Mechanical Properties and Formaldehyde Release of Boards Manufactured with Hygrothermally Treated Tepa (Laureliopsis Philippiana Looser) Particles

Mehanička svojstva i oslobađanje formaldehida ploča proizvedenih od hidrotermički obrađenog iverja drva tepe (Laureliopsis philippiana Looser)

Original scientific paper • Izvorni znanstveni rad
Received – prispjelo: 3. 3. 2017.
Accepted – prihvaćeno: 23. 5. 2018.
UDK: 630*863.21

ABSTRACT • Three layer particleboards were manufactured using hygrothermally treated and untreated tepa (Laureliopsis philippiana) particles with urea-formaldehyde (UF) resin. Hygrothermal treatment consisted of autoclaving particles at 150 °C for 90 min (430 kPa) in a steam saturated atmosphere. A decrease in density and slenderness ratio in treated particles was observed. Modulus of elasticity (MOE) and modulus of rupture (MOR) in static bending, internal bond (IB), and formaldehyde release were determined. An increase in MOE, MOR and IB, as well as a decrease in formaldehyde release, was reported in boards made from heat treated particles.

Key words: Particleboard, hygrothermal treatment, Laureliopsis philippiana, urea-formaldehyde, particle geometry, mechanical properties, formaldehyde release

SAŽETAK • Za potrebe ovog ispitivanja proizvedene su troslojne ploča iverice od neobrađenog i hidrotermički obrađenog iverja drva tepe (Laureliopsis philippiana) primjenom urea-formaldehidnog (UF) ljepila. Hidrotermička se obrada sastojala od zagrijavanja iverja pri 150 °C tijekom 90 minuta u atmosferi zasićenoj vodenom parom (pri tlaku 430 kPa). Nakon hidrotermičke obrade čestica točno je smanjenje gustoće i vitkosti čestica. Troslojinim su pločama ivericama određeni modul elasticnosti (MOE) i modul loma (MOR) pri statičkom savijanju, čvrstoća raslo-

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1 INTRODUCTION

1. UVOD

The production of boards with *L. philippiana* particles using UF as adhesive does not produce favourable results. If an excess of catalysts is not used, bonding failures are produced in the core of the board, leading to blowout (Poblete and Peredo, 1990; Pinto and Poblete, 1992). The high pH value of this species prevents proper hardening of the adhesive (Poblete, 2001). Thermal treatment using pressurized vapour results in the formation of organic acids that catalyze the hydrolysis of hemicellulose, and to a lesser extent, amorphous cellulose (Mitchell, 1988). The formation of acetic and formic acids at temperatures between 100 and 200 °C reduces the pH value of wood (Garrote et al., 1999; Nuopponen et al., 2004), encouraging the hardening of UF and improving mechanical properties.

The main objection to using thermally treated wood in the manufacturing of particleboards is the loss of mechanical strength. Paul and Ohlmeyer (2010) reported that the effects on the MOR of the boards are negligible, while the effects on IB strength, and particularly on MOE, are higher.

1.1 Effect of thermal treatment on mechanical properties of the boards

1.1. Utjecaj toplinske obrade na mehanička svojstva ploča

Thermal treatment of particles produces a 20 to 25 % increase in bending strength in boards manufactured with UF (Tomek, 1966). However, it has also been reported that MOE, MOR and density of thermally treated MDF decrease with increasing temperature (Ayrilmis et al., 2009). Water steam treatment of particles and fibers at temperatures over 200 °C may result in a reduction of the wood-adhesive adhesion (Boonstra et al., 2006), which reduces the IB of the boards (Mohebby et al., 2008, Ayrilmis et al., 2011), since thermal treatment may alter the adhesive distribution on the surface of the wood and the penetration within the porous structure of the wood (Sernek et al., 2008). Paul et al. (2007) state that wood, usually hydrophilic, becomes hydrophobic after thermal treatment. The loss of mechanical properties can be related to the formation of soluble chemical products by the degradation of hemicellulose, such as formic acid and acetic acid, which accelerate the depolymerisation of carbohydrates (Garrote et al., 2001; Sundqvist et al., 2006). The apparent contradictory results reported in the literature, may be because the thermal treatment of boards affects their whole structure and the interaction wood-adhesive-wood, degrading the adhesive bonds. Other important aspects are the temperature and conditions of thermal treatment employed.

When high temperatures and pressures were employed, a higher degradation of the wood components is caused, reducing the mechanical properties.

1.2 Effect of thermal treatment on formaldehyde release

1.2. Utjecaj toplinske obrade na oslobađanje formaldehida

Roffael (2012) reported that, when defibration temperature is increased, it results in acidification of fibres and a reduction in the release of formaldehyde from MDF. Petersen et al. (1973) reported that particleboards manufactured from wood with low pH value emit lower quantities of formaldehyde than boards manufactured from wood with greater pH value.

1.3 Objectives

1.3. Ciljevi

The main objective of this study was to determine the effect of a hygrothermal treatment in steam saturated atmosphere on *L. philippiana* particles and the implications on properties of three layer particleboards. Specific objectives included the study of changes in particle geometry that can take place during gluing process. In particleboards, changes of MOE and MOR in static bending, IB, and formaldehyde release were determined.

2 MATERIALS AND METHODS

2. MATERIJALI I METODE

2.1 Particles production and hygrothermal treatment

2.1. Proizvodnja iverja in hidrotermička obrada

*L. philippiana* wood was harvested from the XIV Region de Los Ríos near to the Andean mountains in Chile. The wood was cut into shavings and then, using a Pallmann PZ2 mill, particles were obtained. A quantity of the particles was treated in the autoclave at 150 °C for 90 min at 430 kPa pressure, in a steam saturated atmosphere.

2.2 Drying of particles

2.2. Sušenje iverja

Hygrothermally treated and non-treated particles were subjected to a drying process, in a Heraeus particle dryer with air circulation at 70 °C, until a moisture content of 4 % was reached.

2.3 Particle properties: dimensions, slenderness ratio, specific surface area and available adhesive quantity

2.3. Svojstva čestica: dimenzije, vitkost, specifična površina in raspoložljiva količina lepila

In order to quantify changes caused by the treatment, particles were subjected to the mechanical action of the blender without adhesive. After this procedure, 120 g of particles were sieved in a Retsch sieving machine using mesh size of 4.0, 2.8, 2.0, 1.4, 1.0, 0.5 and <0.5 mm. Each particle class was weighed to determine the proportional participation. In 30 particles of every class, length and thickness were determined using a micrometer and caliper. Basic density of the par-
particles according to TAPPI T 258 om-94 was calculated. With the particle length, thickness and density data, slenderness ratio (Poblete and Peredo, 1990), specific surface area and available adhesive quantity (Poblete, 2001) were determined by the formula:

\[ \text{Specific surface area} = \frac{0.2}{\text{Thickness (mm) x Density (g/cm}^3) \right] \]

\[ \text{Available adhesive} = \frac{\text{Density (g/cm}^3) \times \text{Thickness (mm)}}{0.2} \text{x Gluing factor (g/m}^2) \]

2.4 Board manufacturing
2.4. Proizvodnja ploča

Hygrothermally treated and untreated *L. philippiana* particles were employed. The percentage of wood in the outer layer was 30 % and in the inner layer 70 %. UF (50 % solid) produced by Georgia Paci Corp. with a molar ratio of 1:1.22 was applied as adhesive. This is a high emission resin, and was used without formaldehyde scavenger and without hardener in order to best determine the variation in formaldehyde release due to hygrothermal treatment. Particleboards with nominal densities of 600, 625, 650, 675 and 700 kg·m⁻³, with dimensions of 53 x 53 cm and 15 mm thickness were produced. The adhesive was applied at 8 % loading (dry basis) spraying it in a Drais FSP-80 blender. Pressing was conducted in a Bürkle LA-160 press at a temperature of 180 °C for 180 s. The maximum pressure used was conducted in a Bürkle LA-160 press at a temperature of 180 °C for 180 s. The maximum pressure used was 3 N·mm⁻² for 30 s and half pressure of 1.5 N·mm⁻² for 150 s. Two boards were produced for each nominal density, a total of 10 boards with non-treated particles along with a change in the slenderness ratio, specific surface area and available amount of adhesive. This modification of the geometry could have an effect on panel properties. As reported by Klauditz (Ginzel and Peraza, 1966) for the determination of the slenderness ratio, specific surface area of the particles and the available amount of adhesive, it is necessary to characterize the particles through the determination of basic density, thickness and length. The average weighted in accordance to the weight proportion of the particles in each sieve mesh for hygrothermally treated and untreated particles is presented in Table 1.

Table 1. Basic density, thickness, length, slenderness ratio, specific surface area and amount of available adhesive for hygrothermally treated and untreated *L. philippiana* particles

<table>
<thead>
<tr>
<th>Particles</th>
<th>Basic density</th>
<th>Thickness</th>
<th>Length</th>
<th>Slenderness Ratio</th>
<th>Specific surface area</th>
<th>Available adhesive</th>
</tr>
</thead>
<tbody>
<tr>
<td>Surface Layer</td>
<td>Untreated <em>neobrađeno</em></td>
<td>442</td>
<td>0.63</td>
<td>6.75</td>
<td>11.19</td>
<td>0.72</td>
</tr>
<tr>
<td>Outer Layer</td>
<td>Thermally treated <em>termički obradeno</em></td>
<td>428</td>
<td>0.60</td>
<td>6.48</td>
<td>10.85</td>
<td>0.78</td>
</tr>
<tr>
<td>Core Layer</td>
<td>Untreated <em>neobrađeno</em></td>
<td>442</td>
<td>0.90</td>
<td>14.55</td>
<td>16.00</td>
<td>0.50</td>
</tr>
<tr>
<td>Inner Layer</td>
<td>Thermally treated <em>termički obradeno</em></td>
<td>428</td>
<td>0.88</td>
<td>13.80</td>
<td>15.75</td>
<td>0.53</td>
</tr>
</tbody>
</table>

2.5 Mechanical properties of the boards
2.5. Mehanička svojstva ploče

Density (EN 323, 1993), MOE and MOR in static bending (EN 310, 1993), and IB (EN 319, 1993) were determined. The mechanical properties were carried out in a Metrotec HM-D/200 universal testing machine. The rate of loading was adjusted so that the maximum load was reached within 60 ± 30 s. The number of test specimens by treatment obtained in all board series for static bending (MOE and MOR) was 100 and for IB 90. The density was determined for every test specimen.

2.6 Formaldehyde release
2.6. Oslobodjeni formaldehid

This was conducted according to the perforator method (EN 120, 1992). Three samples of the average nominal board density were analysed.

The perforator value was corrected in accordance with EN 312 (2010).

2.7 Statistical analysis
2.7. Statistička analiza

A simple linear regression analysis was conducted, relating each mechanical property to the density of the respective test specimen. Additionally, an analysis of variance was conducted with a 99 % confidence interval (p <0.01) (statistical program R), to determine differences among averages of properties. In the case of formaldehyde release determinations, because of the amount of tests, no statistical analysis was done, and for the results and discussion, the average of the three tests was considered.

3 RESULTS AND DISCUSSION
3. REZULTATI I RASPRAVA

3.1 Particle geometry, specific surface area and quantity of available adhesive
3.1. Geometrija čestica, specifična površina i količina raspoloživog ljepila

The change of the particles geometry comes along with a change in the slenderness ratio, specific surface area and available amount of adhesive. This modification of the geometry could have an effect on panel properties. As reported by Klauditz (Ginzel and Peraza, 1966) for the determination of the slenderness ratio, specific surface area of the particles and the available amount of adhesive, it is necessary to characterize the particles through the determination of basic density, thickness and length. The average weighted in accordance to the weight proportion of the particles in each sieve mesh for hygrothermally treated and untreated particles is presented in Table 1.
The results of hygrothermal treatment showed a decrease in the basic density of particles. As a consequence, thermally untreated particles presented a larger quantity of available adhesive, as total grams of adhesive per surface, square meter of particles. The decrease in the basic density of particles agrees with the decrease in density reported by other investigators (Schmidt, 1982; Feist and Sell, 1987). This effect is due to the thermal degradation of wood components, such as deacetylation of hemicellulose and lignin condensation, and to the lower equilibrium moisture content, since thermally treated wood becomes hydrophobic (Boonstra et al., 2007).

Thickness and specific surface area of the particles did not change. Length, and hence slenderness ratio, decreased. This decrease was more significant in larger particles. Generally, it can be concluded that during the mechanical action of the gluing machine, it is the larger particles that undergo a reduction in size. The decrease in length of the particles may cause a change in the mechanical properties of the boards, causing a reduction of MOE and MOR, and increasing IB.

### 3.2 Mechanical properties of the boards

#### 3.2.1 Mechanical svojstva ploča

Table 2 shows the averages, standard deviation, coefficient of variation and p-values, according to the analysis of variance for boards manufactured with hygrothermally treated and untreated particles. The mechanical properties of the boards manufactured with hygrothermally treated and untreated particles present significant differences ($p<0.01$). The strength of the boards in static bending is correlated with the quality of the bonds achieved on the surface of the boards (Poblete, 2001).

The linear regression analysis registered significance at $p<0.01$. Coinciding with previous studies, when the density of the boards increased, the MOE and MOR also increased (Poblete, 2001). The regression analysis for the relationship between density and MOE

![Figure 1](image_url)

**Figure 1** Relationship between MOE and density in *L. philippiana* boards manufactured with hygrothermally treated and untreated particles

**Slika 1.** Odnos modula elastičnosti i gustoće ploča proizvedenih od hidrotermički obradenoga i neobrađenog iverja drva *L. philippiana*

**Table 2** Analysis of variance of the mean values of mechanical properties of boards manufactured with hygrothermally treated and untreated *L. philippiana* particles

<table>
<thead>
<tr>
<th>Mechanical property</th>
<th>Analysis of variance / Analiza varijance</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Untreated / Neobrađeno</td>
</tr>
<tr>
<td></td>
<td>$x$ N·mm$^{-2}$</td>
</tr>
<tr>
<td>MOE in static bending / MOE pri statičkom savijanju</td>
<td>2023</td>
</tr>
<tr>
<td>MOR in static bending / MOR pri statičkom savijanju</td>
<td>12.62</td>
</tr>
<tr>
<td>IB</td>
<td>0.24</td>
</tr>
</tbody>
</table>

$s$ – mean / srednja vrijednost; $sd$ – standard deviation / standardna devijacija; $cv$ – coefficient of variation / koeficijent varijacije; * – Significant at $p<0.01$ / signifikanto pri $p<0.01$. 
is shown in Figure 1, and the relationship between density and MOR in Figure 2.

The regression for the relationship between density and IB is presented in Figure 3. Correlations were significant at $p < 0.01$. IB is directly related with the bond quality of the particles in the core of the board (Poblete and Burgos, 2010). The results confirm the strong relationship between board density and IB, reported in many studies (Kollmann et al., 1975; Poblete, 1985; Poblete, 2001; Poblete and Burgos, 2010).

**Figure 2** Relationship between MOR and density in *L. philippiana* boards manufactured with hygrothermally treated and untreated particles

**Slika 2.** Odnos modula loma i gustoće ploča proizvedenih od hidrotermički obradjenoga i neobradjenog iverja drva *L. philippiana*

**Figure 3** Relationship between density and IB of *L. philippiana* boards manufactured with hygrothermally treated and untreated particles

**Slika 3.** Odnos gustoće i čvrstoće raslojavanja ploča proizvedenih od hidrotermički obradjenoga i neobradjenog iverja drva *L. philippiana*
According to the analysis of variance, boards manufactured with treated *L. philippiana* particles presented better mechanical properties. In the static bending, the MOE increased by 38.4 %, while the MOR increased by 37.0 %, IB increased by 48.9 %. The increase in MOE, MOR and IB is in agreement with values reported by other studies (Tomek, 1966; Kakaras and Papadopoulos, 2004; Colak et al., 2007). When determining mechanical properties of particleboards, the wood-adhesive-wood interaction is evaluated, which is influenced by the type of adhesive and the environment in which it hardens. The pH value is what determines the success or failure of the bond. In a previous work about the effect of hygrothermal treatment on the chemistry of the same *L. philippiana* particles (Crespo et al., 2013), a pH value of non-treated particles near neutral (6.7) was reported. This condition could inhibit UF curing if a hardener is not used. It should be noted that in this study a hardener was not applied. Pinto and Poblete (1992) studied *L. philippiana* particles, and acidified the environment with the addition of a catalyst, with positive effects on mechanical properties.

Colak et al. (2007) have confirmed that the pH values of the particles obtained from vaporized logs decrease, generating an optimum pH environment during the curing of UF, and increasing the IB of the boards. Crespo et al. (2013) reported for the same *L. philippiana* particles that hygrothermal treatment decreased the pH value from 6.7 to 4.8, while the quantity of volatile acids, water soluble acids and buffering capacity, increased. These changes allow for better hardening of UF and a better wood-adhesive-wood bond, which explains the increase in bending strength properties and IB in the tested boards (Roffael and Parameswaran, 1986; Roffael, 1987).

The decrease in length and slenderness ratio in thermally treated particles observed in the current study may have also influenced the increase in IB of the boards. Coincidentally, Kakaras and Papadopoulos (2004) reported that treated particles produced an increase in the proportion of smaller particles, which is associated with an increase in IB. In the current study, considering that MOE and MOR increased, the contribution of the particles size reduction to the increase of IB is less than the change in acidity.

### 3.3 Formaldehyde release

3.3. Oslobadanje formaldehida

Table 3 shows the average values of formaldehyde release (perforator value) with and without moisture content correction.

<table>
<thead>
<tr>
<th>Particles Iverje</th>
<th>Equilibrium moisture content</th>
<th>Formaldehyde release / Ostobodení formaldehid</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Ravnovesni sadržaj vode</td>
<td>Uncorrected value</td>
</tr>
<tr>
<td></td>
<td>%</td>
<td>Nekorigirana vrijednost</td>
</tr>
<tr>
<td>Untreated / Neobradeno</td>
<td>8.2</td>
<td>17.1</td>
</tr>
<tr>
<td>Thermally treated / Toplinski obradeno</td>
<td>6.2</td>
<td>9.8</td>
</tr>
</tbody>
</table>

The equilibrium moisture of the boards following acclimatization was different, and according to the standard (EN 312, 2010), the release value should be adjusted considering this parameter. Determinations with and without moisture content adjustments show that the hygrothermal treatment decreased the release of formaldehyde. Giebeler (1983) confirmed that, after the treatment of wood with moisture, heat and pressure, the most important improvement is the decrease by over 50 % in formaldehyde release of particleboards. Petersen et al. (1973) reported that boards manufactured from species with lower pH values (3.8) released less formaldehyde than boards made from species with higher pH values (4.8 and 5.5). There was no correction by moisture content of the boards reported in either of these studies. The results of Petersen et al. (1973) support that the decrease in the pH value of hygrothermally treated particles of *L. Philippiana*, which was reported by Crespo et al. (2013) in a former study, reduce the formaldehyde release of boards made from these particles.

Roffael (2012) reported that, when working with MDF manufactured with UF, an increase in delamination temperature from 140 to 175 °C results in a positive effect on formaldehyde release, which is attributed to the higher acidity of the thermo-mechanical pulp produced at 170 °C. Colak and Colakoglu (2004) claim that the acetic acid generated from the hydrolysis of non-cellulose polysaccharide acetyl groups reacts with the free formaldehyde in the resin, fixing this compound. As reported by Crespo et al. (2013) in a former study with the same particles of *L. philippiana*, hygrothermal treatment of particles increases the amount of volatile acids, water soluble acids and buffering capacity, significantly reducing the pH value. Considering the results of the current study, it can be confirmed that the reduction in formaldehyde release in boards manufactured with hygrothermally treated particles is due to the reduction in pH value, to the production of acids, and consequently to better setting of the UF.

### 4 CONCLUSIONS

4. ZAKLJUČAK

Hygrothermal treatment of *L. philippiana* wood particles resulted in lower basic density, length, slenderness ratio and quantity of available adhesive, while the specific surface area increased relative to untreated particles. The hygrothermal treatment increases mechanical properties in particleboards, while decreasing formaldehyde release. This effect is due to an acidification of the particles and an increase in the amount of...
extractable compounds. Overall, hygrothermal treatment of L. philippiana particles at 150 °C for 90 min at the pressure of 430 kPa is a good alternative to improve mechanical properties, as well as to reduce formaldehyde release in particle boards manufactured from this species.

Acknowledgments – Zähvala

This study was supported by a scholarship from the Comisión Nacional de Investigación Científica y Tecnológica (CONICYT) from Chile, and by the Secretaria de Educación Superior, Ciencia, Tecnología e Innovación (SENESCYT) from Ecuador.

6 REFERENCES

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