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THE QUALITY COMPARISON OF PARTICLEBOARDS PRODUCED FROM HEARTWOOD AND SAPWOOD OF EUROPEAN LARCH

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ABSTRACT

In this paper, the impacts of heartwood and sapwood usage on the physical, mechanical, and surface properties and formaldehyde emission of particleboard are investigated. European Larch (Larix decidua) trees are chosen as a raw material. The logs are divided into three segments: sapwood, heartwood and total wood. The highest amounts of cellulose (51,54%), and hemicelluloses (22,24%) in the sapwood, followed by total wood, and the heartwood, respectively. However, the highest amount of lignin (30,54%) was found in the heartwood. The highest extractives values are obtained from heartwood, followed by total wood, and the sapwood, respectively. While the lowest pH value (3,03) is found in heartwood, the sapwood samples provide the highest values (4,95). The highest ash (0,49%) content and amount of condensed tannin (13,89%) are extracted from heartwood, followed by total wood, and sapwood, respectively. The test panels manufactured from sapwood have the smoothest surface (7,49 μ m (R_a) 48,86 μ m (R_a), and 35,12 μ m (R_a)) and the lowest contact angles (67,8°), while the roughest surface (14,20 μ m (R) 68,05 μ m (R), and 50,02 μ m (R)) and highest contact angle (96,9°) are obtained from the panels of heartwood. The thickness swelling (19,88%) and formaldehyde emission (7,28%) values of the panels manufactured from heartwood are significantly lower than the panels manufactured from the total wood and sapwood. The highest modulus of rupture (MOR), modulus of elasticity (MOE), and internal bond (IB) values are observed on sapwood, respectively, 15,60 MPa (MOR), 2201 MPa (MOE), and 0,523 MPa (IB). These mechanical strength values (MOR, MOE, and IB) are followed by total wood, and the heartwood, respectively. Surface smoothness and wettability of the particleboards manufactured from sapwood are better than those of total wood and heartwood.

Keywords: Contact angle, formaldehyde emission, *Larix decidua*, mechanical properties, physical properties, roughness.

INTRODUCTION

Particleboard is a very popular engineered wood based panel product manufactured from wood particles and synthetic resins or other suitable binders (Baharoglu *et al.* 2013). Another definition of particleboard is given as a wood-based panel composite consisting of varying sizes and shapes of particles from lignocellulosic materials bonded together with an adhesive under high temperature and pressure (Sari *et al.* 2013). The production of particleboards has positive effects on the environment due to using wood residues (Garay *et al.* 2009)

Particleboard is a wood-based panel composite used in the manufacturing of floor underlayment, shelving, tables, counters, wall and ceiling, stair treads, bulletin boards, home constructions, kitchen worktops, interior decoration, cabinets and furniture (Nemli and Demirel 2007).

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Particleboards are mostly used in three layers. Demand for particleboard has been increasing steadily in recent years. The reason for this increase may be due to the fact that the particleboard is a low cost product and can be used in many applications.

Many studies have been performed on the effects of various factors on the quality properties of particleboard. These factors are; the usages of various raw materials such as wood bark (Blanchet *et al.* 2000), pine cone (Ayrilmis *et al.* 2009), cotton (*Gossypium hirsutum*) stalks (Nazerian *et al.* 2016), canola (*Brassica napus*) straws (Kord *et al.* 2016), and needle litter (Nemli *et al.* 2008), different parameters of the wood such as log position (Muhcu *et al.* 2015), permeability of wood (Lyman 1969), anatomical and chemical properties of wood (Baharoglu *et al.* 2013), moisture content of raw material (Baharoglu *et al.* 2012), acidity of the particles (Akyuz *et al.* 2010), wood extractives (Foster 1967), density of wood (Maloney 1977), mature and juvenile wood (Wasniewski 1989), and raw material growth region (Bardak *et al.* 2017). The parameters of the particleboard production, such as dimensions of the particles (Motted 1967), residue type and tannin content (Moubarik *et al.* 2013), paraffin application technique (Baharoglu *et al.* 2014), hardener type, urea usage and conditioning period (Atar *et al.* 2014), solid content of the adhesive and panel density (Sari *et al.* 2013), decor paper properties (Bardak *et al.* 2011), and shelling ratio (Nemli and Ozturk 2006) are investigated.

In this study, it is aimed to understand the effect of sapwood and heartwood usage on the physical (thickness swelling) and mechanical (modulus of rupture (MOR), modulus of elasticity (MOE), and internal bond (IB)) properties, surface roughness, and formaldehyde emission of particleboard panels. Besides, how chemical properties of sapwood, heartwood, and total wood affected quality properties of particleboards are investigated.

MATERIALS AND METHODS

Ten European Larch (*Larix decidua* Mill.) trees were felled from a private forest in Trabzon, Black Sea region of Turkey. The age and breast height diameter of the trees were 45 years old and 30 cm, respectively. The bark of logs was removed before chipping. The logs were divided into three segments: sapwood (diameter: 19 cm), heartwood (diameter: 11 cm), total wood (diameter: 30 cm): Segments were chipped using a ring type flaker. The chips were then reduced into smaller particles using a hammer mill. The particles were dried to 3% moisture content in a dryer. The dried particles were classified into two sizes using a 3,0-1,5-0,8 mm openings vibrating screen for the core and face layers. Based on oven dry weight of particle weight, 9% and 11% urea formaldehyde resin was applied for core and face particles, respectively. The ratio of the face thickness to the total thickness of a panel known as the shelling ratio was 0,35 for all samples. Ammonium sulphate (concentration: 20%) was used as a hardener during the blending process by about 1% based on the solid amount of adhesive. No wax or any other additives were used for the panel manufacture. The specifications of the use urea formaldehyde are presented in Table 1.

| Solid Content | 65% |
|---------------|--------|
| Gel Time | 94 s |
| Viscosity | 178 cp |

 Table 1: The specifications of urea formaldehyde.

Mats formed manually in a frame with a size of 550 x 550 mm were hot pressed at a temperature of 150 °C using a pressure of 2,5 MPa for 6 min. All panels were pressed to a nominal thickness of 12,0 mm and an average target density of 0,70 g/cm³. A total of 9 experimental panels, 3 panels for each type of particleboard, were produced. The experimental design of the study is illustrated in Table 2.

| Panel Types | Wood Types | |
|-------------|------------|--|
| А | Heartwood | |
| В | Sapwood | |
| С | Total wood | |

Table 2: The experimental design of the study.

Until the panels reached equilibrium moisture content, they were kept in a conditioned room with a relative humidity of 65% and a temperature of 20°C. Mechanical properties- MOR (the samples with dimensions of 300 mm x 50 mm x 12 mm) and MOE (the samples with dimensions of 300 mm x 50 mm x 12 mm) (EN 310 1993), and IB (the samples with dimensions of 50 mm x 50 mm x 12 mm) (EN 319 1993)), and physical property- thickness swelling (TS) (the samples with dimensions of 50 mm x 50 mm x 50 mm x 12 mm) (EN 317 1993) of particleboards were determined according to European standards. Thirty samples were replicated and used for physical (TS) and mechanical (MOR, MOE, and IB) tests.

Determination of the chemical properties of the woods and preparation of the test specimens were carried out according to TAPPI standard TAPPI T M-45 (1992) standard. Alcohol-benzene TAPPI T 204 cm-97 (1997), hot and cold water (2 g air-dried sample) TAPPI T 207 om-88 (1988) and dilute alkali (1% NaOH) solubility's (2 gair-dried sample) TAPPI T 212 om-98 (1998), lignin content (1 g air-dried sample) TAPPI T 222 om-02 (2002) and amount of ash (4-5 g air-dried sample) TAPPI T 211 (1993) were determined according to the cited standards. Holocellulose (5 g air-dried sample exposed by alcohol-benzene extraction) and cellulose contents (2 g air-dried sample exposed by alcohol-benzene extraction) were determined by chlorite and nitric acic methods (Wise and Karz 1962). The acidity was measured in an extract solution made by 3 g wood flour added to 100 ml water and boiled for 30 min (Prasetya 1989). Amount of condensed tannins was determined according to the method developed by Tisler and friends (Tisler *et al.* 1986). Three samples were replicated and used for determination of chemical properties.

The samples with the dimensions of 50 mm \times 50 mm \times 12 mm used for surface roughness tests were sanded with a sequence of 100 and 150 grit sand papers. The samples to be used in surface roughness were conditioned in a climate chamber at 20°C and 65% RH. A Mitutoyo SJ-301surface roughness tester, a stylus type profilometer, was employed for the surface roughness tests. Three roughness parameters characterized by ISO 4287 (1997) standard, which are average roughness (Ra), mean peak-to-valley height (Rz), and maximum peak-to-valley height (Ry), were considered to evaluate the surface characteristics of the panels. A total of 60 roughness measurements were performed, with 4 measurements for each type of particleboard of 15 samples. Two of the measurements were performed parallel to the sand marks while the other two were performed perpendicular to the sand marks.

The wetting behavior of the particleboard samples conditioned at 65% RH and 20°C was characterized by the CA method (goniometer technique). Contact angle (CA) measurements were performed using a CAM 101 Optical Contact Angle Meter (KSV Instruments Ltd., Helsinki), equipped with a video camera. Using the sessile drop method, which is the most widely used procedure, the contact angle was determined simply by aligning tangent with the sessile drop profile at the point of contact with the solid surface. An imaging system was used to measure contact angle and droplet shape for the tested surfaces of the samples. The image of the liquid drop was captured by a video camera, and the CA was measured by the digital image analysis software. After 5-mL droplet of the distilled water was placed on the sample surface, CAs from the images were measured for 5 s. A total of 30 contact angle measurements, two from each of 15 samples, were performed for each type of panel.

Three samples with the dimensions of 20 mm x 20 mm x 12 mm were randomly taken from each type of particleboard for the determination of formaldehyde emission (FE) by the perforator method based on EN 120-1 (1994) standard. In this method, the free formaldehyde in the board is determined by extraction. According to the standards, free formaldehyde is fed into the distilled water from the board samples boiled in toluene and the amount of formaldehyde in the aqueous solution is determined photometrically and proportioned to the weight of the full dry board (Bardak 2014).

One-way analysis of variance, ANOVA, was conducted ($p \le 0.05$) to evaluate the effect of sapwood, heartwood usage on the quality properties of the panels. Significant differences between the mean values of the

panel types were determined using Newman-Keuls's test.

RESULTS AND DISCUSSION

Chemical properties

The highest amounts of cellulose (49,12%), hemicelluloses (22,24%) were found in the sapwood, followed by total wood, and the heartwood, respectively. However, the highest amount of lignin amounts (30,54%) was found in the heartwood. The average values of chemical properties of furnishes are presented in Table 3. According to the results of statistical analysis, all the chemical properties of the samples were significantly affected by the furnish type. The highest solubility values were obtained from heartwood, followed by total wood, and the sapwood, respectively. While the lowest pH of extract value (3,03) was found in heartwood, the sapwood provided the highest pH of extract values (4,95). The highest ash content (0,49%), and amount of condensed tannin (13,89%) were found in heartwood, followed by total wood, and sapwood, respectively. Generally, the heartwood contains higher amounts of extracts than sapwood (Ors and Keskin 2001, Sivrikaya 2008). In addition, holocellulose and cellulose amounts in sapwood of pinus nigra, fir and oak were higher than the one in the heartwood. However, lignin amount, cold and hot water, alcohol and 1% NaOH solubilities in heartwood of pinus nigra, fir and oak were found to be higher than the one in the sapwood (Atac 2009). In the study conducted by Campbell et al. 1990, it was found that the extractive content of heartwood was significantly higher than sapwood in *latifolia* (3,30% vs. 2,03%) and *murrayana* and the alpha-cellulose content of sapwood was also significantly higher than that of heartwood in *latifolia* (49,38% vs. 46,02%) and *murrayana* (49,52% vs. 44,17%) (Campbell et al. 1990). Based on these findings, we can safely express that our results agree with the previous studies.

| Chemical properties Wood types | | | |
|---------------------------------------|------------|------------|------------|
| | Total wood | Heartwood | Sapwood |
| pH of extract | 4,58 | 3,03 | 4,95 |
| | (0,03) (a) | (0,08) (b) | (0,04) (c) |
| Solubility in dilute alkali (1% NaOH) | 15,56 | 25,51 | 12,33 |
| (%) | (0,21) (a) | (0,43) (b) | (0,32) (c) |
| Extractives (%) | 5,68 | 8,73 | 5,02 |
| | (0,14) (a) | (0,25) (b) | (0,16) (c) |
| Solubility in cold water (%) | 2,54 | 4,87 | 2,13 |
| | (0,08) (a) | (0,05) (b) | (0,03) (c) |
| Solubility in hot water (%) | 5,45 | 7,89 | 5,02 |
| | (0,09) (a) | (0,15) (b) | (0,11)(c) |
| Cellulose (%) | 49,12 | 45,02 | 51,54 |
| | (0,17) (a) | (0,10) (b) | (0,06)(c) |
| Hemicellulose (%) | 19,02 | 17,59 | 22,24 |
| | (0,13) (a) | (0,22) (b) | (0,34) (c) |
| Lignin (%) | 27,31 | 30,54 | 21,66 |
| | (0,07) (a) | (0,12) (b) | (0,18)(c) |
| Ash (%) | 0,34 | 0,49 | 0,22 |
| | (0,02) (a) | (0,05) (b) | (0,04) (c) |
| Amount of condensed tannin (%) | 10,78 | 13,89 | 8,03 |
| | (0,19) (a) | (0,15) (b) | (0,17) (c) |

| Table 3: Chemical properties of total wo | ood, heartwood and sapwood. |
|--|-----------------------------|
|--|-----------------------------|

Note: Numbers in the parenthesis are standard deviations. Different letters in the same line represent statistical differences at 95% confidence level.

Surface properties

The surface roughness and contact angles of the test panels are presented in Table 4. The test panels manufactured from sapwood had the smoothest surface (7,49 μ m (R_a) 48,86 μ m (R_y), and 35,12 μ m (R_z)) and the lowest contact angles (67,8°), while the roughest surface (14,20 μ m (R_a) 68,05 μ m (R_y), and 50,02 μ m (R_z)) and highest contact angle (96,9°) were obtained from the panels manufactured from heartwood. This may be due to high amount of extractives (Table 2) in the heartwood. Extractives and ash negatively affect the surface quality of particleboard. These affect negatively the bonding of wood. Poor bonding increases the surface roughness of particles (Lehman and Geimer 1974, Muhcu *et al.* 2015). One of the most important factors affecting the contact angle results is surface roughness. If the contact angle value measured on the surface of the material generally exceeds 80°, the contact angle value increases as the surface roughness increases (Buscher *et al.* 1984, Onda *et al.* 1996).

| Panel Types | R _a (μm) | R _y (μm) | R _z (μm) | СА |
|-------------|---------------------|----------------------------|----------------------------|------------------|
| | | | | Degree(°) at 5 s |
| Hearthwood | 14,20 | 68,05 | 50,02 | 96,9 |
| | (0,38) a | (0,79) a | (0,98) a | (0,99) a |
| Sapwood | 7,49 | 48,86 | 35,12 | 67,8 |
| _ | (0,22) b | (0,59) b | (0,77) b | (1,30) b |
| Total wood | 10,88 | 54,98 | 40,81 | 82,9 |
| | (0,23) c | (0,69) c | (0,79) c | (1,32) c |

Table 4: Average surface roughness and contact angles.

Note: Numbers in the parenthesis are standard deviations. Different letters in the same column represent statistical differences at 95% confidence level.

Physical and mechanical properties and formaldehyde emission

The physical and mechanical properties and formaldehyde content of particleboards are presented in Table 5.

| Panel Types | MOR (MPa) | MOE (MPa) | IB (MPa) | TS (%) | FE (mg CH ₂ O) |
|----------------|--------------|--------------|-------------|-----------|---------------------------------|
| Hearthwood | 10,10 | 1369 | 0,225 | 19,88 | 7,28 |
| | (0,38) a | (98) a | (0,030) a | (0,23) a | (0,02) a |
| Sapwood | 15,60 | 2201 | 0,523 | 33,85 | 9,05 |
| | (0,43) b | (61) b | (0,019) b | (0,29) b | (0,06) b |
| Total wood | 12,74 | 1723 | 0,329 | 26,77 | 8,51 |
| | (0,25) c | (85) c | (0,013) c | (0,48) c | (0,03) c |

Table 5: Mechanical properties, water resistance, and formaldehyde content of test panels.

Note: Numbers in the parenthesis are standard deviations. Different letters in the same column represent statistical differences at 95% confidence level.

Maderas. Ciencia y tecnología 21(4): 511 - 520, 2019

The 12,5 MPa and 13 MPa are the minimum requirements of MOR of particleboard for general usage and interior fitments (including furniture), respectively, while the minimum MOE for interior fitment is 1800 MPa, based on EN 312 (2005) standard. The minimal requirements of IB strength for general purpose and furniture manufacturing are 0,28 MPa and 0,40 MPa, respectively. The panels made from sapwood had the required levels of MOR, MOE and IB for general purposes and furniture manufacturing. The particleboards made from total wood met the required levels of MOR, MOE and IB only for general purposes. Panels made from heartwood did not have the required levels of mechanical strength properties. According to the test results, the test panels did not have the required level of TS property according to EN 312 (2005) standard due to no usage of water-repellent agents, such as paraffin, in the manufacturing process. The maximum permissible formalde-hyde content for E_1 quality particleboard is 8 mg $CH_2O/100$ g dry particleboard sample (EN 120-1 1994). The panels made from heartwood met the required level of FE for E_1 quality.

The thickness swelling and formaldehyde emission values of the panels manufactured from heartwood are significantly lower than the panels manufactured from the total wood and sapwood. The highest mechanical strength values (modulus of rupture, modulus of elasticity and internal bond strength) were obtained from sapwood, followed by total wood, and the heartwood, respectively. Density of the heartwood is higher than that of sapwood. When the wood at high density is chipped, the particles are rough and thick. Rough and thick particles absorb the adhesive. Therefore, there is not enough adhesive on the particle surfaces. So that the mechanical strength properties decrease (Lyman 1969). The pH of the extract should be 4-5 for good adhesion and blending operation. The pH of extract of the heartwood was found as 3,03 (Table 3). Low pH of the extract causes precuring of adhesive before hot pressing application, and decreases the strength properties (Maloney 1977). The skeleton of the wood is cellulose. Cellulose and lignin are wood components that give strength to the wood (Baharoglu *et al.* 2013). In this study, sapwood had higher amounts of cellulose. For these reasons, panels from sapwood had higher mechanical strength properties.

The water is connected to the OH groups of cellulose and hemicellulose (Baharoglu *et al.* 2013). Heartwood had higher lignin and amount of condensed tannin. Lignin and consended tannin have hydrophobe structure. However, hemicellulose has hydrophilic structure. Hemicelluloses absorb a greater amount of water than cellulose. (Sari *et al.* 2012). Particleboards made from sapwood and total wood had higher thickness swelling values due to a higher amount of hemicelluloses. The adhesive has to wet, flow and penetrate the cellular structure of wood in order to establish intimate contact between molecules of wood and adhesive. Heartwood had higher amounts of extractives than those of total wood and sapwood. The extractives, such as wax, phenolic compounds, liphophilic, and condensed tannin, positively affect the dimensional stability and formaldehyde emission. Tannin and phenolic extractives fix the formaldehyde as a formaldehyde scavenger. (Akbulut 1995). For these reasons, the usage of heartwood reduces the amount of thickness swelling and formaldehyde content. The positive effects of these extractives on the resistance to water (Marshall *et al.* 1974, Sari *et al.* 2012) and formaldehyde emission (Sari *et al.* 2012) were mentioned in a previous work.

High amount of extractives decreases the wettability of wood particles. Extractives cause poorer mechanical properties due to breaking down the adhesive linkage. They negatively affect adhesive bonding and adhesion. Decreasing contact angle and roughness increase the adhesion and wettability of the wood particles. The panels from sapwood had smoother surfaces and lower contact angle (Table 4). Increasing adhesion causes high adhesive bonds and mechanical properties. Some extractives are translated to the wood surfaces by the water during the drying and pressing processes. When the water is vaporized, these extractives stayed in solid form on the particle surfaces. This condition negatively affects quality of gluing and adhesion (Wasniewski 1989, Christiansen 1990, Ayrilmis and Winandy 2009, Huang et al. 2011). The ash content of heartwood is significantly higher than those of sapwood and total wood (Table 3). The ash has lower wettability than the wood, which decreases the bond quality. A non-polar surface of the ash makes it hard for the water-based adhesive to wet and penetrates into the cellular structure, resulting in the poor adhesion between the resin and wood (Muhcu et al. 2015). This is the other reason for the lower strength properties of the particleboards manufactured from heartwood. In a previous study (Pan et al. 2007), MOR, MOE and IB of the particleboard manufactured from saline eucalyptus heartwood were found to be 13,6 MPa; 1564,2 MPa and 1,31 MPa, respectively. 2 hours and 24 hours thickness of physical properties were 31,26% and 38,28%, respectively. In the same study, the thickness swelling from 2 hours and 24 hours were found to be 31,26 and 38,28%, respectively. In another study (Duchesne et al. 2016), it was found that heartwood proportion did not have any significant effect on MOE and MOR. In a 1994 study, it was found that particleboards made of sapwood especially had a higher bending strength (Roffael and Dix 1994).

CONCLUSIONS

In this work, it is aimed to understand the effect of sapwood and heartwood usage on the quality features of particleboards. Thickness swelling, mechanical and surface properties, wettability, and formaldehyde emission of particleboards were significantly affected by the usage of sapwood and heartwood. Nowadays, there is a shortage of raw materials in particleboard production. The results revealed that European Larch trees can be used to produce particleboards in the case of the addition of water-repellent agent. The best mechanical strength, surface smoothness and wettability values were obtained from the particleboards manufactured from sapwood. The thickness swelling and formaldehyde emission of particleboards are generally evaluated in general use and furniture production. When we consider the mechanical properties of the work we have done, it is determined that the particleboards made from sapwood are suitable for furniture and general use, but the particleboards made from heartwood do not fit into any use. Therefore, it may not be appropriate to use the heartwood alone.

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