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FORESTRY SCIENCE

Colour changes and equilibrium moisture content on thermomechanical densified wood

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Abstract: Color change associated with significative positive improve in physical properties is a challenge in wood research. This study investigated the changes in the color of the *Gmelina arborea* wood which underwent a thermomechanical densification process. The process was performed by applying three different temperatures (140 °C, 160 °C and 180 °C) with thickness reduction of 20% and 40% using 2.5 MPa equivalent pressure. The color change of the pieces was analyzed through the CIEL * a * b * system. The relationship between the color and the equilibrium moisture content of the densified material was also analyzed. The process reduced the lightness and yellow hue of the wood, with increased red pigment resulting in darker coloration of the treated pieces. The higher temperature used resulted in more significant changes in wood tone and lower equilibrium moisture. The process proved to be effective to change the color and significantly reduce the equilibrium moisture content in wood samples where the density was increase by 20% and 50% compared to natural wood.

Key words: Colorimetry, moisture content, densification, *Gmelina arborea*.

INTRODUCTION

Aesthetics factor have the important function of allowing conscious and appropriate use of lignocellulosic products, reflecting the surface characteristics of the raw material. The color, factor related to the aesthetics and decoration potencial of wood-based products, can the value added to the final product.

Different color shades of wood of a specific species can be controlled using variations in processes that use temperature as a modifying agent, as occurs in thermomechanical densification. This process which causes changes in the chemical composition and structure of the wood cell wall, and consequently in the properties of wood.

To determine the color and its changes, can be used the system CIEL*a*b*, which uses colorimetric parameters such as: luminosity (L*), chromatic coordinates of green - red (a*) and yellow - blue (b*) axes, color saturation (C*) and hue angle (h) for evaluation and description of color shades on surfaces. With the application of this system, one has the possibility of qualitative classification of the raw material. In the case of woods, this technology makes it possible to classify to group clones with similar color chacacteristics (Moya-Roque & Marín 2011), influence of heat treatments (Cademartori et al. 2014) and evaluations of the effectiveness of surface wood protections against the action of photodegradation (Yuan et al. 2019).

The color of wood can be a decisive factor for choice of a final consumer wood, due to its visual appearance often prevailing (Esteves & Pereira 2009), in addition to increasing value in dark-hued woods compared to wood with light colored (Herrera et al. 2018). The darker shades obtained in processes that use temperature are mainly due to the oxidation of phenolic compounds (Hidayat et al. 2015), formation of secondary compounds, migration of surface extractives and degradation of hemicelluloses (Cademartori et al. 2014, Tomak et al. 2014).

The color becomes important for the use of wood, especially when transparent coatings are applied, adding aesthetic and economic value, as these parameters are used to evaluate the quality of finished wood products (Bekhta et al. 2018). Unlike the heat treatments and others process used to change the certain properties of wood, the thermomechanical densification process enable the material color change, changes in physical and mechanical properties, such as increase in the density and resistance. These can reach different levels within the wood due to the combination of pressure and temperature.

Such alterations potentiate the use of this technique, especially when applied to light colored and low density woods, such as *Gmelina arborea* Roxb.. Therefore, changes obtained by the process parameters may be an alternative to wood application options, based on aesthetic and structural factors, thus adding value to the final product.

In this sense, the present study aimed to evaluate the superficial color changes and their relationship with the equilibrium moisture content of *Gmelina arborea* Roxb, submitted to thermomechanical densification process.

MATERIALS AND METHODS

The study was developed from the application of thermomechanical densification process using a hydraulic press (Siempelkamp, Germany) equipped with plates heated by electric resistance. Temperature and pressure were applied in 10 samples by treatment of dimensions 25 x 100 x 2500 mm (thickness x width x length), prepared from the wood of *Gmelina arborea* Roxb.

The samples were compressed perpendicular to the fibers, using press plate temperatures of 140 °C, 160 °C and 180 °C. Pressure was applied to achieve two final thickness levels, 20 mm and 15 mm, corresponding to a thickness reduction of 20% and 40%, respectively. Were used lateral restrictions to homogenize thickness. Control analysis and five treatments were performed (Table I).

The pieces were accommodated between the previously heated plates and applied pressure, following a 0.5 MPa ramp every minute until reaching 2.5 ± 0.3 MPa. The press was kept closed with pressure and temperature for a period of 30 minutes. After this period the temperature of plates was off, following by a temperature and pressure reduction ramp until reaching 0.5 MPa, keeping in this condition for 240 minutes to reduce the wood temperature and stress relief and internal vapor pressure (Figure 1).

The evaluation of colorimetric parameters was performed according to the system CIEL*a*b* using a KONICA MINOLTA CM-5

Table I. Outline of treatments used in the wood of *Gmelina arborea.*

Treatments	Control T0	140 °C		160 °C		180 °C
		T1	T2	T3	T4	T5
Final thickness	25 mm	20 mm	15 mm	20 mm	15 mm	20 mm
Thickness reduction	-	20%	40%	20%	40%	20%

spectrophotometer coupled to a microcomputer. The colorimetric parameters were obtained from 4 random points, on the abaxial and adaxial sides, from six samples per treatment. Were determined: L* (luminosity); a* (red-green axis coordinate); b* (blue-yellow axis coordinate); *C* (saturation) indicating the color purity and h* (hue angle) indicating the tone dominance.

Changes in wood color after treatment were determined by the total color variation (ΔE^*), according to Equation 1.

Where: EMC = equilibrium moisture content (%); IM = initial mass (g); FM = final mass (g); *d* = bulk density ($g.cm^{-3}$); M = mass acclimatized (g); V = volume acclimatized (cm^3).

Data were tabulated in a spreadsheet and analyzed for variance homogeneity (Bartlett and Hartley's) and data normality (Kolmogorov - Smirnov). The analysis of the parameters was conduct through the Analysis of Variance (ANOVA), and where $p \leq 0.05$ was observed, the tukey test ($p \leq 0.05$) was also performed.

RESULTS AND DISCUSSION

Wood samples of *Gmelina arborea* natural and treated were characterized and compared by the system CIEL*a*b*, enabling to identify that from the natural wood to T5, there was a reduction of lightness values (L*), associated with the intensification of the red pigment (a*) and reduction of the yellow color (b*). Consequently, the wood color became darker, and this change was observed with greater magnitude in T5 (Figure 2).

It can be seen from the visual analysis of the wood pieces that there was a highlight of the staining in the late wood of the treated samples, resulting in some color heterogeneity, evidenced in treatments T3, T4 and T5. This fact is attributed to the heterogeneous chemical composition of the rings (Atik et al. 2013), especially the greater presence of extractives in this region of the wood (late wood), consequently greater migration of these components to the surface with the application of temperatures above 160 °C, acquiring a darker shade as a consequence

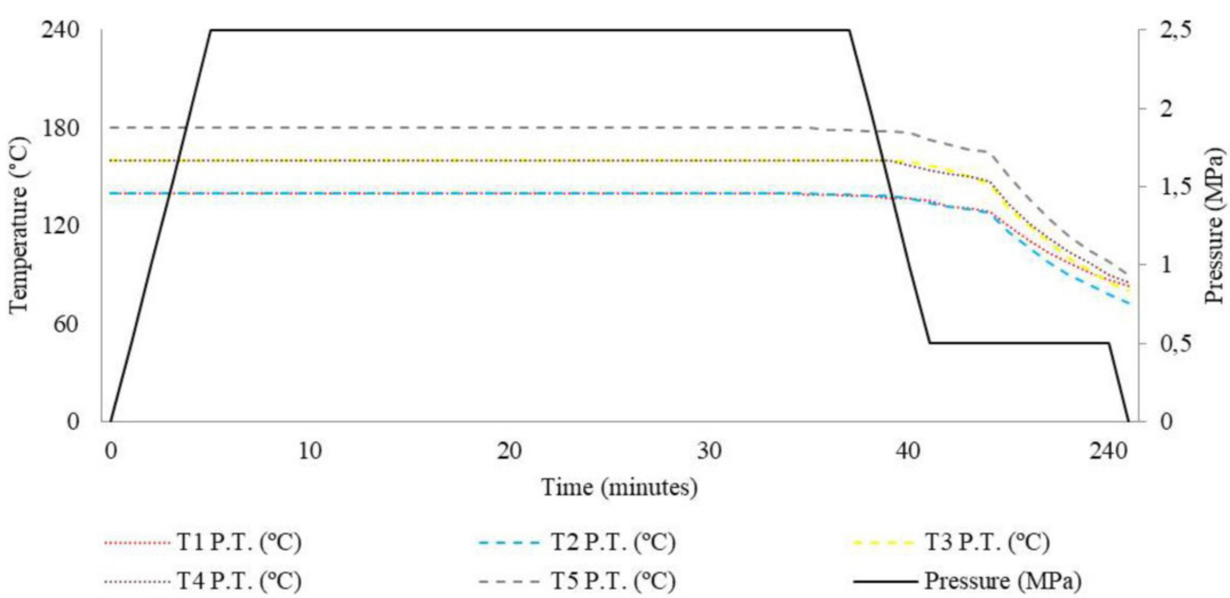

Figure 1. Diagram of the thermomechanical densification process with the temperatures of the hydraulic press (P.T. = Press temperature).

of the temperature action on the wood (Diouf et al. 2011, Sheshmani 2013).

The temperature of 140 °C used in treatments T1 and T2 may not have caused the total migration of the extractives to the surface, resulting in tones more similar to the natural color of the wood. T1 showed a slightly darker color than T2, which may be explained by the complex and heterogeneous nature of the wood, especially by the higher levels of extractives in the samples used in this treatment.

The hue angle is the result of the chromatic matrices a* and b* showing the treatment behavior more similar to the yellow - blue chromatic coordinate (b*), because the value of this parameter is larger compared to the green - red chromatic coordinate (a*). The reduction of the hue angle is attributed to reduction of differences in shades after treatment (Sodero Martins Pincelli et al. 2012), which occurs as temperature increases, replacing of yellow tones with red tones.

It is noticed that the hue angle curve followed the trend of the parameter L* curve. This occurs gradually depending on the treatment applied, as there is minimization of lighter shades (yellow) and increase of darker shades (red). Saturation (*C*), which is dependent on the chromatic matrices a* and b*, showed a behavior similar to that observed in the parameter h*, since, generally, the higher values of the b* matrix had a greater influence on this parameter in comparison to matrix a*, tending to darks tones.

The increase of matrix a*, especially from T4 to T5, favored the change of the samples tones, since reddish tones are associated with the darkening of the sample. Thus, the

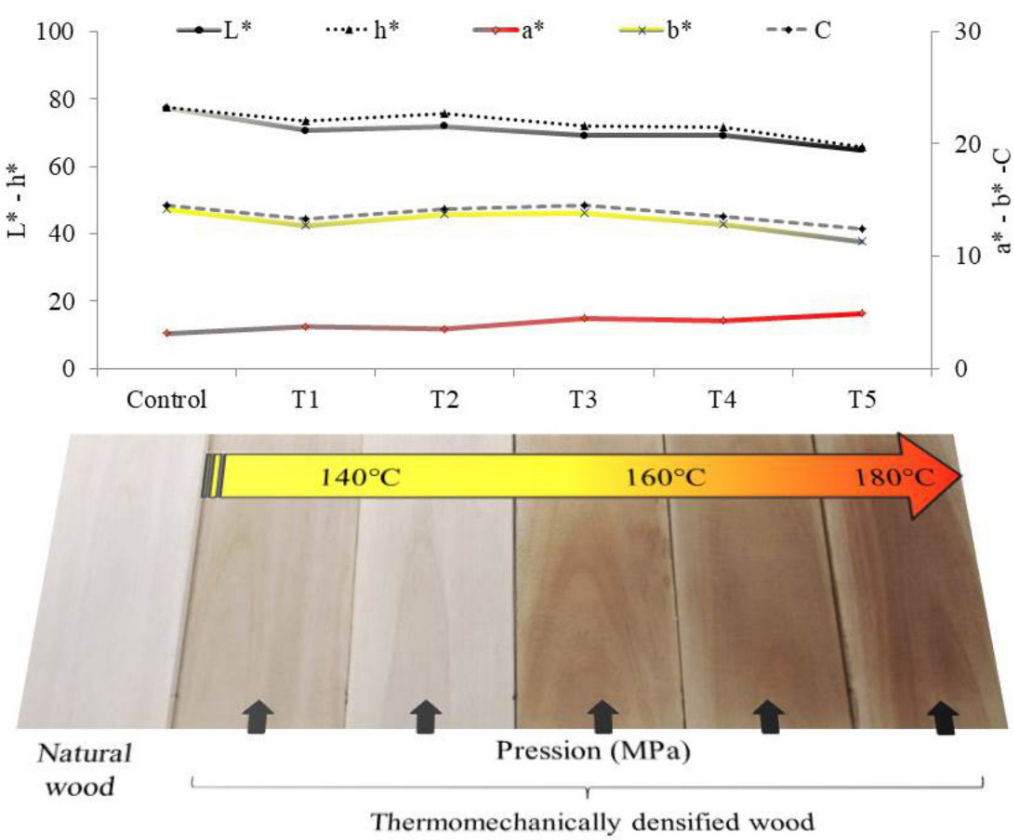

Figure 2. Parameter behavior: L*, a*, b*, *C* E h* of the samples of *Gmelina arborea* in natural condition and after application of thermomechanical densification process.

color of wood has become less vivid and with greyish appearance, as the temperature of the treatments increases. The above changes can be observed in the L* x *C* diagram (Figure 3).

Natural wood is pale to very pale. With the application of the treatments, the tone is changed to tones closer to gray, with greater evidence in the T5 treatment, which has a higher temperature.

The luminosity (L*) and the chromatic coordinate a* (red) were statistically different between the control and the treatments, indicating that the process changed the luminosity and increased the red pigmentation of the wood, because there was an increase in hue a * of the treatments in relation to control (Table III).

From the average values of wood luminosity (L*), it was possible to note that this parameter was modified with the application of treatments, reducing with increasing temperature, and consequently, making the wood darker. It was observed that the compaction did not change this parameter, since the luminosity between treatments with the same temperature did not present statistically significant difference between them. Significant differences were identified between temperature variations, and it was statistically equal only between T1 and T3 and T4 temperatures.

Such differences were also perceived for the parameter: a* as temperature changes, forming a relation between the increase of this property value and the increase of temperature, changing the intensity of the pigmentation of the wood tones to red.

The control samples presented a higher intensity of yellow pigmentation (b*) compared to the treated pieces, but were not statistically different from T2 and T3, indicating that there was no variation of yellow pigmentation between such treatments. The T5 presented the lowest average, with aless intense shade ofred color, statistically different from the others.

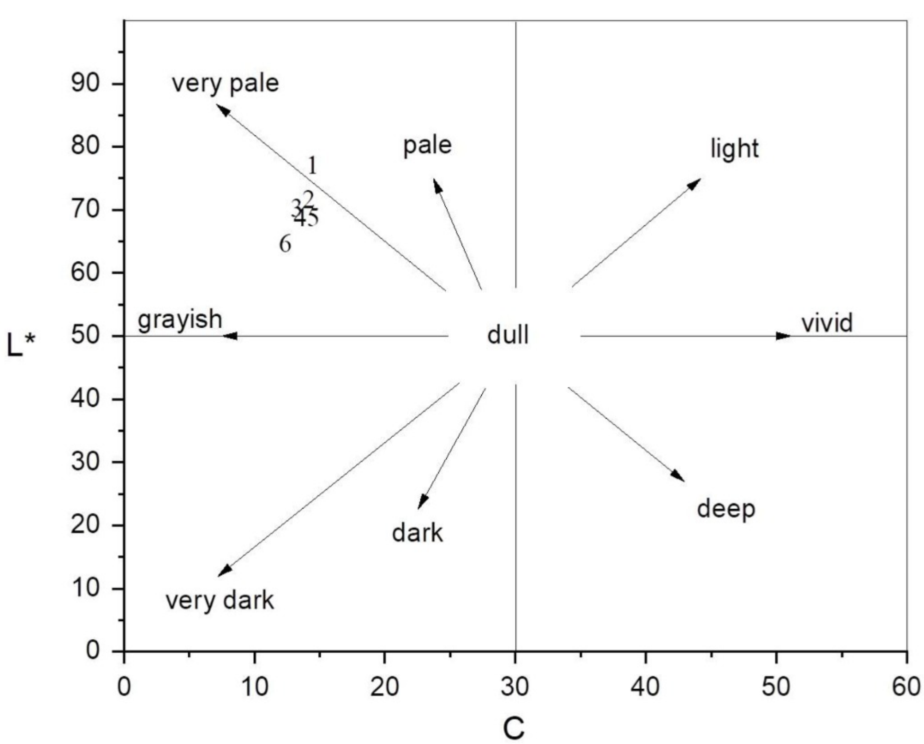

Figure 3. Diagram of luminosity (L*) x saturation (*C*) of *Gmelina arborea* samples in natural condition and treated. Note: 1=Control; 2=T1; 3=T2; 4=T3; 5=T4; 6=T5.

The parameter ΔL^* presented a decreasing configuration as the process temperature increased, and the more negative the value of ΔL^* the greater the color modification. Parameter Δa^* increased with temperature, while for parameter Δb^* there was a reduction in relation to the control for all treatments, but there was no orderly sequence due to temperature increase and thickness reduction (Table IV).

The color neutrality was less evident in the T5 x Control ratio, which associated with the luminosity variation (ΔL^*) of -15.94% indicates a darker and more intense coloration of red pigments. Regarding the relationship between T2 x Control, it is noted that from the chromatic combination, the color of the wood approached to more neutral tones, that is, with a color less dark.

Analyzing the variations of the colorimetric parameters, it can be observed that for ΔL^* all treatments presented negative values, with the lowest variation corresponding to -6.99% for T2 x Control ratio. This ratio also presented lower value of Δa^*, corresponding to 10.48%.

For Δb^* the lowest variation was observed between T3 and control (-2.26%). The highest differences between colorimetric values were observed for T5 x Control ratio, corresponding to -15.94%, 56.51% and -20.11% for the parameters ΔL^*, Δa^* and Δb^*, respectively. Thus, the treatment that applied higher temperature resulted in higher change of the colorimetric parameters in relation to the natural color of the wood, therefore, presenting the superior value of the total color variation (ΔE^*), mainly influenced by the reduction of the luminosity of the wood (ΔL^*), corresponding to -12.31, followed by the reduction of the matrix Δb^* (-2.85), due to the elevation of the matrix Δa^* (1.78), causing increased red pigmentation (Figure 4).

This sequence of influence on the total color variation was not observed only between T3 x Control, but the hue Δa^*(1.31) presented greater influence than the matrix Δb^* which indicates

Table II. **Total color variation rating (ΔE^*) of woods.**

Color variation (ΔE*)	Classification
0.0 – 0.5	Insignificant
0.6 – 1.5	Slightly noticeable
1.6 – 3.0	Remarkable
3.1 – 6.0	Appreciable
6.1 – 12.0	Very appreciable

Table III. **Mean values of the colorimetric parameters of the samples *of Gmelina arborea* in natural condition and treated.**

Treatment	L*	a*	b*	*C*	h*
Control	77.24 a	3.15 a	14.17 a	14.52 a	77.45 a
T1	70.49 bc	3.75 b	12.79 b	13.34 c	73.51 b
T2	71.84 b	3.48 b	13.75 a	14.19 ab	75.75 a
T3	69.03 c	4.46 c	13.85 a	14.57 a	72.05 bc
T4	68.97 c	4.28 c	12.88 b	13.59 bc	71.50 c
T5	64.93 d	4.93 d	11.32 c	12.39 d	65.78 d

Where: L* = luminosity; a* and b* = chromatic coordinates; *C* = saturation; h* = hue angle. The values in a column followed by the same letter show no statistical difference by Tukey's Mean Test at 5% probability of error.

a more reddish pigmentation of the wood in relation to the control and a practically neutral value of the matrix Δ*b**, equivalent to -0.32, that is, very similar to the natural shade of wood for this parameter. According to the table proposed by Hikita et al. (2001), the color variation of T2 in relation to the control is considered appreciable. For the other treatments, the change was more significant, considered very appreciable (Figure 4).

Such changes in wood coloration are attributed to changes in chemical compounds by the effect of temperature (Cademartori et al. 2014, Tomak et al. 2014, Hidayat et al. 2015), and it is intensified with the increase of the applied temperature, considered the most influential factor in color change in processes that use pressure and temperature (Bekhta et al. 2014). This colorimetric change is understood as a positive factor, as it can be more aesthetically attractive.

Associated with the color change with temperature application, the crystallinity index increases in wood due to the reduction of the amorphous portion of hemicelluloses (Navi & Sandberg 2012). This fact results in a relative increase in the content of other chemical components. This directly reflects in a change in the equilibrium moisture of the samples, reducing this property with increasing temperature, with significant difference between the control and the treated parts, especially T5, which presented the lowest equilibrium moisture content (Table V).

Based on the equilibrium moisture content and colorimetric parameters, strong correlations between the variables of each treatment were noted (Table VI). The strong and positive correlation for luminosity and equilibrium moisture content (0.9996) stands out, indicating that the change of one causes a practically linear modification in the other variable.

These correlations are related to chemical change, because during the densification process there are changes in the chemical composition of the wood (Neyses et al. 2016, Yin et al. 2017), such as extractive migration and hemicelluloses degradation, changing its hue and reducing the accessibility of water to wood.

Parameter a* showed a strong and negative association with equilibrium moisture content, in other words, a reduction in this property caused the opposite effect on the a* coordinate. These results indicate that color change presupposes a change in the equilibrium moisture content, corroborating the literature (Budakçi et al. 2012, Baar et al. 2019).

In addition, the changes in the density of treated wood in relation to natural wood were highlighted, corresponding to an approximate increase of 20% for treatments with 20% reduction in thickness and 50% for treatments with 40% reduction in thickness (Table V). That is, the initial density of 0.45 $g.cm^{-3}$ showed significant gain after the treatments, which indicates that the treatments positively modify, besides the color, the wood properties.

Table IV. Percentage variations between the values of the colorimetric parameters of the samples of *Gmelina arborea* after the application of the treatments in relation to the control wood.

Relation	Δ*L** (%)	Δ*a** (%)	Δ*b** (%)
T1 x Control	-8.74	19.05	-9.74
T2 x Control	-6.99	10.48	-2.96
T3 x Control	-10.63	41.59	-2.26
T4 x Control	-10.71	35.87	-9.10
T5 x Control	-15.94	56.51	-20.11

Where: ΔL* = luminosity variation; Δa* = red parameter variation and Δb* = yellow parameter variation.

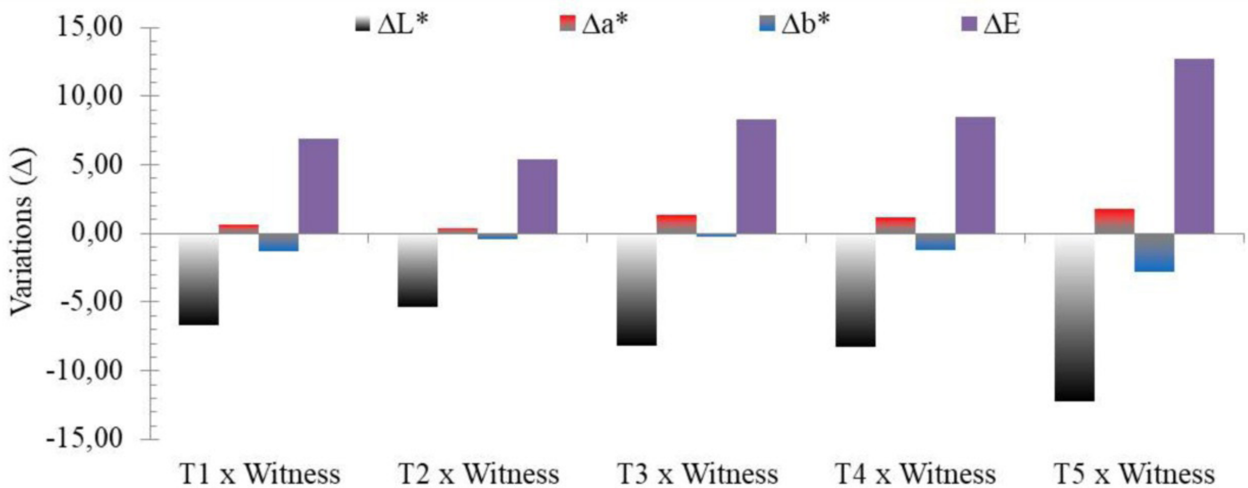

Figure 4. Variations between the values of the colorimetric parameters of the wood samples of *Gmelina arborea* after the application of the treatments in relation to the control wood.

Table V. Equilibrium moisture content and density of *Gmelina arborea* wood samples in natural condition and thermomechanically treated.

Treatments	Equilibrium moisture content (%)	Density (g.cm^{-3})
T0	13.06 (3.00) e	0.45 (1.58) c
T1	10.93 (5.01) d	0.56 (5.59) b
T2	10.34 (8.47) cd	0.68 (6.67) a
T3	9.27 (11.75) b	0.54 (3.58) b
T4	9.46 (6.52) bc	0.67 (3.71) a
T5	7.62 (4.56) a	0.54 (4.11) b

Where: In parentheses the coefficient of variation (%). Values followed by the same letter show no statistical difference by Tukey's Mean Test at 5% probability of error.

Table VI. Pearson correlation between equilibrium moisture content (EMC) and colorimetric parameters L *, a *, b *, *C* and h *.

	EMC	L*	a*	b*	*C*	h*
EMC	1					
L*	0.9963	1				
a*	-0.9599	-0.9397	1			
b*	0.8114	0.8132	-0.7336	1		
C	0.7213	0.7291	-0.6163	0.9867	1	
h*	0.9524	0.9385	-0.9564	0.8941	0.8097	1

Where: EMC = Equilibrium moisture content; L* = luminosity; a* and b* = chromatic coordinates; *C* = saturation; h* = hue angle.

CONCLUSIONS

The application of the thermomechanical process in wood changed their colorimetric parameters and the equilibrium moisture content. The lightness and yellow tone of the wood were reduced with an increase in the red pigment, giving a darker coloration to the treated woods.

The higher temperature applied in the wood densification of *Gmelina arborea* resulted in greater surface darking and lower equilibrium moisture.

The application of thermomechanical densification treatments increased the density, not limiting the superficial color change, and it is considered an efficient technique for such modifications.

The application of the thermomechanical densification process is usable to obtain different colorimetric shades of wood associated with positive modifications of physical properties, enhancing the use for a same species according to the preference of the consumer market.

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