WOOD RESEARCH doi.org/10.37763/wr.1336-4561/66.5.762776
e-ISSN 2729-8906
66(5): 2021 762-776 pp.

SELECTED PROPERTIES OF COMPREGNATED WOOD USING LOW MOLECULAR WEIGHT PHENOL FORMALDEHYDE AND SUCCINIC ANHYDRIDE

SARAH AUGUSTINA[1], IMAM WAHYUDI[1], WAYAN DARMAWAN[1], JAMALUDIN MALIK[2], NAOKI OKANO[3], TAIYO OKADA[3], KAZUSHIGE MURAYAMA[4], HIKARU KOBORI[3], YOICHI KOJIMA[3], SHIGEHIKO SUZUKI[5]

[1]IPB UNIVERSITY, INDONESIA
[2]FOREST PRODUCTS RESEARCH AND DEVELOPMENT CENTER, INDONESIA
[3]SHIZUOKA UNIVERSITY, JAPAN
[4]FORESTRY AND FOREST PRODUCTS RESEARCH INSTITUTE, JAPAN
[5]SHIZUOKA PROFESIONAL UNIVERSITY OF AGRICULTURE, JAPAN

(RECEIVED DECEMBER 2020)

ABSTRACT

The aim of this study was to investigate the effect of impregnating materials (low molecular weight phenol formaldehyde or LmwPF and succinic anhydride or SA), their concentrations (5 and 10%), and compression ratios (20 and 40% from initial thickness) on improvement of specific gravity (SG) and dimensional stability on nyatoh, sepetir, and pisang putih wood; and then compared them to control and densified wood. The results showed that SG and dimensional stability of compregnated wood were affected by all parameters studied. Higher compression ratio and concentration will result in a greater improvement. In general, SG and dimensional stability of compregnated wood were better than the control. SG of LmwPF- and SA-compregnated wood increased by 10.69–22.31% and 6.96–23.09%, respectively. Utilization of LmwPF and SA has significantly reduced the spring-back, but the latter is better. The compression-set recovery after compregnation was 18.34–33.99%, while after densification was 47.86–71.49%.

KEYWORDS: Compregnation, compression ratio, lesser-used wood species, low molecular of phenol formaldehyde, succinic anhydride, spring-back.

INTRODUCTION

Densification is a process that has been widely applied to modify the properties of wood. After processing, the wood becomes denser and its specific gravity (SG) increases significantly (Inoue et al. 1991, Kutnar et al. 2009, Tu et al. 2014, Pelit and Yalçin 2017). This process in general can be divided into four stages: starting with softening of cell wall or plasticizing, then flattening the wood constituents due to compression, followed by setting in deformed condition, and ending with fixation which prevents the wood from returning to its original shape and size (Morsing and Hoffmeyer 1998). The effectiveness of densification is influenced and depends on internal and external factors. Anatomical characteristics and moisture content are the main internal factors to be considered (Tabarsa and Chui 2000, Nairn 2005, Kamke and Kutnar 2010, Darwis et al. 2017), while pre-treatment, direction of compression, compression ratio (CR), temperature, as well as closing and holding time during treatment are the external factors (Shams et al. 2006, Rautkari et al. 2011).

Hot immersion before pressing is commonly used as a pre-treatment because it can accelerate the softening of the lignin so that the wood is easily flattened and compacted (Navi and Sandberg 2012, Khalil et al. 2014). According to Kutnar and Sernek (2007), Kamke and Kutnar (2010), lignin begins to soften when the wood temperature is above its glass transition temperature (Tg). According to Kelley et al. (1987), Furuta et al. (1997), Kong et al. (2017), Tg of lignin is between 50°C to 100°C depending on the species and age of the wood.

Apart from pre-treatment, the applied compression ratio must also be considered. An appropriate compression ratio level will significantly increase the strength, SG, and dimensional stability of the wood; conversely, too high a compression ratio level will not only cause excessive deformation but also cause severe damage to the wood, thereby reducing its strength and dimensional stability (Bao et al. 2017). The correct compression ratio levels can also reduce the ability of the wood to return to its original shape and size (spring-back), especially if wood is put into the room with high humidity. According to Sadatnezhad et al. (2017), spring-back is related to the hygroscopicity of wood and the release of internal stresses that occur during densification. Its value will increase as the compression ratio increases. There is severe spring-back, up to 75%, on poplar wood after the wood is processed using techno-hydro-mechanical (THM) at 50% compression ratio (Bao et al. 2017), while according to Pelit et al. (2016), the set recovery produced by 25% compression ratio was lower than 50% compression ratio on densified Uludağ fir wood (4–8% versus 18–24%).

In order to reduce the spring-back phenomenon in densified wood, the wood must be impregnated with proper materials prior to compaction. This process is called compregnation which is a combination of densification (compaction) and impregnation. Impregnated with synthetic resins such as low molecular weight phenol formaldehyde (LmwPF) or with anhydride such as succinic anhydride (SA) will increase the hydrophobicity of wood and at the same time strengthen its fixation (Shams et al. 2004, Khalil et al. 2014). Apart from being a bulking agent, the presence of impregnation materials also plays a role in the formation of covalent bonds between wood components so that internal stresses remain stored in the microfibrils and the cell wall matrix (Morsing and Hoffmeyer 1998). According to Wan and Kim (2006), Gabrielli

(2009), Lahtela and Karki (2014), Bao et al. (2016), compregnated wood has better strength and dimensional stability than compacted wood without impregnation (densified wood).

Studies on compregnated wood especially using the lesser-used tropical species are very limited. Therefore, this study aims to analyze nyatoh (*Palaquium* spp.), sepetir (*Sindora* spp.) and pisang putih (*Mezzettia* spp.) wood which had been impregnated with LmwPF or SA prior to pressing. The main focus of this study is on the changes occurred in SG and dimensional stability due to the treatment applied and the phenomenon of spring-back. Comparison of similar characteristics to those of densified wood and control wood are also discussed.

MATERIAL AND METHODS

Logs 120 cm in length and 50 cm in diameter at breast height from the bottom of the trunk of nyatoh, sepetir, and pisang putih trees was used as the main samples. The trees came from a forest concession area in North Kalimantan Province, Indonesia. The logs were cut into 3 cm thick boards and air-dried for 2 months before converting into wood samples in eleven treatment categories. Sample size for each category is presented in Tab. 1. Only the clear wood samples namely free from knots, mold, fungi, and visible defects were tested.

Tab. 1: Sample size of each treatment category.

Treatments		**Size (cm)**
Category	**Code**	
1. Untreated	C	2 x 2 x 2
2. Hot immersion without impregnating material but densified at 20% CR	WR–0–20	2 x 2 x 2.4
3. Impregnated and hot immersion within 5% LmwPF and densified at 20% CR	PF–5–20	2 x 2 x 2.4
4. Impregnated and hot immersion within 10% LmwPF and densified at 20% CR	PF–10–20	2 x 2 x 2.4
5. Impregnated and hot immersion within 5% SA and densified at 20% CR	SA–5–20	2 x 2 x 2.4
6. Impregnated and hot immersion within 10% SA and densified at 20% CR	SA–10–20	2 x 2 x 2.4
7. Hot immersion without impregnating material but densified at 40% CR	WR–0–40	2 x 2 x 2.8
8. Impregnated and hot immersion within 5% LmwPF and densified at 40% CR	PF–5–40	2 x 2 x 2.8
9. Impregnated and hot immersion within 10% LmwPF and densified at 40% CR	PF–10–40	2 x 2 x 2.8
10. Impregnated and hot immersion within 5% SA and densified at 40% CR	SA–5–40	2 x 2 x 2.8
11. Impregnated and hot immersion within 10% SA and densified at 40% CR	SA–10–40	2 x 2 x 2.8

The number of samples for densification without impregnation at 20 and 40% compression ratio was 18 pieces (2 levels of compression ratio x 3 replications x 3 species), meanwhile for compregnation they were 360 pieces (8 categories x 15 replications x 3 species).

Densification and compregnation process

Before proceed into the next steps, sample dimensions were measured to obtain its volume and then weighed. For densification treatment (without impregnation), after hot soaking within water at 80°C for 3 hours the samples were directly compacted using machine at 160°C with specific pressure of 2.672 MPa for 15 min with 20 and 40% compression ratio levels from initial thickness. After that the samples were conditioned overnight before testing. For compregnation treatment, the samples were impregnated by 5 and 10% (w/w) of LmwPF and SA solutions separately using –98 kPa vacuum pressure for 15 min then followed by 350 kPa pressure for 4 hours. After that, the samples were hot (80°C) immersed in each impregnating solutions for 3 hours, then immediately compacted using machine at 160°C with a specific pressure of 2.672 MPa for 15 min with 20 and 40% of compression ratio. The samples were also then conditioned overnight prior to testing.

Measurement of specific gravity (SG)

Densified and compregnated wood samples were weighed (Wo) and their volumes were measured (Vo), then oven-dried at 103 ± 2°C for 24 hours to obtain oven-dried weight (W_1). The SG measurements were carried out before and after treatment to record changes in value. The value was calculated according to Eq. 1:

$$SG = \frac{W_1 / Vo}{\rho_{water}} \tag{1}$$

where: W_1- sample weight in oven-dried condition; before and after treatment (g), *Vo*- sample volume in air-dried condition; before and after treatment (cm^3), ρ_{water}- density of water 4°C (= 1 $g \cdot cm^{-3}$).

Measurement of weight percent gain (WPG)

The WPG was calculated according to Eq. 2:

$$WPG\ (\%) = \frac{W_1 - Wo}{Wo} \times 100 \tag{2}$$

where: W_o- sample weight in oven-dried condition before treatment (g), W_1- sample weight in oven-dried condition after treatment (g).

Measurement of compression-set (C-set)

Compression set is used to determine the thickness variation after densification. The value was measured according to Eq. 3:

$$C\text{-}set\ (\%) = \frac{To - Ta}{To} \times 100 \tag{3}$$

where: T_o- sample thickness before densification (mm), T_a- sample thickness in oven-dried condition after densification (mm).

Measurement of dimensional stability

Dimensional stability in this study was determined by compression-set recovery (CSR) and thickness swelling (TS) after water soaking process. Densified and compregnated-wood samples were oven-dried at 103 ± 2°C for 24 hours to obtain their oven-dried thickness before swelling. Oven-dried samples were then immersed in water at room temperature for 24 hours and then oven-dried again at 103 ± 2°C for 24 hours. The thickness in oven-dried condition was measured before and after soaking. The CSR and TS were measured according to Eqs. 4 and 5:

$$CSR\ (\%) = \frac{Tss - Tsb}{To - Tsb} \times 100 \tag{4}$$

$$TS\ (\%) = \frac{Tss - Tsb}{Tsb} \times 100 \tag{5}$$

where: To- sample thickness before densification (mm), Tsb- sample thickness in oven-dried condition before swelling (mm), Tss- sample thickness in oven-dried condition after swelling (mm).

Scanning electron microscopy (SEM) observation

SEM images are used to investigate microscopic changes occurred in densified and compregnated samples. The samples were vacuum dried at 40°C for 24 hours and its cross-sections were sputter coated with gold for 120 s and 30 mA, and observed under SEM (JEOL JSM-6510LV, Japan) at 15 kV.

Data analysis

Experimental design used was a completely randomized design with four factors namely wood species, compression ratio, impregnating material, and the concentration of impregnated material solution; all in three levels. Analysis of variance (ANOVA) was performed to evaluate the effect of species, compression ratio, impregnating material, and its concentration on SG and dimensional stability of treated woods. If there are significant differences from each factor and the interactions, it will be followed by Duncan's multiple distance test.

RESULTS AND DISCUSSION

Specific gravity (SG)

The average SG after treatment is presented in Tab. 2. It can be seen that interaction among wood species, impregnating material, and compression ratio has a significant effect on SG at the 5% significance level (Tab. 3).

Tab. 2: Specific gravity of three wood species after treatment.

Treatment category	Wood species					
	Nyatoh		Sepetir		Pisang putih	
	Before	After	Before	After	Before	After
Untreated	0.48 ± 0.06^{de}		0.36 ± 0.03^{a}		0.52 ± 0.02^{efg}	
WR–0–20	0.49 ± 0.07	0.56 ± 0.07^{gh}	0.35 ± 0.05	0.40 ± 0.05^{abc}	0.53 ± 0.01	0.57 ± 0.02^{gh}
SA–5–20	0.48 ± 0.03	0.56 ± 0.03^{gh}	0.35 ± 0.04	0.42 ± 0.04^{abcd}	0.54 ± 0.01	0.59 ± 0.02^{gh}
SA–10–20	0.48 ± 0.05	0.59 ± 0.06^{gh}	0.36 ± 0.03	0.45 ± 0.02^{bcd}	0.54 ± 0.01	0.62 ± 0.02^{hi}
PF–5–20	0.49 ± 0.05	0.56 ± 0.07^{gh}	0.35 ± 0.05	0.40 ± 0.05^{abc}	0.53 ± 0.01	0.57 ± 0.01^{gh}
PF–10–20	0.48 ± 0.04	0.55 ± 0.05^{fgh}	0.31 ± 0.05	0.37 ± 0.05^{a}	0.54 ± 0.02	0.58 ± 0.03^{gh}

WR–0–40	0.47 ± 0.07	0.62 ± 0.08^{hi}	0.37 ± 0.02	0.49 ± 0.04^{def}	0.51 ± 0.01	0.66 ± 0.03^{i}
SA–5–40	0.49 ± 0.06	0.58 ± 0.08^{gh}	0.32 ± 0.05	0.39 ± 0.05^{ab}	0.49 ± 0.06	0.55 ± 0.05^{gh}
SA–10–40	0.46 ± 0.05	0.57 ± 0.05^{gh}	0.33 ± 0.03	0.45 ± 0.03^{bcd}	0.49 ± 0.05	0.59 ± 0.04^{gh}
PF–5–40	0.49 ± 0.05	0.59 ± 0.06^{h}	0.36 ± 0.02	0.46 ± 0.03^{cde}	0.53 ± 0.01	0.59 ± 0.01^{h}
PF–10–40	0.45 ± 0.02	0.55 ± 0.03^{fgh}	0.34 ± 0.01	0.45 ± 0.02^{bcd}	0.51 ± 0.03	0.58 ± 0.03^{gh}

Note: SG values are averages of 15 replicates with standard deviation. Values within a column followed by the same letters are not significantly different at 5% significance level using Duncan's multiple range test.

The highest increase in SG was found in sepetir wood. For all wood species, succinic anhydride (SA) resulted in a higher SG improvement compared to LmwPF, and SG increased with increasing compression ratio level. Result also showed that compregnation and densification treatments could significantly increase the SG of the woods compared with untreated (control). However, SG of the compregnated wood was smaller than that of densified wood. The SG tended to increase slightly with the increase in the concentration of impregnating material solution.

Tab. 3: ANOVA results of each parameters and factors.

Parameter	Factors														
	JK	CM	CC	CR	JK × CM	JK × CC	JK × CR	CM × CC	CM × CR	CC × CR	JK × CM × CC	JK × CM × CR	JK × CC × CR	CM × CC × CR	JK × CM × CC × CR
SG	**	ns	ns	**	ns	ns	ns	**	**	ns	ns	*	ns	ns	ns
WPG	**	**	**	ns	**	**	ns	**	ns	ns	*	ns	ns	ns	ns
C-set	**	**	**	**	**	ns	ns	ns	ns	ns	*	ns	ns	ns	ns
CSR	**	**	**	**	*	ns	*	**	ns	**	ns	*	ns	**	ns
TS	**	**	**	**	**	ns	**	ns	**	ns	ns	**	ns	ns	ns

Note: ns = not significant; * = significantly different at 5% significance level; ** = significantly different at 1% significance level; JK = wood species; CM = impregnating material, CC = concentration, CR = compression ratio.

Generally, it can be said that SG increases after treatment. In case of nyatoh wood there was an increase of 14.13 and 18.83% for 20 and 40% compression ratio, respectively; while in sepetir and pisang putih wood there was 15.58 and 23.06% as well as 8.46 and 14.56%, respectively. Sepetir wood has the highest improvement in SG at both compression ratio and impregnating material, and the values were significantly different from the other species. This phenomenon is related to its anatomical features, i.e. greater void volume dues to larger vessel diameter and more frequent number of the vessels (Augustina et al. 2020). Higher portion of void volume resulted in a higher level of deformation in the cell lumens during compaction. And as the result, the wood is more compacted. This finding is coincided with Darwis et al. (2017). They stated that the increasing SG after densification is mainly influenced by the porosity or proportion of cell cavities. Higher void volume leads to a higher level of deformation in the cell lumens, as well as a large reduction in void volume (Tu et al. 2014, Bao et al. 2017). It also can be convinced by its C-set value. As shown in Fig. 1, sepetir wood had higher C-set value (18.07%) than nyatoh (17.15%) and pisang putih (12.87%) wood.

According to Tab. 2, SG values generated by 20 and 40% compression ratio were 0.52 and 0.54, respectively. These results are related to the increasing number of flattened cells during compression. The same phenomenon was also reported by Inoue et al. (1991). The SG of modified sugi wood increased from 0.36 to 0.50 at 30% compression ratio and doubled at 60% compression ratio. Therefore, there is no doubt that the compregnation will resulted in a significant reduction in lumen diameter of vessels and fibers, thereby reducing the sizeable portion of the void volume.

At 20% compression ratio, the average increase in SG of compregnated and densified woods were 10.68–18.33% and 7.02–12.50%, respectively; and were 13.93–22.31% and 22.73–24.49% at 40% compression ratio. This results indicate that the higher the compression ratio, the more impregnating material that come out of the wood. This phenomenon was supported by the fact that WPG at 40% compression ratio is lower than 20% compression ratio. According to Zhao et al. (2015), a higher compression ratio will result in greater internal stress. The greater internal stresses will push more impregnating materials out of the wood (Lykidis et al. 2019). This resulted in a reduction in deposited and solidified impregnating material in the cell cavity. This phenomenon is also related to the lower C-set generated in compregnated wood compared to densified wood. At 20% compression ratio, the average value of C-set of compregnated and densified wood were 8.82–14.17% and 12.75–17.19%, respectively; while at 40% compression ratio they were 14.17–21.48% and 26.8–28.07%. This can occur because of the variation in the internal stresses that is formed for each compression ratio which is induced by a decrease in humidity (Pelit et al. 2014). The presence of impregnating substances can also prevent deformation in wood.

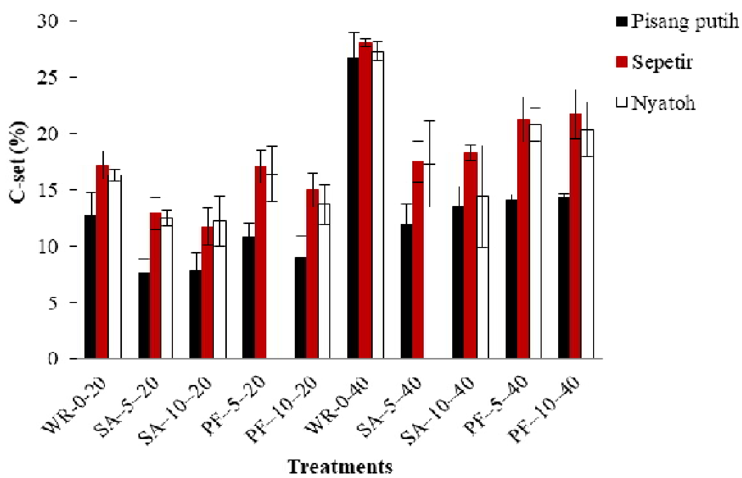

Fig. 1: Compression set of each treatments.

Average SG for compregnated wood using SA as impregnating material (SA-compregnated wood) increased by 10.69–18.33% at 20% compression ratio and 13.93–22.31% at 40% compression ratio, respectively; while those impregnated with LmwPF (LmwPF-compregnated wood) were 6.96–14.36% and 11.12–23.09%, respectively. This phenomenon is also convinced by the WPG (Fig. 2).

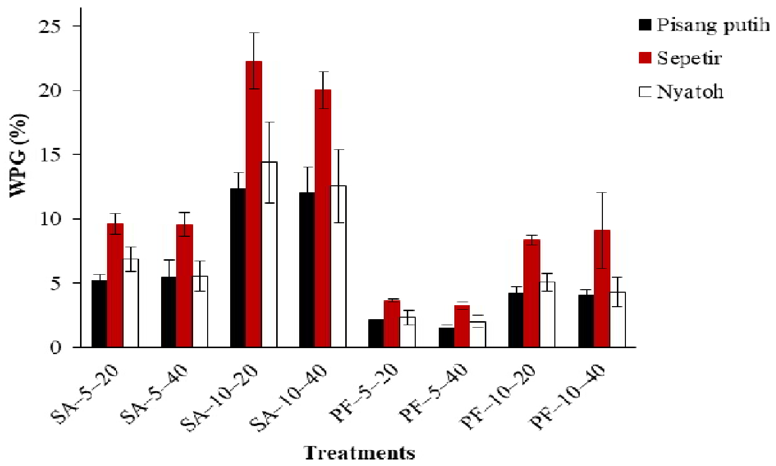

Fig. 2: Weight percent gain of each treatments.

The WPG of the SA-compregnated wood was higher than that of the LmwPF-compregnated wood. It can be said that the changes in microstructure are the result of the SA which covers higher portion of the cell cavity and acted as a blocking agent compared to the LmwPF. According to Chen et al. (2014), modification with SA will result in replacement of the hydroxyl group with a succinic group that is covalently bound. The higher WPG in succinilation is due to the higher molecular weight of SA and adduction of a 4-carbon chain with a carboxyl group to the lignin. Succinilated is also capable of causing the lignin to expand more effectively, making reactive chemicals sites more accessible, and therefore enhancing the rate of modification.

Average SG for the SA-compregnated wood increased by 9.69–17.31% for 5% concentration and 14.93–23.33% for 10% concentration, while those of the LmwPF-compregnated wood increased by 8.59–17.12% and 9.48–20.33%, respectively. The WPG of the SA-compregnated wood increased by 5.45–9.56% and 12.05–20.02% for 5 and 10% concentration, respectively; while those of the LmwPF-compregnated wood were 1.48–3.23% and 4.05–9.10%. This phenomenon indicates that an increase in the impregnating concentration causes an increase in the WPG, which in turn will also result in an increase in SG. The positive correlation between WPG and SG was also confirmed by Nur et al. (2011).

Dimensional stability

Dimensional stability in this study is approached by compression-set recovery (CSR) and thickness swelling (TS) after water soaking process. The CSR and TS of the three wood species with 5 and 10% of LmwPF and SA solutions at 20 and 40% compression ratio are shown in Figs. 3 and 4. It can be seen that sepetir wood generated higher dimensional stability, as indicated by lower the CSR and TS values. CSR and TS increased slightly with an increase of compression ratio. The compregnation resulted in lower CSR and TS than densification. These two values decrease significantly with increasing concentration. This phenomenon was confirmed by the ANOVA results (Tab. 3).

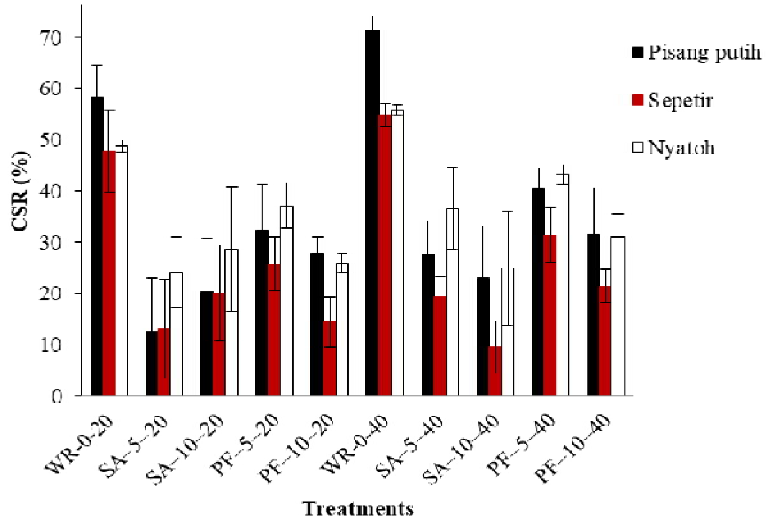

Fig. 3: Compression-set recovery of each treatments.

Average CSR values for nyatoh, sepetir, pisang putih wood were 32.95 and 38.37%, 24.24 and 27.32%, and 30.29 and 38.89% for 20 and 40% compression ratio, respectively; while average TS were 11.17 and 17.10%, 9.92 and 15.58%, and 10.90 and 15.96%, respectively. The lower CSR and TS on sepetir wood are due to increased SG and greater deformation during compression. The higher SG indicates more cells flattening during compression, and consequently increased dimensional stability. The same phenomenon also confirmed by Augustina et al. (2020).

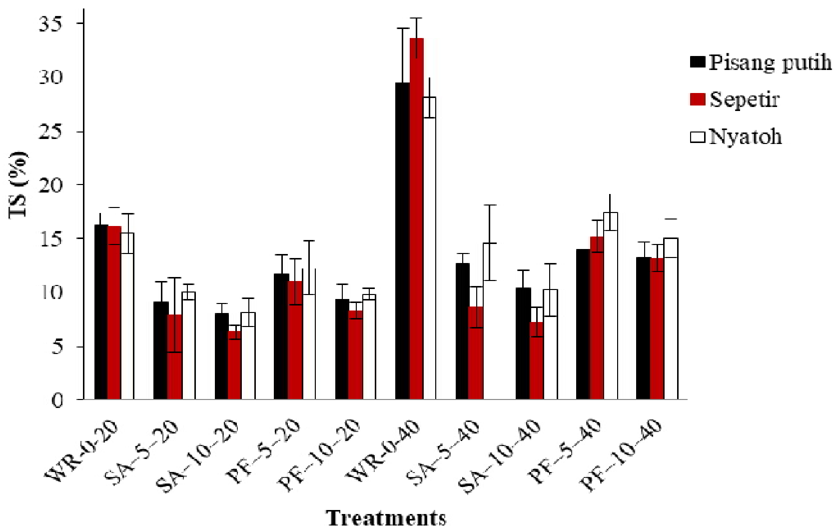

Fig. 4: Thickness swelling of each treatment.

According to Miyoshi et al. (2016), there are 4 different types of cell lumens deformation during compression, namely type I (S-shape cells), type II (cells that have 3-4 projections), type III (cells with protrusion in center or tip), and type IV (elongated shape cells). Sepetir wood has a uniform type of deformation and is categorized as type I and II (Fig. 5). This can lead to permanent and irreversible structural changes and deformations.

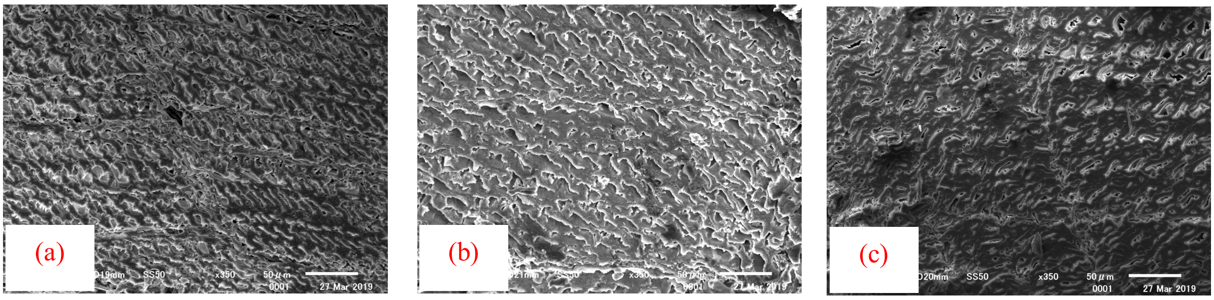

Fig. 5: Type of deformation during compression: a) nyatoh, b) sepetir, and c) pisang putih. Scale bar = 50 µm.

Irreversible swelling occurs due to the breaking of the covalent bonds between hemicellulose and lignin mechanically when the stress exceeds the bond strength (Ohlmeyer and Paul 2010). It was also confirmed by Miyoshi et al. (2016). They stated that the S-shape cells are difficult to swell and tend to increase the dimensional stability of compressed wood. The higher CSR and TS on nyatoh and pisang putih wood are due to higher initial SG which led to uneven distribution of internal stresses during compression. Bao et al. (2017) added that recovery of wood cells depends on distribution of internal stresses.

Average CSR values generated by 20% and 40% compression ratio were 24.24–32.95% and 27.32–38.89%, respectively (Fig. 3); while those of TS were 9.92–11.17% and 15.58–17.10% (Fig. 4). It can be seen that CSR and TS increased with an increase of compression ratio (Tab. 2). A larger compression ratio will produce a greater reaction force inside wood which led the cells to return to its original shape easily. This was especially the case at 40% compression ratio. The same phenomenon was confirmed by Bao et al. (2017) and Darwis et al. (2017). They stated that the THM process induces the deformation of fibers and microfibrils which results in an increase in internal stresses. These stresses allow those fibers and microfibrils to recover during water soaking process, although not 100% recover. It indicated that our treatments provide improvements, especially in dimensional stability.

Average CSR after compregnation and densification treatment at 20% compression ratio were 18.34–28.99% and 47.86–58.28%, respectively; while at 40% compression ratio they were 20.44–33.99% and 54.82–71.49% (Fig. 3). Average TS after compregnation and densification at 20% compression ratio were 8.37–10.09% and 15.5–16.21%, respectively; while at 40% compression ratio they were 11.06–14.35% and 28.10–33.62% (Fig. 4). It can be seen that CSR and TS of compregnated wood are lower compared to those of densified wood. These findings are related to the treatments applied. Bao et al. (2017) and Dos Santos et al. (2018) stated that hydrothermal treatment, especially at temperature above T_g lignin can cause damage to cellulose cross-links, breaking of hemicellulose chains and slight cleavage of lignin that may release the internal stresses. Impregnating materials not only blocks the entry of water into the wood cells by covering the void volume, but also contributes to holding the wood back to its original shape and size (recovery) due to its role as bulking agent. It was also agreed by Ghorbani and Bavaneghi (2016) which stated that impregnating material contributes to reduce the number of hydroxyl group and occupies the intermolecular space in the wood cell wall, thereby decreasing hygroscopicity of wood and consequently increasing the dimensional stability.

The average CSR for the SA-compregnated wood and LmwPF-compregnated wood were 15.51–28.62% and 23.27–34.36%, respectively; while average of their TS were 7.52–10.78% and 11.92–13.66%. It can be seen that both CSR and TS of the SA-compregnated wood are lower than LmwPF-compregnated wood. This phenomenon is correlated with a higher WPG and characteristic of SA which can substitute hydroxyl group with succinic groups. Hill (2006) and Scarica et al. (2018) stated that the application of SA as the impregnating agent can increase stress relaxation because it can act as an internal plasticizer. These facts do support our findings. In case of the LmwPF-compregnated wood, the lower WPG is not sufficient to hold water during the water soaking process. Kajita et al. (2004) stated that LmwPF can easily penetrate into the cell walls, and almost all of them are located in the cell walls and little or no resin visible in the cell lumina.

The average CSR for the SA-compregnated wood at concentrations of 5 and 10% were 16.23–30.40% and 14.80–26.85%, respectively; while for the LmwPF-compregnated wood they were 28.59–40.23% and 17.94–29.69%, respectively. On the other hand, average TS for the SA-compregnated wood at concentration of 5 and 10% were 8.27–12.36% and 6.77–9.20%; while for LmwPF-compregnated wood they were 12.84–14.91% and 10.72–12.42%. It can be seen that in both type of impregnating material, the values of CSR and TS decrease with increasing impregnating concentration. Negative correlation between CSR and impregnating concentration as well as between TS and impregnating concentration can also be described by the WPG (Fig. 6). It was strongly supported by Ghorbani and Bavaneghi (2016). Acetylation affects the spring-back of the board and causes a significant increase, in particular TS and water absorption, along with the increase in WPG.

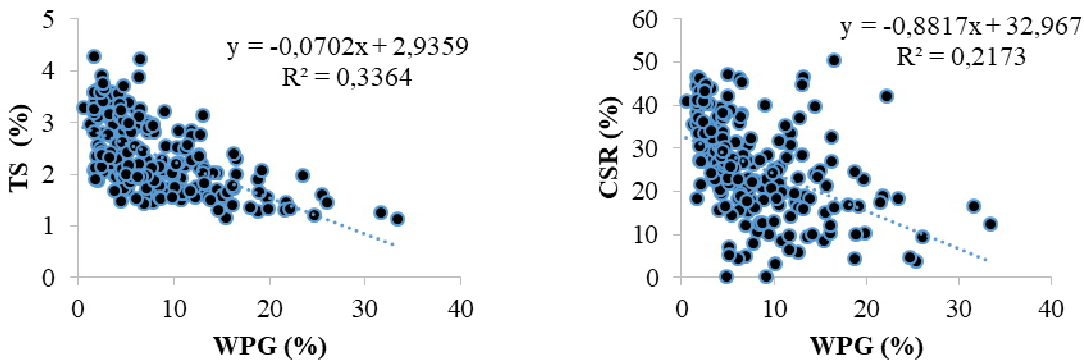

Fig. 6: Correlation between WPG and TS as well as between WPG and CSR.

CONCLUSIONS

The compregnation using LmwPF and SA as impregnating agents can significantly increase the specific gravity (SG) and dimensional stability of the wood. SG of LmwPF- and SA-compregnated wood increased by 10.69–22.31% and 6.96–23.09%, respectively. Application of a higher level of compression ratio will result in better wood characteristics. For 20 and 40% compression ratios, the use of LmwPF and SA as impregnating agents can significantly reduce the spring-back especially the compression-set recovery (CSR) and thickness swelling (TS) of wood; or in other words, dimensional stability of the wood is significantly improved. CSR after compregnation was 18.34–33.99%, while after densification was 47.86–71.49%. SA-compregnated wood has lower CSR and TS than LmwPF-compregnated wood. Increasing the concentration of the impregnating solution will result in a greater decrease in CSR and TS. The SG and dimensional stability of sepetir-compregnated wood are better than those of nyatoh and pisang putih wood.

ACKNOWLEDGMENTS

The authors are grateful for the support of the Ministry of Research, Technology and Higher Education of the Republic of Indonesia through PMDSU No. 1053/IT3.11/LT/2017.

REFERENCES

1. Augustina, S., Wahyudi, I., Darmawan, W.I, Malik, J., Basri, E., Kojima, Y., 2020: Specific gravity and dimensional stability of boron–densified wood on three lesser–used species from Indonesia. Journal of Korean Wood Science and Technology 48(4): 458–471.
2. Bao, M., Huang, X., Jiang, M., Yu, W., Yu, Y., 2017: Effect of thermos–hydro–mechanical densification on microstructure and properties of poplar wood (*Populus tomentosa*). Journal of Wood Science 63(6): 591–605.
3. Bao, M., Huang, X., Zhang, Y., Yu, W., Yu, Y., 2016: Effect of density on the hygroscopicity and surface characteristics of hybrid poplar compreg. Journal of Wood Science 62: 441–541.
4. Chen, Y., Nichole, M., Stark, Zhiyoung, C., Charles, R., Frihart, Linda, F., Lorenz, R.E.I., 2014: Chemical modification of kraft lignin: effect on chemical and thermal properties. Bioresources 9(3): 5488–5500.
5. Darwis, A., Wahyudi, I., Dwianto, W., Cahyono, T.D., 2017: Densified wood anatomical structure and the effect of heat treatment on the recovery of set. Journal of Indian Academy of Wood Science 14(1): 24–31.
6. Dos-Santos, P.C., Del-Menezzi, C.H.S., 2017: Effect of the densification process on properties of commercial oriented strand boards. European Journal of Wood and Wood Products 76: 1707–1713.
7. Furuta, Y., Aizawa, H., Yano, H., Norimoto, M., 1997: Thermal-softening properties of water-swollen wood: IV. Effects of chemical constituents of the cell wall on the thermal-softening properties of wood. Mokuzai Gakkaishi 43(9): 725–730.
8. Gabrielli, C.P., Kamke, F.A., 2010: Phenol-formaldehyde impregnation of densified wood for improved dimensional stability. Wood Science and Technology 44(1): 95–104.
9. Ghorbani, M., Bavaneghi, F., 2016: Effect of press cycle time on application behavior of board made from chemically modified particles. Drvna Industrija 67(1): 25–31.
10. Hill, C.A.S., 2006: Wood modification: Chemical, thermal, and other processes. Chichester. John Wiley and Sons. Sussex. 249 pp.

11. Inoue, M., Norimoto, M., Otsuka, Y., Yamada, T., 1991: Surface compression of coniferous wood lumber II: Permanent set of compression wood by low molecular weight phenolic resin and some physical properties of the products. Mokuzai Gakkaishi 35(3): 227–233.
12. Kajita, H., Furuno, T., Imamura, Y., 2004: The modification of wood by treatment with low molecular weight phenol-formaldehyde resin: a properties enhancement with neutralized phenolic resin and resin penetration into wood cell walls. Wood Science and Technology 37(5): 349–361.
13. Kamke, F.A., Kutnar, A., 2010: Transverse compression behavior of wood in saturated steam at 150-170°C. Wood and Fiber Science 42(3): 377–387.
14. Kelley, S.S., Timothy, G.R., Glasser, W.G., 1987: Relaxation behavior of the amorphous components of wood. Journal of Materials Science 22(2): 617–624.
15. Khalil, A.H.P.S., Dungani, R., Mohammed, I.A., Hossain, S.Md., Aprilia, S.N.A., Budiarto, E., Rosamah, E., 2014: Determination of the combined effect of chemical modification and compression of Agathis wood on the dimensional stability, termite resistance, and morphological structure. Bioresources 9(4): 6614–6626.
16. Kong, L., Zhao, Z., He, Z., Yi, S., 2017: Effect of steaming treatment on crystallinity and glass transition temperature of *Eucalyptuses grandis* x *E. urophylla*. Results in Physics 7: 914–919.
17. Kutnar, A., Sernek, M., 2007: Densification of wood. Zbornik Gozdarstva In Lesarstva 82: 53–62.
18. Kutnar, A., Kamke, F.A., Sernek, M., 2009: Density profile and morphology of viscoelastic thermal compressed wood. Wood Science and Technology 43(1): 57–68.
19. Lahtela, V., Karki, T., 2014: Effects of impregnation and heat treatment on the physical and mechanical properties of Scots pine (*Pinus sylvestris*) wood. Wood Material Science and Engineering 11(4): 217–227.
20. Lykidis, C., Kotrotsiou, K., Tsichlakis, A., 2019: Reducing set-recovery of compressively densified poplar wood by impregnation–modification with melamine–formaldehyde resin. Wood Material Science and Engineering 15(5): 1–9.
21. Miyoshi, Y., Furutani, M., Furuta, Y., 2016: Swelling behavior of cells in compressed wood. Journal of the Society of Materials Science 65(5): 343–346.
22. Morsing, N., Hoffmeyer, P., 1998: Densification of wood: The influence of hygrothermal treatment on compression of beech perpendicular to gain. Technical University of Denmark. Kgs. Lyngby, 138 pp.
23. Nairn, J.A., 2005: Numerical simulation of transverse compression and densification in wood. Wood and Fiber Science 38(4): 576–591.
24. Navi, P., Sandberg, D., 2012: Thermo hydro mechanical processing of wood. EPFL Press. Lausanne, 280 pp.
25. Nur, I.F.A., Zaidon, A., Rabia'tol, A.M.A., Bakar, E.S., Paridah, M.T., Mohd, H.S., Anwar, U.M.K., 2011: Enhancing the properties of low-density hardwood *Dyera costulata* through impregnation with phenolic resin admixed with formaldehyde scavenger. Journal of Applied Sciences 11: 3474–3481.

26. Ohlmeyer, M., Paul, W., 2010: Optimization of the properties of wood materials by means of thermal modification methods (Work Report No. 2010/2). Hamburg, Germany, 228 pp.
27. Pelit, H., Budakçi, M., Sönmez, A., 2016: Effect of heat post-treatment on dimensional stability and water absorption behaviours of mechanically densified Uludağ fir and Black poplar woods. Bioresources 11(2): 3215–3229.
28. Pelit, H., Sonmez, A., Budakci, M., 2014: Effect of Thermowood® process combined with thermo-mechanical densification on some physical properties of Scot pine (*Pinus sylvestris* L.). Bioresources 9(3): 4552–4567.
29. Pelit, H., Yalçin, M., 2017: Resistance of mechanically densified and thermally post–treated pine sapwood to wood decay fungi. Journal of Wood Science 63(5): 514–522.
30. Rautkari, L., Laine, K., Laflin, M., Hughes, M., 2011: Surface modification of Scots Pine: the effect of process parameters on the through thickness density profile. Journal of Materials Science 46(14): 4780–4786.
31. Sadatnezhad, H.S., Khazaeian, A., Sanberg, D., Tabarsa, T., 2017: Continuous surface densification of wood: A new concept for large-scale industrial processing. Bioresources 12(2): 3122–3132.
32. Scarica, C., Suriano, R., Levi, M., Turri, S., Griffini, G., 2018: Lignin functionalized with succinic anhydride as building block for biobased thermosetting polyester coating. ACS Sustainable Chemical & Engineering 6(3): 3392–3401.
33. Shams, M.I., Kagemori, N., Yano, H., 2006: Compressive deformation of wood impregnated with low molecular weight phenol formaldehyde (PF) resin IV: species dependency. Journal of Wood Science 52: 179–183.
34. Shams, M.I., Yano, H., Endou, K. 2004: Compressive deformation of wood impregnated with low molecular weight phenol formaldehyde (PF) resin I: effects of pressing pressure and pressure holding. Journal of Wood Science 50(4): 337–342.
35. Tabarsa, T., Chui, Y.H., 2000: Characterizing microscopic behavior of wood under transverse compression. Part II. Effect of species and loading direction. Wood and Fiber Science 33(2): 223–232.
36. Tu, D., Su, T., Zhang, T., Fan, W., Zhou, Q., 2014: Thermo-mechanical densification of *Populus tomentosa* ver. *Tomentosa* with low moisture content. Bioresources 9(3): 3846–3856.
37. Wan, H., Kim, M.G., 2006: Impregnation of southern pine wood and strands with low molecular weight phenol-formaldehyde resins for stabilization of oriented strand boards. Wood and Fiber Science 38(2): 314–324.
38. Zhao, Y., Wang, Z., Iida, I., Huang, R., Lu, J., Jiang, J., 2015: Studies on pre-treatment by compression for wood drying I: effect of compression ratio, compression direction and compression speed on the reduction of moisture content in wood. Journal of Wood Science 61(2): 113–119.

IMAM WAHYUDI*, WAYAN DARMAWAN
IPB UNIVERSITY
FACULTY OF FORESTRY AND ENVIRONMENT
DEPARTMENT OF FOREST PRODUCTS
JALAN ULIN KAMPUS DARMAGA
BOGOR 16680
INDONESIA
*Corresponding author: imyudarw16@yahoo.com

SARAH AUGUSTINA
IPB UNIVERSITY
FOREST PRODUCTS SCIENCE AND TECHNOLOGY STUDY PROGRAM
JALAN ULIN KAMPUS DARMAGA
BOGOR 16680
INDONESIA

JAMALUDIN MALIK
FOREST PRODUCTS RESEARCH AND DEVELOPMENT CENTER
JALAN GUNUNG BATU N^{O}. 5
BOGOR 16610
INDONESIA

NAOKI OKANO, OKADA TAIYO, HIKARU KOBORI, YOICHI KOJIMA,
SHIZUOKA UNIVERSITY
FACULTY OF AGRICULTURE
DEPARTMENT OF WOOD BIOMASS UTILIZATION
836 OHYA, SURUGA-KU, SHIZUOKA 422-8529
JAPAN

KAZUSHIGE MURAYAMA
FORESTRY AND FOREST PRODUCTS RESEARCH INSTITUTE
DEPARTMENT OF WOOD-BASED MATERIALS
1 MATSUNOSATO, TSUKUBA, IBARAKI 305-8687
JAPAN

SHIGEHIKO SUZUKI
SHIZUOKA PROFESIONAL UNIVERSITY OF AGRICULTURE
678-1 TOMIGAOKA, IWATA, SHIZUOKA 438-0803
JAPAN