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Original Article

# Improvement of Fast-Growing Wood Species Characteristics by MEG and Nano $SiO_2$ Impregnation[1]

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**ABSTRACT**

Jabon (*Anthocephalus cadamba*) is a fast-growing wood species that is widely utilized for light construction and other purposes in Indonesia. The objectives of the current study were to determine the effects of monoethylene glycol (MEG) and $SiO_2$ nanoparticles (nano $SiO_2$) impregnation treatment on the dimensional stability and density of jabon wood and to identify the characteristics of impregnated jabon wood. Wood samples were immersed in water (as untreated), MEG, 0.5% MEG$SiO_2$, then impregnated by applying 0.5 bar of vacuum for 60 min, and then applying 2.5 bar of pressure for 120 min. The results showed that impregnation with MEG and Nano $SiO_2$ had a significant effect on the dimensional stability of jabon wood. Polymers can fill cell walls in wood indicated by increasing weight percentgain, antiswelling efficiency, bulking effect, and density, then decreasing in water uptake value. Jabon wood morphology by using SEM showed that MEG$SiO_2$ polymers can cover part of the pitsin the wood vessel wall of jabon. This finding was reinforced by EDX results showing that the silicon content was increased due to the addition of $SiO_2$ nano. The XRD diffraction pattern indicated that MEG$SiO_2$ treatment increased the degree of crystallinity in wood samples. Overall, treatment with 0.5% MEG$SiO_2$ led to the most improvement in the dimensional stability of 5-year-old jabon wood in this study.

***Keywords:*** impregnation, monoethylene glycol, nano $SiO_2$, jabon, density, dimensional stability

## 1. INTRODUCTION

The development of community forests has been one of the solutions for overcoming the shortage of raw material for wood industries in Indonesia. These forests areusually planted with fast-growing species, such as jabon (*Anthocephalus cadamba*). Jabon naturally grows not only in Indonesia, but also in Australia, China, India, Malaysia, Papua New Guinea, the Philippines, Singapore, and Vietnam (Orwa *et al.*, 2009). Jabon wood is widely used as wood packaging but it has numerous other potential applications, including raw material plywood, pulp and paper, building construction, and flooring. It has several drawbacks, however, namely low density (0.3 g/cm$^3$) (Lestari *et al.*, 2018), poor strength, and poor durability, which are primarily having a large proportion of juvenile wood. The large proportion of juvenile wood in jabon caused it can be

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harvested at 5,6, and 7 year-old (Martawijaya, 2005). In a previous study, Rahayu *et al.* (2014) found that 5 year-old jabon consists of 100% juvenile. Because the quality of juvenile wood is much lower than that of mature wood, it is not recommended for structural purposes (Bowyer *et al.*, 2007). However, its quality could be improved through technological modifications of its structure.

One method of wood modification is impregnation, a process through which a molecular compound is penetratedinto the wood structure and there by increases the density and hardness of the wood. Impregnation of organic and inorganic compounds into wood structure by soaking or using vacuum pressure tube can be performed to improve physical properties of wood (Hadi *et al.*, 2018; Hartono *et al.*, 2016; Oh and Park, 2015). Impregnation research on fast-growing wood species in Indonesia has been widely performed, using styrene (Darma *et al.*, 2002; Jasni *et al.*, 2004) and methyl methacrylate (MMA) (Wardani *et al.*, 2012; Hadi *et al.*, 2013, 2015), wood vinegar and an animal adhesive (Arsyad *et al.*, 2019), but little research has been done on impregnation with monoethylene glycol (MEG) and $SiO_2$ nanoparticles (nano $SiO_2$). MEG is a hygroscopic substance and can absorb twice its weight in water at 100% relative humidity (Budavari, 1989). Based on its perfect water-soluble characteristics and its relatively low cost, we selected MEG for use in this study. $SiO_2$ nanoparticles (Nano $SiO_2$) are single particles of silica dioxide, an inorganic metal oxide, with a diameter less than 100 nm. Nano $SiO_2$ has been used as a polymer to increase the strength and improve the properties of modified wood (Dong *et al.*, 2014). Nano particles resumably produce uniform distribution and penetration in wood, as well as providing low-viscosity properties (Fufa and Hovde, 2010). Impregnation of poplar (*Populus* spp.) wood with furfuryl alcohol (FA) and nano $SiO_2$ was shown to effectively improve the properties of the wood (Dong *et al.*, 2014). Further, Rahayu *et al.* (2019) found that impregnation of the wood of another fast-growing species, sengon (*Falcataria moluccana*), by MEG and nano $SiO_2$ enhanced its dimensional stability. In this study, we used MEG and nano $SiO_2$ impregnation technology to improve the quality of jabon wood.

The objectives of our research were to determine the effects of MEG and nano $SiO_2$ impregnation treatment on the dimensional stability and density of jabon wood to identify the characteristics of impregnated jabon wood.

## 2. MATERIALS and METHODS

### 2.1. Materials

Five-year-old jabon wood was originated from the community forest in Bogor, West Java, Indonesia. The wood was cut to sample sizes of 2 cm × 2 cm × 2 cm based on British Standard (BS) 373 (BS, 1957) for testing, which assessed weight percent gain (WPG), antiswelling efficiency (ASE), water uptake (WU), bulking effect (BE), and density. The materials used in this study were MEG and nano $SiO_2$ (diameter 15 ± 5 nm; Anhui Elite Industrial Co, Ltd, China).

### 2.2. Methods

The impregnation process began with drying samples at a temperature of 103 ± 2°C. Three treatment solutions were used in this study: water-treated as untreated; 50% MEG (vol/vol) in an aqueous solution; and 0.5% MEG$SiO_2$, which contained 0.5% (wt/vol) $SiO_2$ nanoparticles in 50% MEG solution (Table 1). A sonicator (Cole Palmer) was used to mix MEG and nano-$SiO_2$ at 40% amplitude for 60 min. The impregnation process was initiated with a vacuum of 0.8 atm for 60 minutes and followed by pressure at 2.5 bar for 120 minutes. The sample was wrapped with aluminum foil, kept at room temperature for 12 hours, and then dried in an

**Table 1.** Composition of MEG and nano $SiO_2$ solutions used for impregnation.

| Treatment | MEG (mL) | Water (mL) | Nano $SiO_2$ (g) |
|---|---|---|---|
| Untreated | 0 | 1000 | 0 |
| MEG | 500 | 500 | 0 |
| 0.5% $MEGSiO_2$ | 500 | 500 | 5 |

oven at 100°C for 12 hours. The polymerization process was then carried out. The sample was then immersed in water for 24 hours for ASE and WU test, followed by drying in an oven at 103 ± 2°C until it reached constant weight. The impregnation method is adopted from Dong *et al.* (2014) and Rahayu *et al.* (2019). Five samples were used to test each treatment.

We used the formulas for calculating WPG, WU, BE (Hill, 2006), and ASE (Rowell and Ellis, 1978):

$$WPG\ (\%) = (W_1 - W_0)/W_0 \times 100$$
$$WU = (W_2 - W_1)/W_1 \times 100$$
$$BE = (V_1 - V_0) / V_0 \times 100$$
$$ASE\ (\%) = (Su - St)/Su \times 100$$

Where,

$W_0$ = the initial oven-dried weight of wood sample
$W_1$ = the oven-dried wood sample weight after treatment
$W_2$ = the weight of the wood sample after immersion in water for 24 h
$V_0$ = the initial oven-dried volume of wood sample
$V_1$ = the oven-dried wood sample volume after treatment
Su = the volume shrinkage of untreated wood sample
St = the volume shrinkage of treated wood sample

The density (Bowyer *et al.*, 2007), $\rho$ , was calculated after treatment, using the WPG and BE values, and evaluation of treatment-related changes used the untreated density as the baseline.

Morphology of jabon wood was examined by SEM coupled with EDX and XRD to assess the characteristics of the wood. The capability of MEG and nano $SiO_2$ to penetrate inside cell wall was analyzed using SEM (Zeiss Evo 50 Type, Germany). Tangential sections of both treated and untreated samples (0.5 × 0.5 × 0.5 $cm^3$) were mounted on conductive adhesives, sputter-coated with gold, and observed by SEM at an accelerating voltage of 15 kV, with a chamber pressure between 10 and 400 Pa, under (low vacuum mode) and a working distance of 8 mm. The impregnated samples were also analyzed by SEM coupled with EDX (Zeiss Evo 50 Type, Germany) using a spectra mode to analyze the chemical content in untreated and treated jabon wood.

The degree of crystallinity in untreated and treated jabon wood was analyzed by XRD. Jabon samples were milled with a Willey mill into a 40–60 mesh powder, and the crystallinity of each sample was evaluated by XRD with an XRD-7000 diffractometer (Shimadzu Co., Ltd., Japan). The measurements were conducted with Cu Kα radiation with a graphite monochromator at a constant voltage of 40 kV and an electric current of 30 mA over a 2θ scanning range of 5°–40° at a scanning speed of 2° $min^{-1}$.

### 2.3. Data analysis

WPG, ASE, WU, BE, and density data were used in an analysis of variance (ANOVA) to characterize the effects of the water, MEG, and 0.5% $MEGSiO_2$ impregnation treatments. The mean differences between the treatments were determined using Duncan's multiple range test.

## 3. RESULTS and DISCUSSION

### 3.1. Dimensional stability and density of impregnated woods

The impregnation treatment with MEG and nano $SiO_2$ had a significant effect on WPG, ASE, BE, WU, and density as shown in Table 2. The ANOVA results

**Table 2.** Results of WPG, ASE, BE, WU, and density

| Treatment | WPG (%) | ASE (%) | WU (%) | BE (%) | Density (g/cm$^3$) |
|---|---|---|---|---|---|
| Untreated | 0$^a$ | 0$^a$ | 112.2 (±11.1)$^c$ | 2.2 (±0.5)$^a$ | 0.31 (±0.03)$^a$ |
| MEG | 46.8 (±10.1)$^b$ | 55.8 (±9.4)$^b$ | 64.4 (±4.0)$^b$ | 4.2 (±0.6)$^b$ | 0.44 (±0.04)$^b$ |
| 0.5% MEGSiO$_2$ | 56.7 (±3.3)$^c$ | 67.2 (±9.0)$^c$ | 31.7 (±6.1)$^a$ | 6.3 (±0.8)$^c$ | 0.46 (±0.04)$^b$ |

WPG, weight percent gain; ASE, antiswelling efficiency; WU, water uptake; BE, bulking effect.
$^{a-c}$ Values followed by the same letters show no real difference based on the Duncan test.

**Table 3.** F-values of variance analysis

| Source | F-value | | | | |
|---|---|---|---|---|---|
| | WPG | ASE | WU | BE | Density |
| Treatment | 109.79** | 88.00** | 116.16** | 44.28** | 26.36** |

WPG, weight percent gain; ASE, antiswelling efficiency; WU, water uptake; BE, bulking effect.
** $p < 0.05$; indicates that treatment factor had a significant effect at a 95% confidence level.

indicated that impregnation had a statistically significant effect on the WPG, ASE, BE, WU, and density ($p < 0.05$) (Table 3). The results of Duncan's multiple comparison test (Table 2) confirm that the WPG, ASE, BE, WU, and densityvalues of the treated samples were significantly different from those of the water- and MEG-treated sample. The WPG, ASE, BE, and density values of samples treated with 0.5% MEGSiO$_2$ were greater than those of samples treated with water or MEG. However, the 0.5% MEGSiO$_2$ samples had lower WU values than samples treated with water or MEG. These findings are similar to those of Dong *et al.* (2014), who found that the WPG of poplar treated with FA and nano SiO$_2$ was greater than that of wood treated with FA treated alone. Similar results was reported by Hartono *et al.* (2016), who concluded that higher WPG Oil Palm trunk after impregnation with phenol formaldehyde (PF) resulted in the increase of dimensional stability as shown by an increase of density. Wood density has a decisive effect on the physical and dynamic properties of wood (Lee and Lee, 2018; Zhang *et al.*, 2018). In accordance with Hill (2006), the findings suggest that impregnation of wood with a polymer causes to penetrate and strengthen cell walls. The strengtheningis believed to occur through the polymer filling the void space within the cell wall.

The increasing ASE values were followed by the increasing WPG values. The highest ASE value of 67.2% and the highest WPG value of 56.7% were achieved with 0.5% MEGSiO$_2$ treatment. The higher the ASE value, the less the wood is able to adsorb water vaporfrom the surrounding environment. Further, the addition of SiO$_2$ nano can replace the bound water that exists within the cell wall. This result is in line with a report by Deka *et al.* (2007), who concluded that ASE values increased as WPG rose in Norway Spruce after impregnation with melamine formaldehyde (MF), it might be due MF resin is blocking the OH-group within the cellulose chain.

The WU values of the samples treated with 0.5% MEGSiO$_2$ were the lowest among all treatments. This outcome shows that the ability of wood to adsorb water decreases with the addition of MEG and nano SiO$_2$. The decrease of water absorption indicates the reduction of wood hygroscopicity (Priadi *et al.*, 2019). Hill's (2006) suggestionthat if the cell wall is filled by a polymer then it will become stronger. A negative correlation was observed between WPG and WU, and

as WPG increased, WU decreased. This finding may have been due to the MEG and $MEGSiO_2$ filling the cell wall, minimizing the availability of space for water molecules.

The increase in the BE values suggests that adding $MEGSiO_2$ via the impregnation process can improve wood dimension stability. The samples treated with 0.5% $MEGSiO_2$ hadthe greatest BE values (6.3%). This finding indicated that the addition of MEG and Nano $SiO_2$ as bulking agents is could be a reliable means of improving wood quality. The increasing BE values were accompanied by increasing WPG and density values. According to Hill (2006) polymerentering cell walls and strengthening can improve dimensional stability, and the extent of this effect likely depends on the amount of bulking agnt used. Bowyer *et al.* (2007) further pointed out that the higher the wood density, the greater the wood content in the cell wall and the cell wall can be make thicker via artificial means, such as thorugh the use of the bulking agents tested in this study.

Previous studies have highlighted that improvements in dimensional stability and mechanical properties in poplar wood were achieved by reducing the porosity of the wood by filling void space with urea formaldehyde and nano $SiO_2$. These addition prevented moisture frompenetrating into wood (Jiang *et al.*, 2013).

## 3.2. Jabon wood morphology (SEM analysis)

SEM analysis revealed the morphology of untreated jabon wood, usingmagnification of ×250 (Fig. 1a) and ×500 (Fig. 1b). Figures 1a and 1b show empty spaces in the pits and cell lumens. With the addition of MEG, jabon wood underwentmorphological changes, as shown in Figure 1c. MEG appears to partially cover the cell wall in a scattered pattern (Fig. 1d). The morphological differences between untreated jabon wood and treated jabon wood can be seen clearly.

Figs. 1e and 1f show that MEG and nano $SiO_2$ penetrated into vessel walls and evenly adhered to and cover cell wall of jabon. The SEM analysis revealed the likely cause of the increases in the WPG, ASE, BE, and density values and the decrease in the WU value in the treated jabon wood. The images illustrate that the addition of nano $SiO_2$ can increase the MEG distribution, allowing the MEG to more readily enter, scatter, and even cover pits and vessel walls. Overall, the 0.5% $MEGSiO_2$ treatment was the most optimal treatment of the threepolymer treatments investigated.

## 3.3. Composition element (Analysis using SEM–EDX)

The chemical composition of wood samples treated with water, MEG, and $MEGSiO_2$ was determined by SEM-EDX, and the results are shown in Table 4. The untreated treatment and MEG resulted in no $SiO_2$ content. In contrast, in the wood samples impregnated with 0.5% $MEGSiO_2$ the silicon content of the wood samples reaching 3.03%. Thus,it can be concluded that there is silicon in the wood treated with $MEGSiO_2$.

## 3.4. XRD analysis

Fig. 2 shows the XRD spectra of the untreated jabon wood samples and the treated samples (MEG and 0.5% $MEGSiO_2$). The XRD wave peaks associated with the untreated treatment, MEG, and 0.5% $MEGSiO_2$ occurred respectivelyat an angle for $2\theta$ of 22.14°, 21.76°, and 22.08°. Furthermore, cellulose peaks were also detected at an angle for $2\theta$ = 15.78°, 15.84°, and 15.30° corresponding to the peak of cellulose in the JCPDS data base $I_{012}$ $2\theta$ = 22.6° and $I_{101}$ $2\theta$ = 17.13°.

Crystallinity is an important properties in polymers that exhibit bonds between molecular chains, resulting in a more orderly arrangement of molecules. In the Fig. 2 it can be seen that the peak of cellulose decreases

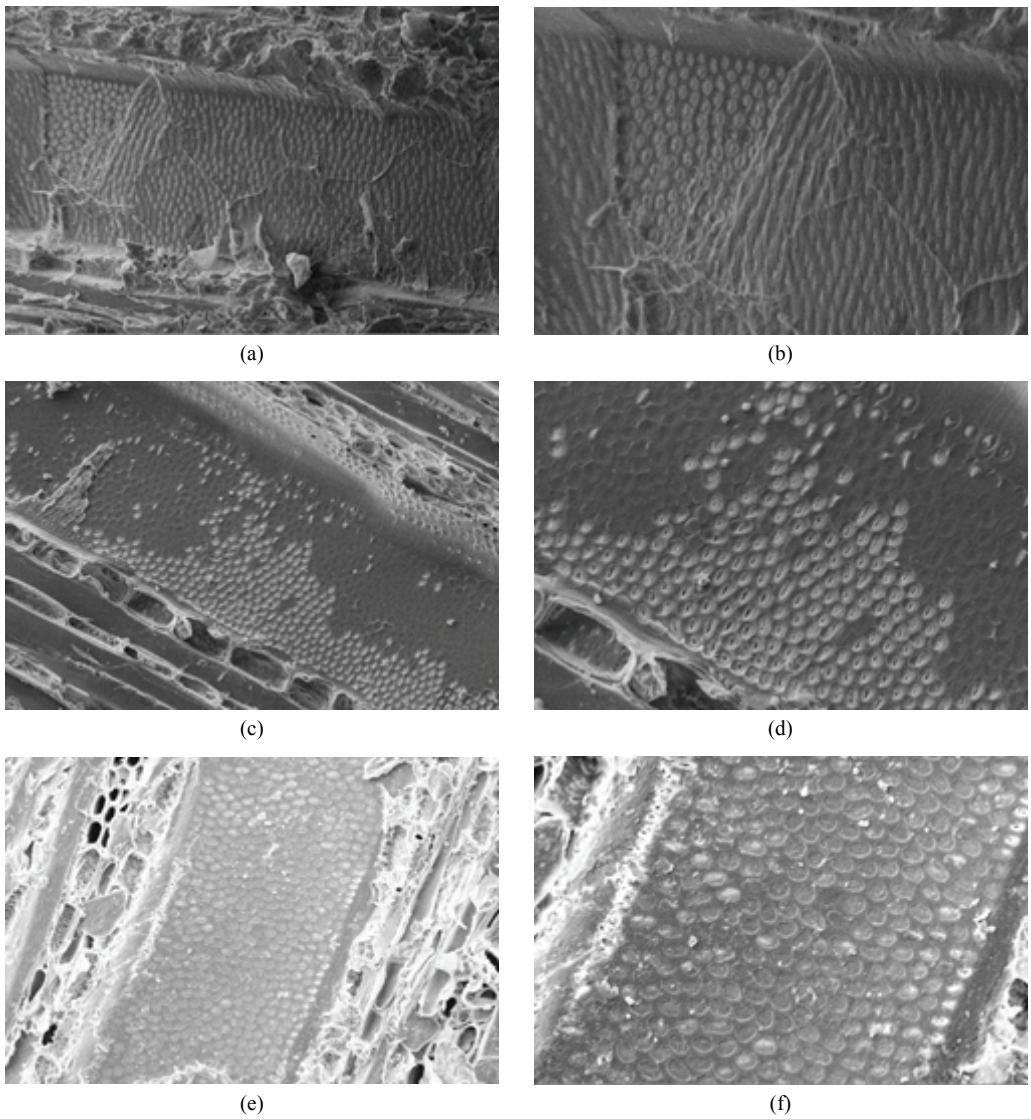


**Fig. 1.** (a) Jabon untreated morphology with 250x magnification (b) Jabon untreated morphology with 500x magnification (c) Jabon MEG morphology with 250x magnification (d) Jabon MEG morphology with 500x magnification (e) Jabon 0.5% $MEGSiO_2$ with 250x magnification (f) Jabon 0.5% $MEGSiO_2$ with 500x magnification.

with the addition of MEG and 0.5% $MEGSiO_2$. It is suspected that MEG and 0.5% $MEGSiO_2$ have an influence on the structure of the jabon wood. The addition of nano $SiO_2$ used has semi-crystalline characteristics that can increase the cristallinity. Similar results were previously reported by Dirna (2019), who found that the degree of crystallinity of jabon wood was increased through MEG and silica treatment. Similar results were shown in Rahayu *et al.* (2019) study, that MEG and $MEGSiO_2$ treatment can increased crystallinity of

**Table 4.** Chemical composition of wood sample of jabon on various treatment of MEG and nano $SiO_2$ impregnation

| Treatments | Carbon (wt. %) | Oxygen (wt. %) | Silicon (wt. %) |
|---|---|---|---|
| Untreated | 61.99 | 28.25 | 0 |
| MEG | 60.02 | 28.01 | 0 |
| 0.5% $MEGSiO_2$ | 55.29 | 31.77 | 3.03 |

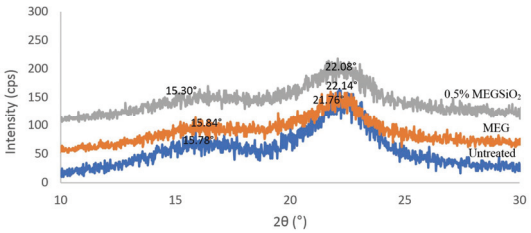


**Fig. 2.** XRD spectra jabon impregnated wood untreated, MEG, and 0.5% $MEGSiO_2$.

sengon wood.

The results of the XRD analysis show the effects of nano $SiO_2$ addition onWPG, ASE, WU, BE, and the resulting density. There effects are supported by the appearance of silicone peaks in the diffraction field that correspond to the ICDC database. The results show that the addition of $SiO_2$ nano increases the degree of crystallinity in the wood. High crystallinity indicates strong and rigid bonds between $MEGSiO_2$ polymers.

## 4. CONCLUSION

MEG and nano $SiO_2$ impregnation of the jabon wood had a significant effect on the resulting dimensional stability of the wood. Increases in WPG, ASE, BE, and density indicates that polymers can fill cell walls in wood, causing a decrease in WU value and a consequent reduction in the ability of jabon wood to adsorb water. Changes in jabon wood morphology with MEG treatment and $MEGSiO_2$ treatment relative to untreateds were demonstrated by SEM analysis results showing that $MEGSiO_2$ polymers can cover part of the pits in the wood vessel wall of jabon. This finding was reinforced by EDX results showing that the silicon content was increased due to the addition of nano $SiO_2$. The XRD diffraction pattern indicated that $MEGSiO_2$ treatment increased thedegree of crystallinity in wood samples. Overall, treatment with 0.5% $MEGSiO_2$ led to the most improvement in the dimensional stability of 5-year-old jabon wood in this sample.

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