) and particleboards (Mendes et al., 2010; Minini et al., 2017; Tinti et al., 2018). Particleboards could be manufactured from different types of wastes and origins, such as from sawmills (Alves et al., 2014), soybean cultivation (Martins et al., 2018), corn straw (Silva et al., 2015) and macadamia nutshells (Ferreira et al., 2014; Wechsler et al., 2013).

Among the crops with high agricultural production, are macadamia, coffee, and papaya. To produce a ton of macadamia nuts, 70% of wastes from the peel and the carpel are generated (30% return rate). Those are usually used in gardens, also for the production of ornamental seedlings and as raw material for energy production (Penoni et al., 2011).

The yield of the coffee process depends on the variety. In the variety "acaia", the amount of peel and husk represents 60% of the weight; in the variety "mundo novo", 53.4% in the "catuai" 48.5%, all of these belong to the *Coffea arabica* species (Sfredo, 2006).

Papaya production represents 10% of the whole production of tropical fruits, about eight million tons, with 39% coming from Latin America and the Caribbean. The main world producers are Brazil, Mexico, Nigeria, India, and Indonesia, while the biggest exporters are Mexico and Malaysia (Food and Agriculture Organization of United Nations, 2012). There are still no studies about the use of papaya stalks in the production of particleboards. This waste is usually incorporated into the soil.

With this background, this study aims to characterize the physical properties, apparent density, water absorption, swelling thickness and the emission of formaldehyde in particleboards produced with the eucalypt sawdust combined with different agro-industrial waste percentages of the macadamia nut carpels, coffee husks, and papaya stalks.

MATERIAL AND METHODS

Raw material

The experiment consisted of using eucalypt wood and ligno-cellulosic agro-industrial wastes. The wood sawdust was obtained from logs of seven years old *Eucalyptus urophylla* × *Eucalyptus grandis*, from a plantation located in Alegre, Southern Espírito Santo State.

The agro-industrial wastes (Figure 1) were collected from papaya farm production (*Carica papaya*) (papaya stalks) and macadamia (*Macadamia integrifolia*) (carpels of macadamia nuts) located near the city of São Mateus, northern Espírito Santo state; and the coffee husks (endocarp), from an Arabica coffee (*Coffea arabica*) production at Dores do Rio Preto, southern Espírito Santo state.



Figure 1. Unprocessed agro-industrial ligno-cellulosic wastes: A – papaya stalks, B - carpels of macadamia nuts, C - coffee husks.

Eucalypt wood sawdust was obtained from log processing plants. All raw materials were air dried until achieving the equilibrium humidity content; and were then reduced to particles in a hammer mill and classified by a knitted sieve of 4.0mm, to obtain the most appropriate particles to produce the particleboard, using only the ones that were retained at a knitted sieve of 2.0mm. Then they were oven-dried at a temperature of 80 +/- 2 °C until the 5% pre-set humidity content and stored in hermetically sealed bags.

Particleboard production

To produce particleboards commercial pure urea-formaldehyde adhesive was used (density: 1.28g cm⁻³; pH 8.1; viscosity: 600 Cp; solids content: 64.8%; gel time: 64s) with the mixture (10%) of commercial tannic extract from *Acacia mearnsii* at 50% (density: 1.40 g cm⁻³, pH: 5.0, viscosity: 2068.50 Cp, solids content: 78.0%, gel time: 91s). The lignocellulosic particles were weighed to quantify the material needed and the adhesive to be applied, according to each treatment.

To determine the used particle weight and determine the particleboard density in each proportion, aiming at a compaction rate of 1.3, the bulk density of each material: papaya stalks (0.19 g cm⁻³), macadamia nut carpels (0.29 g cm⁻³) and coffee husks (0.35 g cm⁻³) were used. For eucalypt wood the basic density (0.56 g cm⁻³) (Equation 1-4) (Martins, 2016) was used.

$$D_p(100\%waste) = 1,3 \times D_{euc} \quad (1)$$

$$D_p(10\%waste) = 1,3 \times \left[\left(\frac{D_{euc} \times 90}{100} \right) + \left(\frac{D_{was} \times 10}{100} \right) \right] \quad (2)$$

$$D_p(20\%waste) = 1,3 \times \left[\left(\frac{D_{euc} \times 80}{100} \right) + \left(\frac{D_{was} \times 20}{100} \right) \right] \quad (3)$$

$$D_p(30\%waste) = 1,3 \times \left[\left(\frac{D_{euc} \times 70}{100} \right) + \left(\frac{D_{was} \times 30}{100} \right) \right] \quad (4)$$

Where: D_p = particleboard density (g cm⁻³) in each composition; D_{euc} = basic density of eucalypt wood (g cm⁻³); D_{was} = Bulk density of the lignocellulosic waste (g cm⁻³)

The particleboard was prepared in a 42.5 cm × 42.5 cm bottomless wooden forming box with an inserted lid. This form was supported on an aluminum plate and the previously glued particles were distributed manually and uniformly in its interior.

After the arrangement of the particles, the particleboard was compacted by applying a force on the forming cap, thereby performing a pre-pressing. This action allowed to reduce the air spaces, providing an improvement in the accommodation of the content on the aluminum blade. The aluminum sheets with the pre-pressed content were spaced with metal bars (10 mm).

The particleboard was pressed in a hydraulic press of horizontal flat plates with electric heating, thus forming the particleboards. Three particleboards were produced per treatment, with a pressing time of 10 minutes; pressing temperature of 160°C and pressure of 40 kgf cm⁻².

The particleboard was conditioned in a acclimatized room, with a temperature of 20 ± 3°C and relative humidity of 65 ± 5%, until equilibrium (72h minimum) (Associação Brasileira de Normas Técnicas, 2006a). Thereafter it was cut and again stored under controlled climatic conditions. Then the particleboards were cut and made into samples to obtain the apparent density (21 repetitions), water absorption (WA) and thickness swelling (TS) - both after 2 and 24 hours of immersion in water (24 repetitions each), and the formaldehyde emission (06 repetitions each adhesive) (European Standard, 1992).

Statistical analysis

The experiment was done using a completely randomized experimental design, in a factorial arrangement of 2 × 9, with two adhesives and nine material compositions, aside from two additional treatments: control (100% eucalypt considering as an adhesive factor), totaling 20 treatments (Table 1).

The data were analyzed using the software Assistant (version 7.7) and were submitted to the Cochran and Shapiro Wilk tests after the results were statistically analyzed by variance analysis (ANOVA) ($p \leq 0.05$).

Table 1. Raw material composition used in the particleboard production.

Initials	Eucalypt (%)	Papaya Stalks CM (%)	Macadamia nut carpels CNM (%)	Coffee husks PC (%)
Control	100	-	-	-
10% CM	90	10	-	-
20% CM	80	20	-	-
30% CM	70	30	-	-
10% CNM	90	-	10	-
20% CNM	80	-	20	-
30% CNM	70	-	30	-
10% PC	90	-	-	10
20% PC	80	-	-	20
30% PC	70	-	-	30

RESULTS AND DISCUSSION

Apparent density and compaction ratio on particleboards

A significant interaction among the adhesive factors and composition (Table 2) with the apparent density ($F_c = 3.91$; $p < 0.01$) and compaction rate ($F_c = 3.67$; $p < 0.01$) was observed.

Table 2. Mean values of apparent density (g cm⁻³) for the particleboards related to the raw material and adhesive used.

Composition	Apparent density	
	Adhesive	
	Urea-formaldehyde (UF)	Tannin-urea-formaldehyde (TUF)
10% CM	0.663 bB ^{ns}	0.698 aA ^{ns}
20% CM	0.640 cA*	0.641 cA ^{ns}
30% CM	0.597 dA*	0.606 dA*
10% CNM	0.657 bB ^{ns}	0.693 aA ^{ns}
20% CNM	0.658 bA ^{ns}	0.663 bA ^{ns}

Table 2. Continuation...

Composition	Apparent density	
	Adhesive	
	Urea-formaldehyde (UF)	Tannin-urea-formaldehyde (TUF)
30% CNM	0.618 cA*	0.628 cA ^{ns}
10% PC	0.693 aA ^{ns}	0.644 cB ^{ns}
20% PC	0.703 aA ^{ns}	0.671 bB ^{ns}
30% PC	0.658 bA ^{ns}	0.645 cA ^{ns}
Control	0.674 A	0.668 A

CM: Papaya stalks; CNM: Macadamia nut carpels; PC: Coffee husks. Means followed by the same lowercase letter in the column do not differ from each other by the Scott-Knott test ($p > 0.05$). Means followed by the same uppercase letter on the same line do not differ from each other by the F test ($A > B$). ** Differs statistically from the control treatment by the Dunnett test ($p \leq 0.05$); Ns: Does not differ statistically from the control treatment by the Dunnett test ($p > 0.05$).

To evaluate the different components inside the adhesive UF a higher apparent density to the treatment originating from the mix of 10% and 20% coffee husks with the eucalypt wood was verified, while with TUF, higher densities were obtained using 10% of wastes of papaya stalks and macadamia nut shells.

On the adhesive comparison inside each composition, it was observed that only at the composition of 20% and 30% of papaya stalks; and 20% and 30% from carpels of macadamia nuts and 30% of coffee husks did not present statistical differences ($p \geq 0.05$) between the adhesive and the density.

In short, the particleboards produced with the mix with lignocellulosic wastes had apparent densities varying from 0.59 g cm^{-3} to 0.70 g cm^{-3} with the adhesive UF, and 0.61 g cm^{-3} to 0.69 g cm^{-3} to the adhesive TUF, whereupon 26% of the particleboards were classified as of medium density (0.64 g cm^{-3} to 0.68 g cm^{-3}), on both adhesives. Exceptions were the compositions with 30% papaya stalks and 30% macadamia carpel nuts, with both types of adhesives, those being classified as of low density ($< 0.64 \text{ g cm}^{-3}$) (American National Standards Institute, 1999).

As expected, when comparing the apparent density averages of each composition with their respective control treatments, it was observed that, for the most part, treatments composed of mixtures of agricultural waste and eucalypt wood had their densities decreased; there was a tendency of decrease in the density of the particleboards with the increase of the proportion of waste.

Despite the clear reduction of the density of the particleboards, the addition of the agricultural waste had little effect on the density, when compared to the controls. With the UF adhesive, the addition of 20% and 30% papaya stalks to the composition of the particleboard reduced it by 5.04% and 11.42% respectively; and the particleboards with 30% of macadamia nut carpels had a density 8.31% lower than the ones produced with 100% *eucalyptus*. With the adhesive TUF, only the composition 30% of waste from papaya had a statistical difference from the control, showing a 10.09% lower density.

The addition of ligno-cellulosic wastes with low density in the particleboard (Table 3) interfered significantly with the density. Also, there was a significant interaction between the adhesive factor and composition to the compaction ratio ($F_c = 3.67$; $p < 0.01$).

Table 3. Mean compaction ratio of particleboards related to the raw material and adhesive.

Composition	Compaction ratio	
	Adhesive	
	Urea-formaldehyde (UF)	Tannin-urea-formaldehyde (TUF)
10% CM	1.27 bB ^{ns}	1.33 aA*
20% CM	1.32 aA*	1.32 aA*
30% CM	1.33 aA*	1.35 aA*
10% CNM	1.23 bB ^{ns}	1.30 aA*
20% CNM	1.30 bA*	1.31 aA*

Table 3. Continuation...

Composition	Compaction ratio	
	Adhesive	
	Urea-formaldehyde (UF)	Tannin-urea-formaldehyde (TUF)
30% CNM	1.28 bA*	1.30 aA*
10% PC	1.29 bA*	1.19 bB ^{ns}
20% PC	1.36 aA*	1.30 aB*
30% PC	1.32 aA*	1.30 aA*
Control	1.20 A	1.19 A

CM: Papaya stalks; CNM: Macadamia nut carpels; PC: Coffee husks. Means followed by the same lowercase letter in the column do not differ from each other by the Scott-Knott test ($p > 0.05$). Means followed by the same uppercase letter on the same line do not differ from each other by the F test ($A > B$). ** Differs statistically from the control treatment by the Dunnett test ($p \leq 0.05$). Ns: Does not differ statistically from the control treatment by the Dunnett test ($p \geq 0.05$).

Higher means of compaction were obtained with the treatments derived from the composition 20% and 30% from papaya stalks and coffee husks to the UFF adhesive. In the TUF adhesive, only the addition of 10% coffee husks resulted in a significant difference from the other treatments showing a lower compaction rate.

To compare the different adhesives inside each composition, it was verified that the adhesive TUF had a higher compaction ratio to the treatment derivate from the composition 10% papaya stalks, 10% of macadamia carpels and 10% and 20% of coffee husks. By contrast, the increase in the compaction ratio produced from 20% and 30% papaya waste, and 20 and 30% of coffee waste were provided by the UF adhesive.

Comparing the treatment means with their respective controls, it was observed that for the UF adhesive, only the 10% papaya stalk and 10% macadamia nuts did not show a difference from the control ($p > 0.05$). For the TUF adhesive, the same behavior was observed with 10% coffee husk.

It is possible to claim that the waste addition to the particleboard composition provided an increase in their compaction ratio when compared to the control. This is due to the lower density of the materials papaya stalks (0.19 g cm^{-3}), macadamia nut carpels (0.30 g cm^{-3}) and coffee husks (0.34 g cm^{-3}) when compared to the eucalypt wood (0.56 g cm^{-3}) and consequent reduction of possible empty spaces, differently from that observed by Ferreira et al. (2014). The addition of the macadamia nutshell (not the macadamia nut carpel) decreased the density of the particleboards, due mainly to the high density of the raw material, which made the panel compaction more difficult, forming a panel with a larger amount of voids, therefore less dense.

The addition of sisal fiber in the formation of particleboards allowed a gradual increase in compaction ratio (Mesquita et al., 2015), but with fixation of the nominal density, differently from the present work, despite the variation found in some treatments. The low density of ligno-cellulosic materials utilized and the inherent characteristics of each allowed for a reduction of the density in some treatments studied, as mentioned by Iwakiri et al. (2014).

Water absorption (WA) after 2h and 24h immersed into water

There was a significant interaction among the adhesive factor and the composition of the absorption of water (WA) after 2h ($F_c = 11.0$; $p < 0.01$) and after 24h ($F_c = 23.5$, $p < 0.01$) of immersion in water (Table 4).

Table 4. Mean values for water absorption (WA), in percentage after 2h and 24h immersion in water of particleboards according to the raw material and adhesive used.

Composition	WA 2h		WA 24h	
	Adhesive		Adhesive	
	Urea-formaldehyde (UF)	Tannin-urea-formaldehyde (TUF)	Urea-formaldehyde (UF)	Tannin-urea-formaldehyde (TUF)
10% CM	18.33 dA*	19.84 eA*	54.28fA*	38.51cB*
20% CM	27.78bA*	29.27cA*	63.77dA*	40.26cB*
30% CM	44.16aB*	47.10aA*	75.96aA*	46.33aB*
10% CNM	16.64eA*	16.64fA ^{ns}	53.51fA*	36.11dB ^{ns}
20% CNM	20.28dA*	21.57eA*	62.10dA*	39.11cB*
30% CNM	27.22bA*	27.38cA*	71.53bA*	42.44bB*
10% PC	15.72eB ^{ns}	19.36eA*	52.36fA*	38.93cB*
20% PC	18.50dB*	23.96dA*	57.59eA*	41.28cB*
30% PC	23.53cB*	34.61bA*	65.68cA*	44.88aB*
Control	13.57A	14.85A	43.34A	35.19B

CM: Papaya stalks; CNM: Macadamia nut carpels; PC: Coffee husks- Means followed by the same lowercase letter in the column do not differ from each other by the Scott-Knott test ($p > 0.05$). Means followed by the same uppercase letter on the same line do not differ from each other by the F test ($A > B$). ** Differs statistically from the control treatment by the Dunnett test ($p \leq 0.05$). Ns: Does not differ statistically from the control treatment by the Dunnett test ($p \geq 0.05$).

When evaluating the means of WA after 2h and 24h of immersion of the different compositions within each adhesive, different behaviors between treatments were observed. In general, the treatments showed a tendency of increase in the means of WA (in both periods of immersion) with the addition of agro-industrial wastes to their composition. Such behavior is due to the lower density of some particle panels, although there is little difference between compaction ratios, being less expressive in the compositions of 10%. Moreover almost all the compositions of raw materials and adhesives tested presented a WA greater than the control; this fact is due to the characteristics of the different wastes.

Particleboard of low-density made with peanut peel and the fiber of the green coconut and Castor oil based on two components polyurethane adhesive an overlap of coconut fiber to the peanut peel was observed, resulting in bigger empty spaces, providing higher values of water absorption (Cravo et al., 2015).

Different compositions of the particleboards practically did not result in a change in compaction ratio, which was expected ($t \leq 0.05$); although the densities and characteristics of the raw materials allowed for a difference between the treatments (10.16% to 60% to 2h WA and 32.83% to 85.57% to 24h WA). With the increase of the percentage wastes of corn cobs and rice husks added to the pine and eucalypt particles, it was reported in the literature that the dimensional stability of the particleboards was impaired, especially after 2h and 24h WA (Silva et al., 2015; Scatolino et al., 2013; Melo et al., 2009).

Another factor that is related to the higher or lower absorption rate is that the agricultural waste, in general, has a higher hemicellulose content in its composition when compared to wood waste (Martins, 2016). Due to the fact that it is more hydrophilic than cellulose and lignin, the hemicellulose shows higher affinities with water, providing a higher water absorption rate (Li et al., 2011), as in the case of the waste of the papaya stalk, on both glues.

The high water absorption in particleboard produced with eucalypt wood with bamboo was an association of anatomical characteristics (high porosity) and chemical (higher hemicellulose content and extractives) of this material (Melo et al., 2015).

In agreement with the literature, it was shown that in absolute terms, the particleboards produced with the addition of papaya stalks presented higher values of water absorption when compared to the rest; this behavior is explained by the lower lignin content (23.19%) and a consequently higher percentage of holocellulose (Martins, 2016).

The addition of tannic extract to the UF adhesive (TUF) did not influence significantly the 2h WA of the produced particleboard with the papaya stalks waste (with the exception the 30% composition) and the macadamia nut carpels. As for the particleboard produced with coffee husks, the 24h WA ratio used with the adhesive TUF contributed to the increase of this property.

Nevertheless, analyzing the adhesive influence at WA 24h, it was noted that the addition of tannic extract to the UF adhesive (TUF), reacted in a positive form for this property. The urea-formaldehyde adhesive has as main disadvantage its high hygroscopic property, necessitating the addition of paraffin or another product, for the reduction of water absorption (Melo et al., 2015).

When there is tannin in the adhesive composition the water absorption ratio (WA), according to Pizzi & Mittal (2003) may not influence higher absorptions due to the presence of hydroxyl groups in the tannic molecule. There is a possibility to modify the tannic molecule, e.g. by the addition of anhydric acetic acid that could block the free hydroxyl on the tannin (Pizzi & Mittal, 2003). On the other hand, the addition of tannic extract to urea-formaldehyde to produce particleboards from eucalypt as observed by Tostes et al. (2004) contributed to the increase of WA 2h and WA 24h (41.74 to 71.33% and 77.79% to 95.26%, respectively).

In all the treatments, the addition of agro-industrial wastes to the wood particles contributed to an increase of 2h and 24h WA ratio for both adhesives. The increase of the 2h WA ratio compared to the control varied from 15.84% (10% PC) and 225.42% (30% CM) to the produced composition with UF and between 12.05% (10% CNM) and 217.7% (30% CM) to the compositions with TUF. The 24h WA ratio varied from 20.81% (10% PC) and 75.26% (30% CM) to UF and 2.61% (10%CNM) and 31.65% (30% CM) to TUF, respectively.

When comparing the treatments and the control, only the treatment composed by 10% of coffee husks with UF adhesive did not show a significant difference ($p < 0.05$) between the control and 2h WA. With TUF, this behavior was observed only in the 10% macadamia nut carpel treatment of the 2h and 24h WA.

This result showed that the addition of a lower proportion of wastes in the board composition does not affect its responses to WA when compared to the ones produced with 100% eucalyptus sawdust. The proportion increase by fewer particles and the composition of the board contribute to the increase of empty space in its structure, contributing to the WA.

Thickness Swelling (TS) after 2h and 24h immersion

The variance analyses show a significant interaction among the adhesive factors and the composition to the TS propriety after 2h ($F_c = 9.27$; $p < 0.01$) and after 24h ($F_c = 7.58$; $p < 0.01$) immersed in water (Table 5).

Table 5. Mean values for thickness swelling (TS), in percentages after 2h and 24 h immersion in water of particleboards according to the raw material and adhesive used.

Composition	TS 2h		TS 24h	
	Adhesive		Adhesive	
	Urea-formaldehyde (UF)	Tannin-urea-formaldehyde (TUF)	Urea-formaldehyde (UF)	Tannin-urea-formaldehyde (TUF)
10% CM	5.15dB*	7.89dA ^{ns}	16.53cB*	25.71bA ^{ns}
20% CM	7.49cB*	9.01cA*	17.98cB*	23.51cA ^{ns}
30% CM	8.62bB*	13.55bA*	15.92cB ^{ns}	23.72cA ^{ns}
10% CNM	5.22dB*	7.85dA ^{ns}	16.45cB ^{ns}	22.64cA*
20% CNM	6.20dB*	8.85cA*	17.32cB*	26.22bA ^{ns}
30% CNM	7.38cB*	9.60cA*	16.50cB ^{ns}	25.25bA ^{ns}
10% PC	5.66dB*	7.65dA ^{ns}	16.91cB*	26.27bA ^{ns}
20% PC	7.14cB*	9.23cA*	20.39bB*	27.07bA*
30% PC	9.95aB*	15.72aA*	23.28aB*	34.82aA*

Table 5. Continuation...

Composition	TS 2h		TS 24h	
	Adhesive		Adhesive	
	Urea-formaldehyde (UF)	Tannin-urea-formaldehyde (TUF)	Urea-formaldehyde (UF)	Tannin-urea-formaldehyde (TUF)
Control	3.93B	7.17A	15.02B	24.79A
Associação Brasileira de Normas Técnicas (2006b)	Maximum 8%		-	
Commercial Standard (1968)	-		30-35%	

CM: Papaya stalks; CNM: Macadamia nut carpels; PC: Coffee husks- Means followed by the same lowercase letter in the column do not differ from each other by the Scott-Knott test ($p > 0.05$). Means followed by the same uppercase letter on the same line do not differ from each other by the F test ($A > B$). ** Differs statistically from the control treatment by the Dunnett test ($p \leq 0.05$). Ns: Does not differ statistically from the control treatment by the Dunnett test ($p > 0.05$).

When evaluating the TS means after 2h and 24h of immersion in water of the different compositions with each adhesive, different behaviors between the treatments were observed. Similar to what occurred with the WA ratio; the addition of agro-industrial waste to the boards provided an increase of TS during the 2h period with both adhesives.

The waste type that was added to the particles creates an increase trend of TS; e.g., when corn straw is added to the eucalypt particles (Silva et al., 2015), maize cob to pine particles (Scatolino et al., 2013) and rice peel to the eucalypt sawdust (Melo et al., 2009). This may influence the dimensional stability of the particleboards since they are also associated with the geometry and anatomical characteristics of the material used.

When analyzing thickness swelling after 24h of water immersion, a significant difference was not verified between the means with the percentage of waste from papaya stalks and macadamia nut carpels when the UF adhesive was used; however, the addition of coffee husks caused an increase on the 24h TS with the same adhesive. The addition of a higher percentage of macadamia nuts carpel waste and coffee husks resulted in a higher 24h TS on the particleboards produced with TUF adhesive, but the addition of papaya stalks decreased the 24h TS.

When evaluating the TS property; it was found that it had higher values in the boards with higher compaction. A higher compaction ratio can liberate more prehensile tension when immersed in water (Kim & Kim, 2005).

In contrast to the literature, which says that when tannin is added to chemical adhesives in the presence of formaldehyde it produces a moisture resistant adhesive (Pizzi et al., 1981; Gonçalves et al., 2008) and results in particleboards of satisfactory stability (Melo et al., 2010); the present work showed the opposite (increase of 11% for WA 2h and 30% for IE 2h). The exception occurred for WA 24h with a 34% reduction. The type of waste may have contributed to this behavior, mainly due to the density and chemical composition of the materials.

When analyzing the treatment means with their respective controls in all treatments, it was found that the addition of agro-industrial waste to eucalypt wood favored the increase of the 2h and 24h ratio in both adhesives, with the only exception of the 10% CNM with TUF adhesive, which showed a reduction of the 24h TS. The increase of 2h TS ratio compared to the control varied between 31.04% (10% CM) and 153.18% (30% PC) in the compositions produced with UF; and between 6.69% (10% PC) and 119.25% (30% PC) in the composition produced with TUF. With the 24h WA, it varied from 5.99% (30% CM) and 54.99% (30% PC) in TUF, respectively.

With the TUF adhesive, only treatments composed of 10% CM, 10% CNM and 10% PC did not give a significant difference ($p < 0.05$) between the 2h TS while the 24h TS in the majority

of the treatments (10, 20, 30% CM; 20 and 30% CNM; 10% PC) did not differ from the control. When evaluating the 24h TS it was observed that the 30%CM and 10 and 30% CNM differed from the control treatment with UF adhesive.

The increase of WA and TS can also be attributed to the higher volume of particles of lower densities added to the panels, allowing greater releases of the compression stresses imposed during the pressing (Iwakiri et al., 2005).

The majority of the particleboards produced with UF adhesive did not exceed the established limit of the norm (Associação Brasileira de Normas Técnicas, 2006b) to the 2h TS (8% maximum), except the 30% CM and 30% PC composition. Nevertheless, the TUF adhesive with a composition of only 10% of wastes had a swelling lower than 8%. Aside from the 10% tannic extract added to the UF adhesive which increased significantly the 24h TS values; those values were in agreement with the norm COMMERCIAL STANDARDS – CS 236-66 (Commercial Standard, 1968) class 1, showing a lower TS at the 30% and a maximum at 35% in the low and medium density boards, respectively.

Formaldehyde emission in particleboards

A difference of the formaldehyde was detected between the control (UF) and FF (13.5; $p < 0.01$) (Table 6). The particleboards that were produced with 100% eucalypt using both types of adhesives were classified in class E2 in formaldehyde emission (8-30 mg 100g⁻¹) (European Standard, 1992). The particleboard had a reduction of 22.48% in the emission of formaldehyde.

Table 6. Emission of formaldehyde from particleboards according to the raw material and adhesive used and the control treatment (100% eucalypt) in mg 100g⁻¹.

Adhesive	Control
Urea-formaldehyde (UF)	11.34 (57.66) a
Tannin-urea-formaldehyde (TUF)	8.79 (26.85) b

a>b by F test; values in parentheses correspond the variation coefficient

With the tannic extract added to the UF adhesive a 22.5% reduction of the formaldehyde was verified. It is estimated that the addition of a higher percentage of tannic extract to the adhesive urea-formaldehyde may further reduce the emission of formaldehyde in particleboards. However, it is necessary to test to which level this addition will bring about its emission benefits without any practical influence on the board properties.

The free formaldehyde reduction on the particleboard might be associated with the chemical composition of the tannin, since the condensation reaction of the formaldehyde with tannin occurs more easily than with urea.

The replacement of adhesives for reduction of formaldehyde emission was abundantly researched by Trianoski et al. (2014), who found that there was a 76% reduction in the emission of formaldehyde in MDF from *Pinus densiflora* when urea-formaldehyde adhesive was replaced by melanin-formaldehyde.

In *Abies cephalonica* particleboards, at humidity $\leq 10\%$, glued with pure urea-formaldehyde, a 12.31 mg kg⁻¹ emission medium value was found in the sample method, showing that the particle humidity might be a limiting factor (Petinarakis & Kavvouras, 2006).

The addition of tannin to the urea-formaldehyde adhesive in particleboards made from rice husk and charcoal reduced the formaldehyde emission level (Eon et al., 2006).

The emission of formaldehyde in particleboards bonded with urea-formaldehyde can be further reduced by the optimization of the production process by controlling the variables pressing temperature, apparent density and moisture content of the particles in admixture with the glue before the pressing phase (Petinarakis & Kavvouras, 2006).

CONCLUSION

The percentage increment of the wastes on the particleboard's composition provided an increase in the compaction ratio, water absorption and thickness swelling.

Regardless of the adhesive and composition, the thickness swelling after 24h of immersion in water met the norms' specification.

The addition of 10% tannin extract on the urea-formaldehyde reduced the emission of free formaldehyde on the particleboards by about 21%.

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ERRATUM

Erratum

In the article “Physical properties and formaldehyde emission in particleboards of *Eucalyptus* sp. and lingo-cellulosic agro-industrial waste”, DOI number <https://doi.org/10.18671/scifor.v48n125.13>, published in **Scientia Forestalis**, vol. 48, no. 125, e2926:

The article title reads:
“lingo-cellulosic”

and should read:
“ligno-cellulosic”

Throughout the article body, header and how to cite the following term reads:
“lingo-cellulosic”

and should read:
“ligno-cellulosic”

