Roughness, Wettability, and Morphological Properties of Impregnated and Densified Wood Materials

Hüseyin Pelit,[a,*] and Umuthan Arısüt [b]

The effects of pre-impregnation on surface roughness, wettability, and morphological structure of densified aspen and fir wood were investigated. Wood specimens were impregnated with paraffin, linseed oil, and styrene after pre-vacuum treatment. The impregnated specimens were densified using compression ratios of 20% and 40% at 120 °C, 150 °C, and 180 °C. The roughness decreased and the contact angle increased in all impregnated specimens (undensified and densified). Compared to paraffin and linseed oil-impregnated specimens, lower roughness and higher contact angle were found in styrene-impregnated specimens. After densification, the roughness and wettability of the wood specimens decreased. More successful results (lower roughness and higher contact angle) were generally obtained in specimens densified with high compression ratio (40%). In both untreated and impregnated specimens, the contact angle increased with increasing compression temperature. While the effect of compression temperature on the roughness of the fir specimens was not significant, the roughness of the aspen specimens (especially styrene-impregnated) decreased with the increase in temperature. Scanning electron microscopy observations indicated that impregnation agents (especially styrene) clung to wood cell walls and partially or completely filled the lumens. This was positively correlated with the determined roughness and wettability. Pre-impregnation facilitated wood densification without significant cell deformations.

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INTRODUCTION

Glue or paint/varnish materials are applied to wood's surface in gluing and finishing processes; thus the physical and chemical properties of the wood surfaces used are important in terms of criteria for success. Wettability is one of the surface characteristics of wood that considerably affects the gluing/coating process. The wettability of wood is the ability of a liquid such as water, glue, paint, dye to come into close contact with the wood surface. Therefore, it plays a critical role in the adhesion between the wood material and the adhesive/coating (Bekhta and Krystofiak 2016). Wettability of wood is mostly determined by measuring the contact angle, which is inversely proportional to wettability (Ünsal *et al.* 2011). The contact angle is also known as equilibrium contact angle when wettability reaches equilibrium (Guancheng 2018). Roughness, another important surface characteristic of wood, is a basic factor in determining the surface quality of wood and wood-based materials. Roughness greatly

affects not only the quality of the final product, but also processes such as bonding and coating (Richter *et al.* 1995; Hiziroğlu *et al*. 2014; Söğütlü *et al.* 2016; Salca *et al.* 2017; Söğütlü 2017). High roughness prevents homogeneous distribution of glue or paint/varnish solutions on the wood surface, increases consumption, reduces adhesion strength, and causes increased costs (Faust and Rice 1986; Richter *et al.* 1995; Candan *et al.* 2010; Bekhta *et al.* 2017). The roughness or wettability characteristics of wood surfaces are separately affected by many factors, such as the wood anatomical properties, density, moisture, compression parameters, and properties of the machining process (Kılıç *et al*. 2006; Diouf *et al.* 2011; Ünsal *et al.* 2011; Arruda and Del Menezzi 2013; Sofuoğlu and Kurtoğlu 2015; Bekhta and Krystofiak 2016; Bekhta *et al.* 2017; İlçe 2018; Pinkowski *et al.* 2018). In addition, surface roughness significantly affects the equilibrium and non-equilibrium contact angles of solid materials (Shupe *et al.* 2001; Büyüksarı *et al.* 2011; Akgül *et al.* 2012; Papp and Csiha 2017; Huang *et al.* 2019; Luo *et al.* 2020).

Several mechanical properties, such as bending performances, hardness, and abrasion resistance of wood are correlated with its density, and therefore density is an important parameter for the selection of suitable usage areas of wood. High-density wood species are often preferred because of their good mechanical properties, especially for structural applications where strength is important. However, high-density wood species are difficult to obtain due to important factors such as high cost, limited availability, and transportation problems. The density of low-density softwood species can be increased through modification, and this process is called densification. As a result of densification, new wood products with high strength properties are developed, and thus low-density species are better utilized (Sandberg *et al.* 2013; Song *et al.* 2018; Fang *et al.* 2019; Laskowska 2020; Sotayo *et al.* 2020; Cabral *et al.* 2022). Densification also improves the surface quality properties of wood such as roughness, contact angle, and gloss (Candan *et al.* 2010; Diouf *et al.* 2011; Kutnar *et al.* 2012; Arruda and Del Menezzi 2013; Büyüksarı 2013; Bekhta *et al.* 2014a, 2017; Şenol and Budakçı 2019). Densification is often achieved by compressing the wood in the transverse direction and under suitable heat and/or steam conditions (Seborg *et al.* 1956; Inoue *et al.* 1993a; Navi and Girardet 2000; Kamke and Sizemore 2008; Sandberg *et al.* 2021; Cabral *et al.* 2022). Additionally, wood material is densified by decreasing the cavity structure of wood after the impregnation of various substances into the lumens or by combining compression and impregnation (Seborg *et al.* 1962; Kollmann *et al.* 1975; Inoue *et al.* 1993b; Gabrielli and Kamke 2010; Pelit and Emiroglu 2021).

One of the most important problems of densified wood, whose void volume is reduced because of compression, is that it is unstable in terms of dimensions. Compressed wood almost completely returns to its initial dimensions when exposed to liquid water or high relative humidity, and this undesirable phenomenon is known as set-recovery (Morsing 2000; Navi and Heger 2004; Rautkari *et al.* 2010; Kutnar and Kamke 2012). Another important problem with compressed wood is the various deformations that occur in the cell wall structure such as collapses, breakage, and cracking (Doğu *et al.* 2010; Ahmed *et al.* 2013; Budakçı *et al.* 2016; Bekhta *et al.* 2017). The amount and type of deformation varies depending on the compression method and process parameters, and deformations generally have a negative effect on the physical or mechanical properties of wood (Navi and Girardet 2000; Kutnar *et al.* 2009; Pelit *et al.* 2018).

In previous studies that may contribute to the results of this study, the effects of pre-impregnation with paraffin, linseed oil, and styrene on some physical and mechanical properties of densified aspen and fir woods were analyzed (Pelit and Emiroglu 2020, 2021). The results of the studies showed that the hygroscopicity of the densified wood specimens (especially styrene pre-impregnated) decreased and the dimensional stability was almost completely achieved. In addition, the hardness and mechanical strength properties of these wood specimens increased greatly. Some untested and needed important properties of these impregnated and densified wood products with newly developed and superior properties were analyzed in the present study. Thus, the usage areas of modified wood products may be determined more clearly. The objective of this study was to determine the effects of pre-impregnation processes on surface roughness, wettability, as well as morphological structure of densified aspen and fir wood.

EXPERIMENTAL

Materials

Wood material

In this study, aspen (*Populus tremula* L.) and fir (*Abies bornmuelleriana* Mattf.) woods, which have relatively low densities (aspen: 0.37 g/cm^3, fir: 0.45 g/cm^3) were used. Wood materials with average dimensions of 3500 × 230 × 100 mm^3 (L × R × T) and with moisture content of 18% to 20% were selected randomly from a timber company in Düzce, Turkey. Aspen and fir specimens were rough cut from sapwood of boards. The specimens were subjected to natural drying to approximately 12% moisture content, and then cut to the dimensions of 300 × 20 mm^2 (longitudinal direction × tangential direction) and three different thicknesses 20 mm (for undensified specimens), 25 mm, and 33.3 mm (radial direction). The test specimens were prepared in a number sufficient to accommodate eight repetitions (n = 8) for each variable in the study. Before impregnation, the specimens were kept in a drying oven until they reached a stable weight at 70 °C and then their weights were recorded.

Impregnation

Paraffin, linseed oil, and styrene, which have high water repellency, were used as impregnation agents. To prepare the impregnation solutions: a) the paraffin in solid form was melted by heating; b) the linseed oil was thinned *via* addition of 1/1 synthetic thinner; c) the styrene monomer was mixed with 1% catalyst (methyl ethyl ketone peroxide) to accelerate the polymerization. Impregnation of wood specimens was completed using a cylindrical tank assembly in accordance with ASTM D1413-99 (1999). A pre-vacuum equivalent pressure of 760 mm Hg was applied to the specimens for 60 min. Afterwards, the specimens were kept in impregnation solutions at atmospheric pressure for 24 h (Fig. 1). To prevent the paraffin from solidifying again, the paraffin solutions were held at 80 °C for 24 h. After impregnation, the paraffin- and linseed oil-impregnated specimens were kept at a RH (relative humidity) of 65 ± 3% and temperature of 20 ± 2 °C. Styrene-impregnated specimens were wrapped in aluminum foil and then incubated in an oven at 90 °C for 2 h to initiate the polymerization process (chain-reactions of monomers). Afterwards, the densification process was started immediately to complete the polymerization.

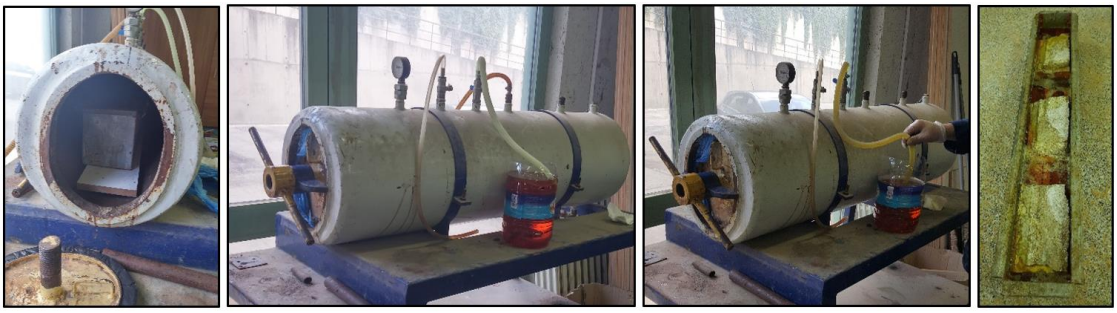

Fig. 1. Impregnation process of wood specimens

Densification

The impregnated specimens were densified using special metal molds in a hydraulic test press. Densification was completed at three different temperatures (120, 150, and 180 °C) and two different compression extents (20% and 40%). Channels 10 mm in depth and 20 mm wide were opened in the metal molds used for densification. The 25-mm- and 33.3-mm-thick specimens were placed separately inside the channels in molds. The molds were then placed in the press machine (SSP-180 T; Cemilusta Wood Working Machinery Ind. Inc., Istanbul, Turkey), where the lower and upper plates were heated at target temperatures and pre-heated for 10 min. Afterwards, the specimens were compressed in the radial direction with a loading speed of 60 mm / min. To achieve the targeted thickness (20 mm), the load was maintained until the metal molds came into contact with each other. The compressed specimens were kept under pressure for 20 min and then were removed from the press together with the molds and cooled to room temperature under an average pressure of 0.5 MPa to minimize the spring-back effect. Densification processes were also described in detail in a previous study (Pelit and Emiroglu 2021).

After the densification, specimens remained at RH 65 ± 3% and 20 ± 2 °C until they reached a stable weight. The impregnated and densified specimens were then cut into smaller specimens (only in the longitudinal direction) for surface roughness and contact angle tests.

Methods

Determination of retention

The retention values of wood specimens impregnated with water repellents were determined using Eq. 1,

$$\text{Retention (kg/m}^3\text{)} = (G \times C) / V \times 10 \quad (1)$$

where G is the amount (g) of water repellent absorbed by the specimens, C is the concentration (%) of the water repellent solution, and V is the volume (cm^3) of the wood specimens.

Roughness measurement

Roughness measurements of wood specimens were performed using a Time TR200 instrument (Time Group Inc., Beijing, China) according to ISO 4287 (1997). The parameters R_a, R_y, and R_z are generally used in the numerical expression of wood surface roughness. The R_a parameter was measured to evaluate the roughness values of the specimens. The R_a parameter is the arithmetic mean of the absolute values of the profile departures. After setting the roughness instrument to a measuring speed of 15 mm/min,

a measuring step length of 2.5 mm, measurements were made perpendicular to the fibers and on the tangential section.

Contact angle measurement

Contact angle (or lower wettability) measurements were made with the sessile drop method using goniometer technique in the Attension Theta Flex equipment (Biolin Scientific Group, Gothenburg, Sweden). After the test specimen was placed on the measuring table, the equipment camera was focused on the specimen surface. The software (OneAttention, Biolin Scientific Group, v4.1.2 (r9576), Gothenburg, Sweden) allows the desired amount of liquid to be dripped onto the specimen. Distilled water, which was chosen as the test liquid in the study, was used in a standard amount of 5 μL. The contact angle value is determined by calculating the angle formed between the right and left contact point of the droplet and the tangent line to the approximate droplet radius on the digital image. For each specimen, measurements were made for 60 s and measurements taken every 1 s. The contact angle value was determined by averaging the values obtained at the end of 60 s.

Scanning electron microscopy (SEM) analysis

Changes in the morphological structure of the impregnated and densified wood specimens were investigated using the FEI Quanta FEG 250 scanning electron microscope (Dawson Creek Drive Hillsboro, OR, USA). Small wood specimens with the dimensions of 8 × 8 × 8 mm^3 were prepared for SEM analysis, and the wood specimen surfaces were sputter coated with gold (Denton Vacuum, LLC, Moorestown, NJ, USA). Microscopic images at different magnifications were obtained from cross-sectional segments of the aspen and fir wood specimens.

Statistical analysis

The MSTAT-C 2.1 software (Michigan State University, East Lansing, MI, USA) was used for statistical analysis. Analysis of variance (ANOVA) tests were conducted to determine the effects of impregnation and densification on the roughness and contact angle of the aspen and fir wood at the 0.05 significance level. Duncan's one-way tests were conducted for comparisons between the means of the measured properties for each wood species.

RESULTS AND DISCUSSION

The retentions determined for aspen and fir wood specimens after impregnation processes are given in Table 1. The results showed that the highest retention value for both wood species was determined in specimens impregnated with styrene and the lowest in paraffin-impregnated specimens.

Table 1. Retention Values

Wood Species	Retention (kg/m^3)		
	Paraffin	Linseed Oil	Styrene
Aspen	155.3 (10.0)	230.2 (21.4)	311.6 (21.7)
Fir	148.1 (9.5)	230.1 (20.4)	300.4 (18.2)

Values in parenthesis are standard deviations

According to ANOVA results, the effect of impregnation agents and densification conditions on surface roughness and contact angle of aspen and fir woods was statistically significant ($p \leq 0.05$).

The highest roughness average regarding impregnation agent was in the untreated specimens (2.862 μm for aspen and 2.101 μm for fir), while the lowest was obtained in the styrene-impregnated specimens (1.638 μm for aspen and 1.371 μm for fir) (Table 2). Surface roughness values decreased in all impregnated specimens (undensified and densified) for both wood species (Fig. 2). Lower roughness values were measured in paraffin-impregnated specimens compared to linseed oil-impregnated specimens. However, the most successful (lower roughness) results in both wood species were determined in styrene-impregnated specimens. It is well known that wood material has a porous structure. It can be said that partial or complete filling of the pores and/or cavities of the wood material by the impregnation agents (especially *in-situ* polymerization of the styrene monomer) is effective in decreasing the roughness value of the specimens. When the SEM images of the impregnated wood specimens (especially styrene-impregnated) were examined, it was observed that the cell lumens were partially or completely filled.

Table 2. Duncan's Test Results for Means of Roughness and Contact Angle Values

Factor	Aspen				Fir			
	Roughness (μm)		Contact Angle (°)		Roughness (μm)		Contact Angle (°)	
	Mean	SG	Mean	SG	Mean	SG	Mean	SG
Impregnation Agent								
Untreated	2.862	a	74.82	d	2.101	a	75.31	c
Paraffin	2.064	c	95.22	b	1.600	c	88.77	b
Linseed oil	2.249	b	90.75	c	1.838	b	89.06	b
Styrene	1.638	d	97.99	a	1.371	d	97.25	a
Densification								
Undensified	3.214	a	83.72	d	2.595	a	82.60	d
120 °C / 20%	2.488	b	87.78	c	1.661	bc	84.35	d
120 °C / 40%	2.066	c	88.50	bc	1.479	e	86.58	c
150 °C / 20%	2.012	c	89.80	b	1.720	b	87.18	bc
150 °C / 40%	1.841	d	88.90	bc	1.536	de	88.59	b
180 °C / 20%	1.961	cd	93.94	a	1.605	cd	92.53	a
180 °C / 40%	1.839	d	95.24	a	1.496	e	91.35	a

SG: statistical group (different letters denote significant differences)

Regarding densification condition, the maximum roughness average for both wood species was determined in the undensified specimens (3.214 for aspen and 2.595 μm for fir). The minimum roughness average for aspen wood was determined in the specimens compressed at the ratio of 40% at 150 and 180 °C (1.841 and 1.839 μm), and for fir wood in specimens compressed at the ratio of 40% at 120 and 180 °C (1.479 and 1.496 μm) (Table 2). The roughness values of all densified specimens (untreated and impregnated) decreased depending on the compression ratio and compression temperature (Fig. 2). Lower roughness values were obtained in the specimens densified with high

compression ratio (40%). After densification, it can be said that the decrease in the void volume of the wood material, depending on the increase in the compression ratio, has an effect on the results. In addition, because compressed wood exhibits a more homogeneous structure compared to natural wood, it may have an effect on the roughness results. In a previous study, it was stated that the surface roughness of the densified wood decreases with the increase in the compression ratio, and this may be due to the tighter structure of the material as a result of the decrease in the intercellular spaces and the increase in density (Pelit *et al.* 2015). Different study results also show that the surface roughness of densified solid wood or veneers decreases depending on the applied densification parameters (Diouf *et al.* 2011; Santos *et al.* 2012; Arruda and Del Menezzi, 2013; İmirzi *et al.* 2014; Bekhta *et al.* 2014b). In addition, it was stated that the densified wood has a more homogeneous structure depending on the compression ratio (applied pressure) (Blomberg *et al.* 2005).

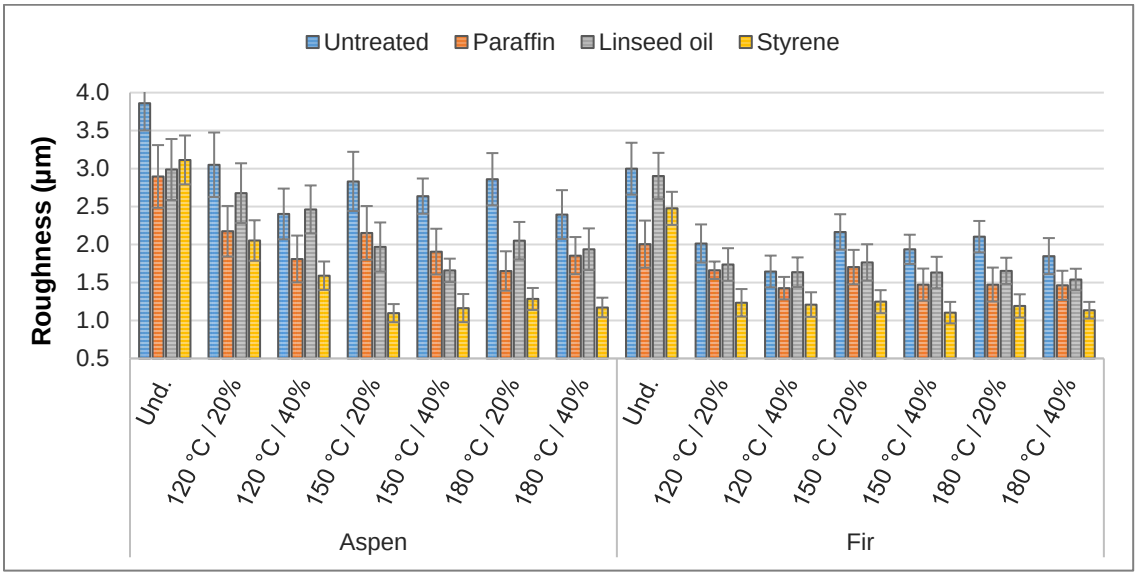

Fig. 2. Roughness values of impregnated and densified wood specimens

In terms of compression temperature, temperature change had no significant effect on roughness values for fir wood specimens. In contrast, surface roughness values tended to decrease in aspen wood specimens, especially in styrene-impregnated specimens, depending on the increase in compression temperature (Fig. 2). This can be explained by the lesser occurrence of cell deformations (fractures, cracking, *etc.*) that may occur in specimens compressed at high temperatures. In previous studies, it has been stated that the amount and size of the deformation in the cell structure occur less in wood materials that are compressed at higher temperatures (Kutnar and Šernek 2007; Budakçı *et al.* 2016). The SEM analyses within the scope of this study also confirmed this situation. In previous studies examining of some properties of densified wood veneers, it was determined that the surface roughness decreased with increasing compression temperature and pressure (Candan *et al.* 2010; Bekhta *et al.* 2017). It was also stated that the increase in compression temperature has a significantly positive effect to reduce the surface roughness of thermo-mechanically densified wood veneers (Arruda and Del Menezzi 2013). After densification, depending on the compression ratio and compression temperature, the roughness value of paraffin, linseed oil, and styrene-impregnated specimens decreased up to 57%, 57%, and 72% for aspen wood, and up to 52%, 49%, and 63% for fir wood, respectively.

With respect to impregnation agent, the highest contact angle average was determined in the styrene-impregnated specimens (97.99° for aspen and 97.25° for fir) and the lowest was found in the untreated specimens (74.82° for aspen and 75.31° for fir) (Table 2). After the impregnation processes, the contact angle values of both undensified and densified aspen and fir specimens increased. The contact angle of paraffin and linseed oil-impregnated specimens were generally similar (Fig. 3). Styrene is a conjugated system that can polymerize quickly and effectively compared to the non-conjugated paraffin and linseed oil. The SEM observations support the results. After impregnation, especially as a result of *in-situ* polymerization of styrene, most of the lumens were filled and a hydrophobic film was formed in the wood structure. Furthermore, it can be said that the increase in the contact angle values of the impregnated specimens is due to the decrease in the roughness values of these specimens and the hydrophobic (water repellent) nature of the impregnation materials used. Huang *et al.* (2019) reported that the density increase in poplar veneers laminated after impregnation with phenolic resins has a reducing effect on the surface roughness and there is also a negative correlation between contact angle and surface roughness. Poplar wood treated with monomers synthesized from styrene and methyl methacrylate has a higher contact angle than untreated poplar (Li *et al.* 2013). In wood materials treated with linseed oil and tung oil, the contact angle increased and more hydrophobic surfaces were obtained (Arminger *et al.* 2020). Compared to untreated (66°) and paraffin emulsion treated wood (94°), the wood treated by paraffin/acrylate compound emulsion showed the highest water contact angle (133°) and better dimensional stability (Jiang *et al.* 2020).

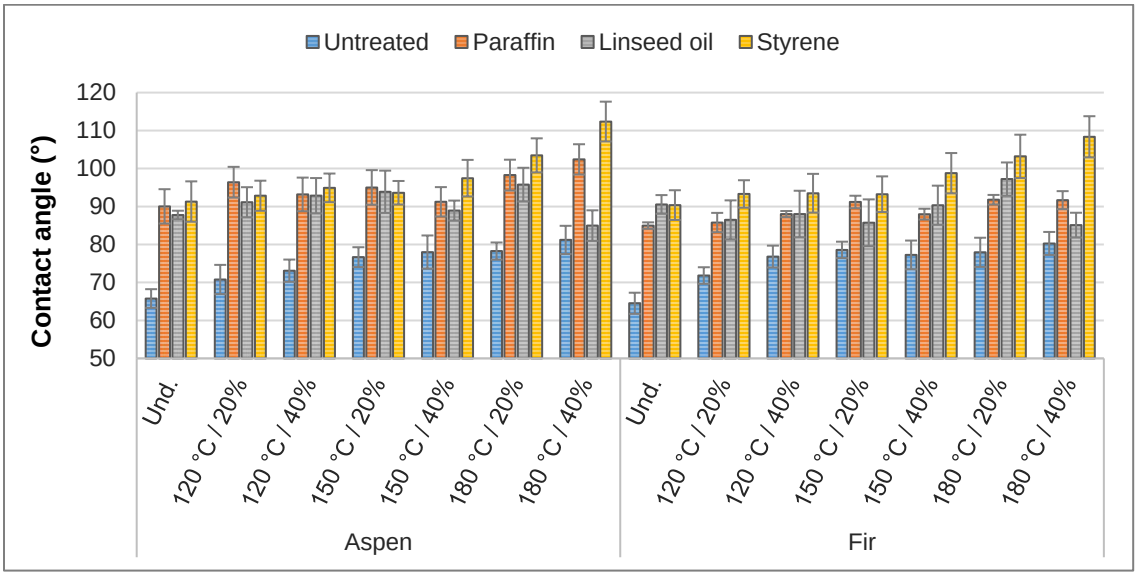

Fig. 3. Contact angle values of impregnated and densified wood specimens

Regarding densification condition, the highest contact angle average for both wood species was in the specimens compressed at 180 °C with the 20% and 40% ratio (93.94° and 95.24° for aspen; 92.53° and 91.35° for fir), while the lowest was obtained in the undensified specimens (83.72° for aspen and 82.60° for fir) (Table 2). Depending on the compression ratio and compression temperature, the contact angle values of untreated and impregnated wood specimens generally increased. The contact angle tends to increase in untreated and styrene-impregnated specimens with increasing compression ratio. The

compression ratio did not have a significant effect on other impregnated wood specimens. The effect of compression temperature on the contact angle was more pronounced (Fig. 3). The reason for the increase in the contact angle of the densified specimens can be shown as the decrease in the void volume of the wood specimens and their transformation into a more homogeneous structure, and also the decrease in the roughness values of the specimens. The SEM analyses show that depending on the increase in the compression ratio, the void volume decreased and the homogeneity increased. In a previous study, examining the chemical structure and wetting behavior of surface densified wood, it was stated that there was no significant chemical change, but the contact angle increased and the wettability decreased in densified surfaces (Rautkari *et al.* 2010). It has been reported that after short-term thermo-mechanical densification, the contact angle of different wood veneers increased and became more hydrophobic, and the effect of temperature on contact angle was more evident than pressure (Bekhta *et al.* 2017).

In terms of compression temperature, contact angle values increased and wettability decreased with temperature increase in both untreated and impregnated aspen and fir wood specimens (Fig. 3). It is thought that the wood surfaces being less hygroscopic with the effect of high temperature during the densification process have an effect on the results. This is because it is well known that the equilibrium moisture content and hygroscopicity of the wood exposed to high temperatures decrease (Esteves and Pereira 2009; Kaygın *et al.* 2009; Fang *et al.* 2012; Kocaefe *et al.* 2015; Boonstra 2016). Numerous studies have reported that the surface wettability of thermally treated wood is reduced due to degradation of hygroscopic components (especially hemicellulose) (Hakkou *et al.* 2005; Esteves *et al.* 2007; Kocaefe *et al.* 2008; Candan *et al.* 2012; Kutnar *et al.* 2012; Bakar *et al.* 2013; Chu *et al.* 2016; Gérardin 2016). The compression temperature increase causes an increase in the contact angle, which is consistent with findings of previous studies (Diouf *et al.* 2011; Ünsal *et al.* 2011; Bekhta and Krystofiak 2016; Bekhta *et al.* 2018). After the densification processes, depending on the compression ratio and temperature, the contact angle of untreated, paraffin, linseed oil, and styrene-impregnated specimens increased up to 23%, 56%, 46%, and 71% for aspen wood, and up to 24%, 42%, 51%, and 68% for fir wood, respectively.

Microscopic observations can help to better understand property changes in treated wood. The SEM micrographs of cross-sections from impregnated and densified fir and aspen specimens are shown in Figs. 4 and 5. Impregnation and thermo-mechanical densification processes significantly affected the morphological structure of the wood specimens. It was observed that the impregnation agents were able to cling to the cell walls and some lumens were partially or totally filled. Compared to the control specimens (Figs. 4a1 and 5a1), especially in the styrene-impregnated specimens, it was observed that most of the lumen cavities were filled (Figs. 4d1 and 5d1). The observation results were also consistent with the retention data in Table 1. As shown in Figs. 4 and 5, buckling the cell walls and reducing the volume of void spaces in all densified specimens (untreated and impregnated). Lumen diameters of the densified wood specimens were considerably reduced compared to the undensified specimens. In addition, the lumen cavities of the pre-impregnated specimens were almost completely closed after the densification process. This is more evident in styrene-pretreated specimens (Figs. 4d2, 4d3, 5d2, and 5d3). The effect of compression temperature on lumen properties was not clear. It can be said that partial or complete closure of the lumens causes a decrease in hygroscopicity and smoother cell walls in the wood material. In this context, SEM images support the surface roughness and contact angle (wettability) results.

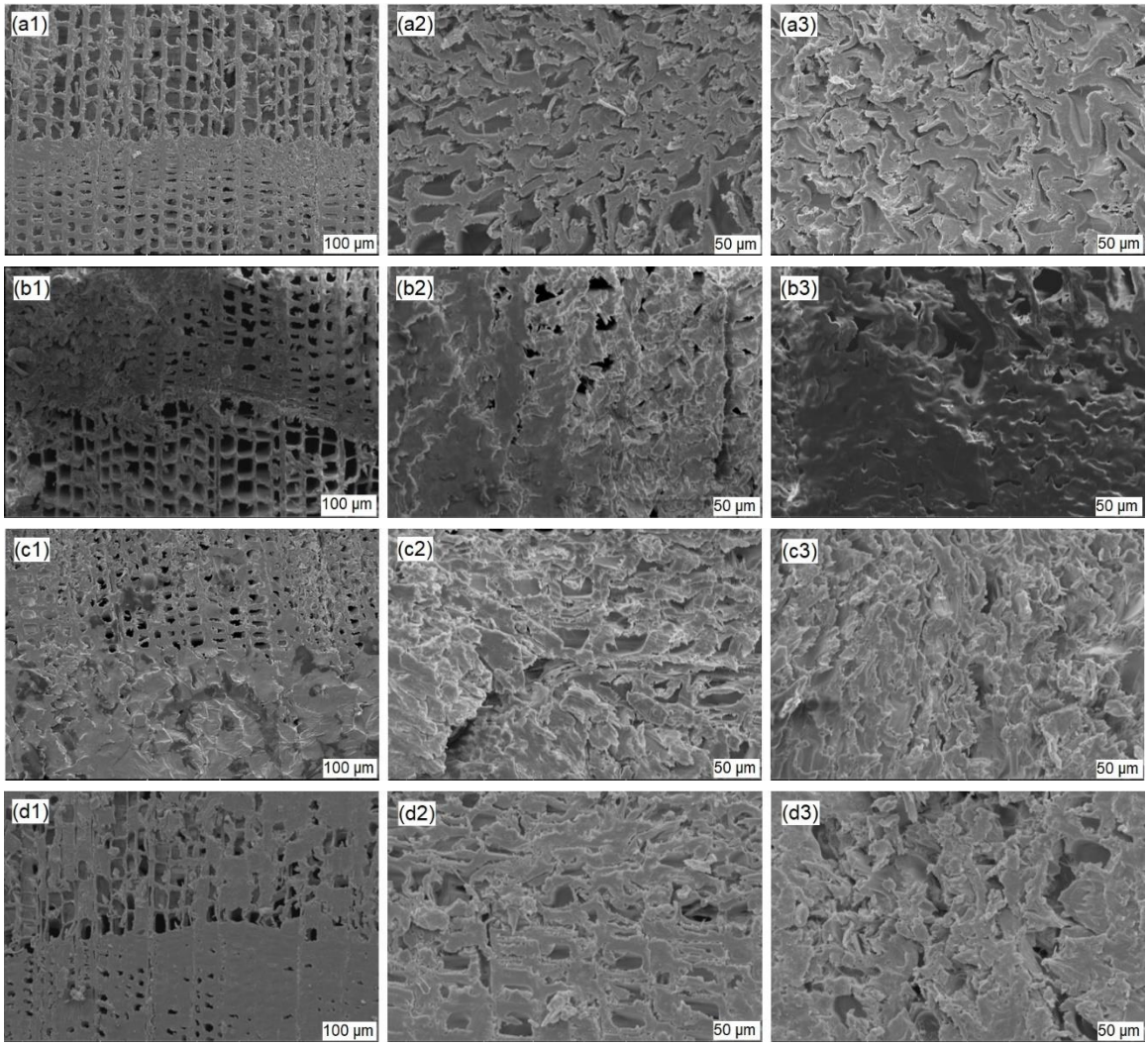

Fig. 4. SEM images of cross-sections of fir wood specimens: (a) untreated (non-impregnated); (b) paraffin-impregnated; (c) linseed oil-impregnated; (d) styrene-impregnated; (1) undensified; (2) 40% compressed at 120 °C; (3) 40% compressed at 180 °C

One of the important problems seen in wood material densified under high pressure is various deformations, such as collapses, breakage, and cracking, which occur in the cell structure (Kultikova 1999; Doğu *et al.* 2010; Ahmed *et al.* 2013; Budakçı *et al.* 2016; Bekhta *et al.* 2017). These deformations that occur during densification adversely affect the physical and/or mechanical properties of the wood material, depending on the type and amount (Navi and Girardet 2000; Kutnar *et al.* 2009; Pelit *et al.* 2018). According to the SEM images, various deformations occurred as a result of the collapse of the cell wall in all densified specimens. In particular, wood cell deformations are clearly seen in the fir specimen compressed at lower temperature (120 ℃), which is untreated (non-impregnated) (Figs. 4a2). In contrast, it was observed that pre-impregnation with paraffin, linseed oil, and styrene reduced the amount of cell deformations and changed the type of the deformation (usually elastic buckling) in densified aspen and fir specimens. Possible reasons for this may be the softening (an improvement in glass transition temperature) of the wood cell wall with the effect of the heat of the impregnation agents, and the

impregnation agents penetrating into the solid wood transmitting the heat applied in the densification process more evenly and with less loss. It has been stated in previous studies that densified wood exhibits a glassy behavior under low temperature, low moisture content, and short treatment time. When the wood temperature is above the glass transition temperature or pre-softening, the amorphous polymers can be compressed without any major deformation or fracture in the cell structure (Wolcott *et al.* 1990, 1994; Kutnar and Sernek 2007).

As a result, pre-impregnation with paraffin, linseed oil, and styrene before densification played an important role in reducing the hygroscopicity of the wood due to the effect of filling the cell lumens and obtaining smoother surfaces, as well as densification without significant cell deformations.

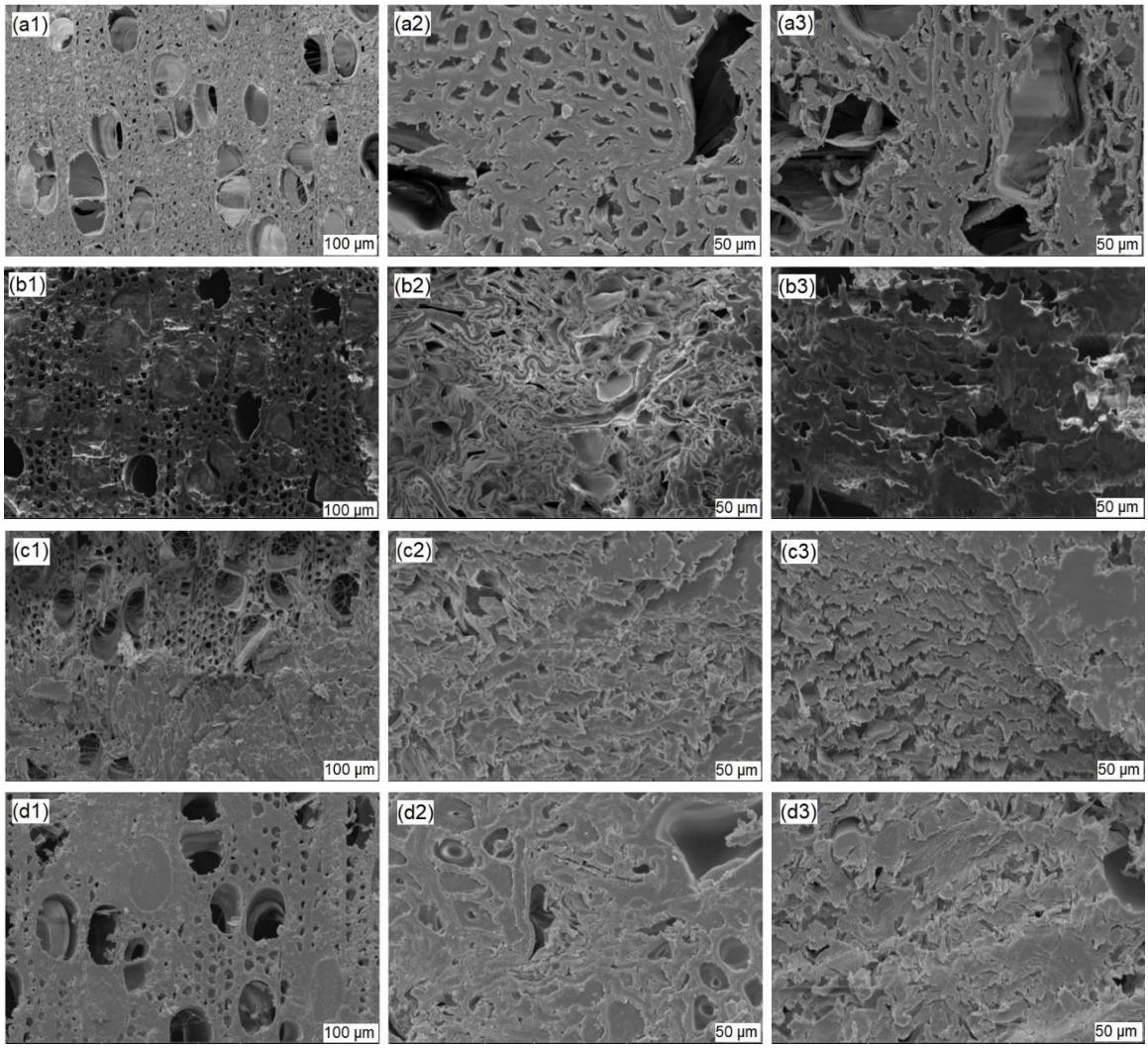

Fig. 5. SEM images of cross-sections of aspen wood specimens: (a) untreated (non-impregnated); (b) paraffin-impregnated; (c) linseed oil-impregnated; (d) styrene-impregnated; (1) undensified; (2) 40% compressed at 120 °C; (3) 40% compressed at 180 °C

A difficulty inherent in the present study or similar studies is that the contact angle results can depend on changes that occur at an extremely small scale, *i.e.* nanoscale changes. For future studies, time-of-flight surface ionization mass spectrometry analysis

and evaluation of nano-scale roughness by atomic force microscopy, as well as chemical compositional analysis can be investigated.

CONCLUSIONS

1. The results of this study show that the pre-impregnation and densification treatments of wood materials caused major changes in wood surface roughness, wettability, as well as in its morphological structure. With impregnation processes, the roughness of both undensified and densified aspen and fir specimens decreased, and the contact angle increased. More successful results (lower roughness and higher contact angle) were obtained in styrene-impregnated specimens compared to paraffin and linseed oil.
2. After densification, the surface roughness of all specimens (untreated and impregnated) decreased depending on the compression ratio and temperature. Lower roughness values were obtained in the specimens densified with high compression ratio (40%). While the change in compression temperature did not have a significant effect on the roughness values of the fir specimens, the roughness values of the aspen specimens (especially styrene-impregnated) usually decreased with increased compression temperature.
3. As a result of the increase in the compression ratio, the contact angle increased in the untreated and styrene-impregnated specimens. The compression ratio did not have a significant effect on other impregnated specimens. The effect of temperature on the contact angle was more evident than that of compression ratio. Contact angle increased with compression temperature increase in both untreated and impregnated specimens.
4. In styrene pre-treated and densified aspen and fir specimens, the surface roughness decreased up to 72% and 63%, respectively, and the contact angle increased up to 71% and 68%, respectively. These rates were lower in paraffin and linseed oil pretreated specimens.
5. According to SEM observations, after impregnation, especially as a result of *in situ* polymerization of styrene, most of the lumens were filled and a hydrophobic film was formed in the wood structure. This supports the determined surface roughness and wettability results. In contrast, impregnation with paraffin, linseed oil, and styrene allowed wood to be densified without significant deformations in the wood cell structure.

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