Improving the Physical Properties of Young Teak Wood through Phenol Formaldehyde Compregnation

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Abstract

Young teak wood exhibits inferior properties due to a high proportion of juvenile wood and sapwood. The modification through compregnation of phenol formaldehyde is required to improve wood qualities. Therefore, this study aimed to observe the possibility of improving the physical properties of young teak wood (15 years) using phenol formaldehyde compregnation at various concentrations (5%, 10%, 15%) and pressing times (15, 30, 60 min). The results showed that phenol formaldehyde concentration produced retention with a maximum value of 31.19 kg/m^3. Based on wood without treatment (controls), the compregnation significantly increased the redness (a*) level by 45.57% and the specific gravity by 7.93%. The decreasing levels after treatment were observed in the brightness (L*) (by 36.56%), the yellowness (b*) (by 38.40%), and air-dried moisture contents by (by 5.44%). Furthermore, the reduction in hygroscopicity was observed in an equilibrium moisture content level in various relative humidity, as well as in increasing the stability dimension, though in a small magnitude.

Keywords: wood modification, pressure, higroscopicity, anisotropy, *Tectona grandis* L.f.

Introduction

The superior characteristics of teak wood include a good physical appearance and weathering resistance. However, young teak wood exhibits inferior properties due to the high content of sapwood and juvenile wood (Hidayati *et al.* 2015). Compared to mature wood, the juvenile has high proportion of early wood which causes lower density and strength. Furthermore, juvenile wood has high microfibril angles in the S2 layer of the cell wall. This form influences high longitudinal shrinkage and reduced transverse shrinkage, causing low dimensional stability (Shmulsky and Jones 2011).

Wood can be modified by physical and chemical methods or a combination of both. Wood modification is carried out through impregnation techniques using chemicals, heat densification, and pressure. There are several modification processes by impregnation based on various resin systems (Hill 2009). The modified wood products have a higher density than the original wood through a densification process where wood is impregnated with resin and compacted under pressure (compregnation). Phenol formaldehyde (PF) resins are commonly utilized in the wood industries as adhesives. Furthermore, phenol formaldehyde has several advantages in improving wood quality (Way *et al.* 2011; Ashaari *et al.*2015; Ang *et al.* 2014; Franke *et al.* 2017). Phenol formaldehyde resin impregnation and compression at high pressures significantly increase the strength of wood, especially flexural resistance, dimensional stability, and biological durability (Shams and Yano 2008; Nabil *et al.* 2016; Hartono *et al.* 2016). Therefore, this paper discussed on the improvement the physical properties of young teak wood by compregnation treatment using phenol formaldehyde.

Materials and Methods

Sample Preparation

The materials used in this study included three teak trees of 15 years old felled from Madiun Forest Management Unit, Perum Perhutani plantation. These trees were from thinning activities in which a total tree height of ± 6 m and diameter breast height of ± 13 cm. Sapwood samples were cut at the base part and sawn across the trunk with a length of approximately 1 m. Afterwards, the logs were sawn into boards in a flat sawn pattern. The boards were converted into specimens with a size of 2.5 × 2.5 × 20 cm for compregnation treatment and untreated (control). From these specimens, samples for moisture content (MC) and specific gravity (SG) (2 × 2 × 2 cm) as well as dimension stability (2 × 2 × 4 cm) were also taken according to British Standard (BSI 1957). Each sample was then coated with a flinkote paint on the cross-section of the wood to prevent compregnant substances from entering longitudinally. The test sample was then air-dried until it reached a MC of 12~15%. After the air-dry condition was achieved, the dimensions and initial weight of the sample were measured. The three individual trees were designed as replications.

Compregnation Process

The phenol formaldehyde (PA-302, PT. Pamolite Adhesive Industry, Probolinggo, Indonesia) was in liquid form with a solid resin content (RS) of 42% and molecular weight of 4000 Mn. The concentration of phenol formaldehyde solution (PF) was determined by the ratio of weight (w/w) of 5, 10, and 15%. Those levels were achieved by diluting PF resin with distilled water (formula $M_1V_1 = M_2V_2$). Furthermore, the compregnation process was preceded by air-drying the samples until 12~15% MC was reached. The initial weight was measured and the samples were then grouped according to the treatments. The test sample and PA solution were filled into the compregnator tank (Figure 1). The initial vacuum of 50-60 cmHg was applied for 15 min followed by pressing (10 kg/cm^2) for 15, 30, and 60 min. After the specified time was reached, the PF solution was removed and a final vacuum of 50-60 cmHg was applied for 15 min. The test sample was then air-dried and weighed.

Figure 1. Compregnation tank

Determination of Color Properties

Measurements were conducted using a spectrophotometer NF777 (Nipon Denshoku Co. Ind. Ltd., Japan). Air-dried woods were measured after compregnation three times for each sample at a different position. Three values were obtained including L* (brightness) with a scale of 0 (black) ~100 (white), a* (redness) with a (+) scale for red and (-) for green, and b* (yellow) with a (+) scale for yellow and (-) for blue.

Determination of Retention and Physical Properties

The retention was calculated from the dry weight of PF solution retained after treatment in a given volume of treated wood (kg/m^3). Measurements were carried out by weighing the wood after compregnation, minus the initial weight before. The MC was determined by oven method and SG by water displacement method (Marsoem *et al.* 2014). Furthermore, measurement of wood shrinkage was carried out in tangential, radial, and longitudinal directions from the condition of the wet MC to the oven-dried content. Anti-shrinkage efficiency (ASE) of each treatment variation was calculated using the following formula:

$$ASE\ (\%) = 100 \times [(Su - St)/Su] \quad (1)$$

Where Su = volumetric shrinkage for untreated wood; St = volumetric shrinkage for treated wood.

Determination of Reduced Equilibrium Moisture Content

The test samples were measured for dimensions and weighed. Different relative humidities were achieved using saturated salt solutions (Young 1967). The test samples in three replications of each parameter were placed in a different desiccator on top of a cup containing ammonium sulfate, magnesium chloride, and potassium hydroxide to produce relative humidity (RH) of 81, 33, and 8% at a temperature of 25°C, respectively. The test samples were then weighed continuously until a constant weight was obtained. Afterwards, the test samples were dried at a temperature of 103 ± 2°C to determine the equilibrium moisture content (EMC). The percentage reduction in equilibrium moisture at saturation:

$$EMC_R\ (\%) = 100 \times [(EM_u - EMC_t)/EMC_u] \quad (2)$$

Where EMC_R = reduction in equilibrium moisture content; EMC_u = equilibrium moisture content for untreated wood; EMC_t = equilibrium moisture content for treated wood.

Statistical Analysis

The effects of PF concentration and pressing time arranged in factorial design were calculated using analysis of variance (ANOVA) GLM procedures. The effects were taken into account only when significant at the 95% level using Type III Sums of Squares. Furthermore, the Tukey (Honestly Significant Difference) test was used to show further significant differences between groups. All statistics were performed using SPSS 12.0 software.

Results and Discussion

Retention Values

By ANOVA, it was observed that there was no significant interaction between the concentration and the pressing time factors (Table 1). The concentration factor affected all parameters except for longitudinal shrinkage and EMC at RH 33%. The pressing time factors had a significant effect only on retention and redness values. Retention is a measure of the concentration of preservative retained after treatment in a specified assay zone. The retention values obtained from the compregnation treatment using PF ranged from 13.47 to 31.19 kg/m^3 while the average highest value was obtained by a concentration of 15% and a pressing time of 30 minutes.

PF impregnation has been proven to increase the durability of the wood against termites or fungi attacks (Gascon-Garrod *et al.* 2015; Nabil *et al.* 2016). This experiment will be followed by bio-assay test to evaluate the PF resin act as a preservative in the next report. The retention value exceeded the minimum required for wood utilization for preservatives i.e. the equivalent of 5.30~15.89 kg/m^3 (Hunt and Garratt 1986). The increasing PF concentration and pressing time also caused an increase in the average retention amount (Figure 1). However, an increase in concentration of 10 to 15% and pressing time from 30 to 60 minutes did not show any significant differences. This is presumably due to the solution becoming saturated in the wood particularly in the lumen.

Table 1. The analysis of variance of of phenol formaldehyde concentration and pressing time effects

Properties	Source of variation		
	Concentration (A)	Pressing duration (B)	A x B
Retention	<0.01**	<0.01**	0.10
Specific gravity	0.01*	0.39	0.73
Air-dry moisture content	0.03*	0.01*	0.23
Longitudinal shrinkage	0.21	0.97	0.99
Radial shrinkage	0.04*	0.60	0.91
Tangential shrinkage	<0.01**	0.24	0.08
EMC–RH8–air-dry	<0.01**	0.49	0.86
EMC–RH8–wet	<0.01**	0.37	0.45
EMC–RH33–air-dry	0.40	0.73	0.74
EMC–RH33–wet	0.02*	0.17	0.41
EMC–RH81–air-dry	<0.01**	0.19	0.72
EMC–RH81–wet	<0.01**	0.68	0.43
Brightness (L*)	<0.01**	0.50	0.29
Redness (a*)	<0.01**	0.01*	0.51
Yellowness (b*)	<0.01**	0.56	0.43

Remark: SG = specific gravity; AD-MC = air-dry moisture content; RH = relative humidity; EMC = equilibrium moisture content

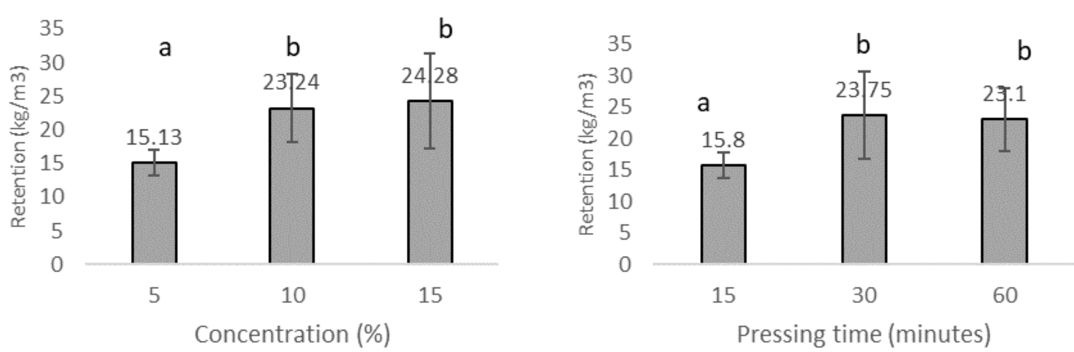

Figure 1. The effect of phenol formaldehyde concentration and pressing time on retention value. Mean of three trees, with the standard deviation error bar. The same letters are not statistically different at $P < 0.05$ by Tukey's test.

Color Properties

Color properties constituted supporting data to determine the effect of the compregnation treatment on young teak. Compregnation using PF caused the appearance of the wood to be darker, similar to the mature wood (Figure 2). The wood color was permanently changed and the solution thinly penetrated into the wood monitored by planing the surfaces. Unfortunately, the distance was not measured to obtain the actual penetration. Furthermore, increasing the concentration of PF up to 15% significantly decreased the values of L* (by 36.56%) and b* (by 38.40%) but increased the values of a* (by 45.57%) from the percentage of control values (Table 2).

PF resin gives red-brown color due to change in the pH of the wood/resin (Furuno *et al*. 2004; Kielmann *et al.* 2018) which is not occured for urea formaldehyde resin or poly-ethylene glycol treatment. The presence of PF had the same effect as extractives in heartwood where the greater the amount, the greater the color change. The increase in pressing time up to 30 min increased the a* value, signifying a more intensive reaction that resulted in the extractives in teak wood to have a reddish color. Teak extractives consisted quinone group with varying colors particulalrly in the heartwood part (Lukmandaru and Ogiyama 2005; Romagnoli *et al.* 2013). Therefore, further research is necessary to explore teak extractive compounds that experience color changes due to phenol formaldehyde impregnation in the sapwood part.

Figure 2. The appearance of teak wood control samples (brighter specimens) and after compregnation treatment (darker specimens) with phenol formaldehyde resin.

Table 2. The color properties of teak wood with compregnation treatment (phenol formaldehyde resin) (mean of three trees).

Concentration (%)	Pressing time (minutes)	L* (brightness)	a* (redness)	b* (yellowness)
5	15	45.2	12.5	14.6
	30	45.4	13.0	14.8
	60	46.6	13.0	13.7
10	15	41.5	13.7	11.9
	30	41.1	14.8	11.5
	60	39.9	15.1	11.7
15	15	34.2	14.6	9.8
	30	34.9	15.1	10.1
	60	32.2	14.9	10.0
Control		63.3	9.7	19.5

Specific Gravity and Moisture Content

The obtained wood SG of the study ranged from 0.61 to 0.68 (Table 3). Compared to the control (0.63), the highest improvement was achieved with a concentration of 10% and a pressing time of 15 min (0.68), although the increase in SG values only reached 7.93%. The impregnation PF in the parenchyma of the wood surface affected the SG increase (Sumardi *et al*. 1999). This relatively low value in this study indicates a few PF substances are able to penetrate into the cell wall structure. It is thought that the comparatively high initial density (0.63) of teak wood in this experiment partially affects the low penetreation rate. In earlier reports, the low initial specific gravity values in other species (0.5>) such as oil palm trunk (Sumardi *et al.* 1999; Aini *et al.* 1999; Hartono *et al.* 2016), sesenduk and jelutong (Adawiah *et al.* 2012; Ashaari *et al.* 2015) showed drastic increases in density levels. Another possibility is the unsuitable molecular weight of PF in this experiment (4000 Mn). PF (low molecular weight) with a concentration of 20% and hot pressing successfully increased wood density from 0.45 to 1.1 g/cm^3 in

Cryptomeria japonica wood (Shams *et al.* 2004). The effect of molecular weight of PF resin has been addressed by several researchers (Furuno *et al*. 2004; Rowell 2005; Aizat *et al.* 2017)

Wood is a hygroscopic material that adsorbs and desorbs water in liquid or vapor form from the atmosphere. The higher is the temperature or the lower the RH of the air, the lower is the EMC and vice versa. It is expected that the modified wood had a lower air-dried MC when compared to the control. It is also assumed that the decrease in MC was due to the presence of PF as a bulking agent that filled the wood cells. The air-dried MC of the treatment produced a range of 14.24~17.23% (Table 3). The lowest value was observed in the compregnation with a concentration of 5% and a pressing time of 15 minutes (14.24%) or a decrease of only 5.44% of the control value (15.60%).

Tabel 3. The specific gravity and moisture content of teak wood with compregnation treatment (phenol formaldehyde resin) (mean of three trees).

Concentration (%)	Pressing time (minutes)	SG	AD-MC (%)	Equilibrium moisture content (%)					
				RH8%-AD	RH8%-Wet	RH33%-AD	RH33%-Wet	RH81%-AD	RH81%-Wet
5	15	0.61	14.24	6.50	6.51	10.44	10.50	22.40	27.65
	30	0.61	14.60	6.46	6.64	10.24	10.75	22.25	26.31
	60	0.62	15.80	6.43	6.61	10.27	10.80	23.78	28.44
10	15	0.68	14.89	6.53	6.80	10.29	10.75	25.31	29.92
	30	0.63	17.23	6.64	6.74	10.62	11.06	28.02	30.73
	60	0.65	16.20	6.54	6.69	10.40	10.88	27.65	32.00
15	15	0.67	15.80	6.29	6.47	10.27	10.66	26.09	28.62
	30	0.65	16.13	6.23	6.53	10.28	10.68	28.11	30.60
	60	0.64	15.73	6.09	6.34	10.09	10.49	27.14	28.46
Control		0.63	15.60	6.33	6.51	10.30	10.80	24.11	30.21

Remark: SG = specific gravity; AD-MC = air-dry moisture content; RH = relative humidity.

At 8% and 33% RH, wood conditioned at the initial air-dried MC did not experience the mechanism of water adsorption from the surrounding environment (adsorption). On the other hand, the wood released water into the air (desorption) until it reached a state of equilibrium. For wood conditioned at different RH, a 5% concentration also showed the highest decrease in EMC at 81% RH (7.71~12.90%) but less significant at 8 and 33% RH. Furthermore, the addition of more than 5% concentration produced a higher EMC level than the control wood. The same result was also observed by Sumardi *et al.* (1999) and Aini *et al.* (1999) in oil palm wood. It is probable that in the wood treated initially with air-dried MC, some of the hygroscopic properties were reduced due to the previous drying process or hysteresis. Technically, resins are deposited in the wood cell walls extensively to reduce swelling on water immersion (Furuno *et al.* 2004). The ineffectiveness of PF in reducing the hygroscopicity of wood is assumed to be due to the few amount of PF that penetrated the cell wall, although it showed a high retention level. This is represented by the relatively small increase in SG values.

Dimensional Stability

The shrinkage parameters observed in this study included three main fiber directions of wood, i.e. longitudinal, radial, and tangential, as well as the value of ASE. The value of wood shrinkage observed was from wet to oven-dried conditions. Furthermore, the concentration factor significantly influenced the wood shrinkage in the radial and tangential directions. The compregnation treatment at a concentration of 5% showed a lower shrinkage value than the control wood and was more effective than other higher concentrations. This longitudinal shrinkage value obtained was in the range of 0.33~0.64%. A comparatively high value indicates the presence of juvenile wood (Shmulsky and Jones 2011). PF compregnation produced a lower shrinkage value than the control, with a percentage decrease of 11.86%; 3.73%; and 7.87% in the longitudinal; radial, and tangential directions, respectively.

The impregnation of phenol resin on wood increases its dimensional stability (Kollmann *et al.* 1975; Rowell 2005; Aizat *et al.* 2017). The penetration into the cell walls are the decisive factor for dimensional stability of the treated wood (Franke *et al.* 2017).This occurs through the penetration and enlargement of wood cell walls. The compregnation of PF reduced the shrinkage value of wood with the best treatment at a concentration of 5%. This agrees with previous studies on rubber wood (*Hevea brasiliensis* Muell Arg.) where compregnation with urea formaldehyde at a concentration of 5% produced the best effect on the physical and mechanical properties (Wedatama *et al.* 2014).

The effectiveness of the compregnation to stabilize wood dimensions was assessed through ASE values. Overall, it was observed that the highest ASE value was produced by a concentration factor of 5% alongside a pressing time of 15 min (43.85%). The lowest ASE value was produced by a concentration factor of 15% and a pressing time of 15 min. It is also noticed that the ASE values were less than 0% in several 10% concentration and 30-minute treatments as well as 15% concentration and 15- or 30-min treatments. For uncertain reason, it indicates those treatments caused less hydrophobic of the wood. The compregnation treatment was quite effective on the cross-section of the wood as evidenced by the higher ASE value in the longitudinal direction, compared to the radial and tangential. This is assumed to be due to the presence of large vessel cells for conducting the solution in a vertical direction. However, the value of anti-shrinkage efficiency (ASE) due to the PF compregnation treatment was still relatively low (50%>) to increase the dimensional stability of young teak wood. This is presumably because the type of PF used had a large molecular size to penetrate the cell wall. PF resin is deposited on the lumen surfaces (Hosseinpourpia *et al.* 2016). PF resin might bulk in cell wall while mostly the resin fill the lumen. Scanning electron microscope analysis will be helpful to confirm the inclusion of PF in the wood structure which causes different physical properties.

Tabel 4. The shrinkage and anti-shrink effficiency of teak wood with compregnation treatment (phenol formaldehyde resin) (mean of three trees).

Concentration (%)	Pressing time (minutes)	Shrinkage (%)			Anti-shrink effficiency (%)		
		Longitudinal	Radial	Tangential	Longitudinal	Radial	Tangential
5	15	0.33	2.67	4.04	43.85	17.25	36.42
	30	0.43	2.65	4.23	27.17	17.80	33.33
	60	0.34	2.62	5.22	42.89	18.54	17.87
10	15	0.54	3.17	5.63	8.71	1.66	11.31
	30	0.54	3.36	8.09	8.40	-4.20	-27.39
	60	0.61	3.10	6.25	-3.99	3.72	1.58
15	15	0.63	3.66	6.66	-7.62	-13.51	-4.88
	30	0.64	3.62	6.54	-8.93	-12.44	-3.05
	60	0.58	3.02	5.99	0.58	6.15	5.73
Control		0.59	3.22	6.35			

Conclusions

The physical properties of the teak sapwood had been improved by the PF treatment (concentration and pressing time). The maximum retention value was produced at a concentration of 10% and a pressing time of 30 min (31.19 kg/m^3) which exceeded the minimum required for wood utilization for preservatives. The increase in the SG value of the control reached 7.93% by a concentration of 10% and a pressing time of 15 min (0.63 to 0.68). The compregnation treatment with a concentration of 5% produced the best results on hygroscopic properties and dimensional stability, although the decrease was not as high as expected, while the concentration of 15% caused a drastic change in color properties. Pressing time only affected significantly air-dried MC and redness (a*) values. Therefore, the compregnation of phenol formaldehyde in young teak wood with the lower molecular weight resin for maximizing penetration into the cell wall of the wood should be explored as a subsequent study.

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