Improvement of Fast-Growing Wood Species Characteristics by MEG and Nano SiO₂ Impregnation

Fitria Cita DIRNA² · Istie RAHAYU³,† · Lukmanul Hakim ZAINI³ · Wayan DARMAWAN³ · Esti PRIHATINI³

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 SiO₂ nanoparticles (nano SiO₂) 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% MEGSiO₂, 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 SiO₂ 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 MEGSiO₂ 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 SiO₂ nano. The XRD diffraction pattern indicated that MEGSiO₂ treatment increased the degree of crystallinity in wood samples. Overall, treatment with 0.5% MEGSiO₂ led to the most improvement in the dimensional stability of 5-year-old jabon wood in this study.

Keywords: impregnation, monoethylene glycol, nano SiO₂, 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 are usually 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/cm³) (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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¹ Date Received August 9, 2019, Date Accepted December 24, 2019
² Master student of IPB University, Bogor (16680), Indonesia
³ Department of Forest Products, Faculty of Forestry, IPB University, Bogor (16680), Indonesia
† Corresponding author: Istie RAHAYU (e-mail: istiesr@apps.ipb.ac.id, ORCID: 0000-0003-4902-564X)
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
penetrated into the wood structure and thereby 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., 2016; 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; Jasi 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
SiO2 nanoparticles (nano SiO2). 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. SiO2
nanoparticles (Nano SiO2) are single particles of silica
dioxide, an inorganic metal oxide, with a diameter less
than 100 nm. Nano SiO2 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 SiO2 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 SiO2 enhanced its
dimensional stability. In this study, we used MEG and
nano SiO2 impregnation technology to improve the
quality of jabon wood.

The objectives of our research were to determine
the effects of MEG and nano SiO2 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
The sample was wrapped with aluminum foil, kept at
room temperature for 12 hours, and then dried in an

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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):

\[
\text{WPG} (\%) = \frac{(W_1 - W_0)}{W_0} \times 100
\]
\[
\text{WU} = \frac{(W_2 - W_1)}{W_1} \times 100
\]
\[
\text{BE} = \frac{(V_1 - V_0)}{V_0} \times 100
\]
\[
\text{ASE} (\%) = \frac{(Su - St)}{Su} \times 100
\]

Where,
\[
W_0 = \text{the initial oven-dried weight of wood sample}
\]
\[
W_1 = \text{the oven-dried wood sample weight after treatment}
\]
\[
W_2 = \text{the weight of the wood sample after immersion in water for 24 h}
\]
\[
V_0 = \text{the initial oven-dried volume of wood sample}
\]
\[
V_1 = \text{the oven-dried wood sample volume after treatment}
\]
\[
Su = \text{the volume shrinkage of untreated wood sample}
\]
\[
St = \text{the volume shrinkage of treated wood sample}
\]

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% MEGSiO₂ impregnation treatments. The mean differences between the treatments were determined using Duncan’s multiple range test.

### Table 1. Composition of MEG and nano SiO₂ solutions used for impregnation.

<table>
<thead>
<tr>
<th>Treatment</th>
<th>MEG (mL)</th>
<th>Water (mL)</th>
<th>Nano SiO₂ (g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Untreated</td>
<td>0</td>
<td>1000</td>
<td>0</td>
</tr>
<tr>
<td>MEG</td>
<td>500</td>
<td>500</td>
<td>0</td>
</tr>
<tr>
<td>0.5% MEGSiO₂</td>
<td>500</td>
<td>500</td>
<td>5</td>
</tr>
</tbody>
</table>

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 SiO₂ 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 cm³) 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.

3. RESULTS and DISCUSSION

3.1. Dimensional stability and density of impregnated woods

The impregnation treatment with MEG and nano SiO₂ 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

<table>
<thead>
<tr>
<th>Treatment</th>
<th>WPG (%)</th>
<th>ASE (%)</th>
<th>WU (%)</th>
<th>BE (%)</th>
<th>Density (g/cm³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Untreated</td>
<td>0 a</td>
<td>0 a</td>
<td>112.2 (±11.1)c</td>
<td>2.2 (±0.5) a</td>
<td>0.31 (±0.03) a</td>
</tr>
<tr>
<td>MEG</td>
<td>46.8 (±10.1)b</td>
<td>55.8 (±9.4)b</td>
<td>64.4 (±4.0)b</td>
<td>4.2 (±0.6)b</td>
<td>0.44 (±0.04)b</td>
</tr>
<tr>
<td>0.5% MEGSiO₂</td>
<td>56.7 (±3.3)c</td>
<td>67.2 (±9.0)c</td>
<td>31.7 (±6.1)c</td>
<td>6.3 (±0.8)c</td>
<td>0.46 (±0.04)b</td>
</tr>
</tbody>
</table>

WPG, weight percent gain; ASE, antiswelling efficiency; WU, water uptake; BE, bulking effect.

Values followed by the same letters show no real difference based on the Duncan test.

Table 3. F-values of variance analysis

<table>
<thead>
<tr>
<th>Source</th>
<th>F-value</th>
</tr>
</thead>
<tbody>
<tr>
<td>WPG</td>
<td>109.79**</td>
</tr>
<tr>
<td>ASE</td>
<td>88.00**</td>
</tr>
<tr>
<td>WU</td>
<td>116.16**</td>
</tr>
<tr>
<td>BE</td>
<td>44.28**</td>
</tr>
<tr>
<td>Density</td>
<td>26.36**</td>
</tr>
</tbody>
</table>

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 density values 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% MEGSiO₂ were greater than those of samples treated with water or MEG. However, the 0.5% MEGSiO₂ 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 SiO₂ was greater than that of wood treated with FA treated alone. Similar results were 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 strengthening is 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% MEGSiO₂ treatment. The higher the ASE value, the less the wood is able to adsorb water vapor from the surrounding environment. Further, the addition of SiO₂ 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% MEGSiO₂ 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 SiO₂. The decrease of water absorption indicates the reduction of wood hygroscopicity (Priadi et al., 2019). Hill’s (2006) suggestion that 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 MEGSiO₂ filling the cell wall, minimizing the availability of space for water molecules.

The increase in the BE values suggests that adding MEGSiO₂ via the impregnation process can improve wood dimension stability. The samples treated with 0.5% MEGSiO₂ had the greatest BE values (6.3%). This finding indicated that the addition of MEG and Nano SiO₂ 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 agent 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 thorough 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 SiO₂. These addition prevented moisture from penetrating into wood (Jiang et al., 2013).

3.2. Jabon wood morphology (SEM analysis)

SEM analysis revealed the morphology of untreated jabon wood, using magnification 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 underwent morphological 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 SiO₂ 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 SiO₂ can increase the MEG distribution, allowing the MEG to more readily enter, scatter, and even cover pits and vessel walls. Overall, the 0.5% MEGSiO₂ treatment was the most optimal treatment of the three polymer treatments investigated.

3.3. Composition element (Analysis using SEM–EDX)

The chemical composition of wood samples treated with water, MEG, and MEGSiO₂ was determined by SEM–EDX, and the results are shown in Table 4. The untreated treatment and MEG resulted in no SiO₂ content. In contrast, in the wood samples impregnated with 0.5% MEGSiO₂ the silicon content of the wood samples reaching 3.03%. Thus, it can be concluded that there is silicon in the wood treated with MEGSiO₂.

3.4. XRD analysis

Fig. 2 shows the XRD spectra of the untreated jabon wood samples and the treated samples (MEG and 0.5% MEGSiO₂). The XRD wave peaks associated with the untreated treatment, MEG, and 0.5% MEGSiO₂ occurred respectively at an angle for 2θ of 22.14°, 21.76°, and 22.08°. Furthermore, cellulose peaks were also detected at an angle for 2θ = 15.78°, 15.84°, and 15.30° corresponding to the peak of cellulose in the JCPDS data base I₀₀₂ 2θ = 22.6° and I₁₀₁ 2θ = 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
with the addition of MEG and 0.5% MEGSiO₂. It is suspected that MEG and 0.5% MEGSiO₂ have an influence on the structure of the jabon wood. The addition of nano SiO₂ 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 MEGSiO₂ treatment can increased crystallinity of
**Table 4.** Chemical composition of wood sample of jabon on various treatment of MEG and nano SiO$_2$ impregnation

<table>
<thead>
<tr>
<th>Treatments</th>
<th>Carbon (wt. %)</th>
<th>Oxygen (wt. %)</th>
<th>Silicon (wt. %)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Untreated</td>
<td>61.99</td>
<td>28.25</td>
<td>0</td>
</tr>
<tr>
<td>MEG</td>
<td>60.02</td>
<td>28.01</td>
<td>0</td>
</tr>
<tr>
<td>0.5% MEGSiO$_2$</td>
<td>55.29</td>
<td>31.77</td>
<td>3.03</td>
</tr>
</tbody>
</table>

**Fig. 2.** XRD spectra jabon impregnated wood untreated, MEG, and 0.5% MEGSiO$_2$.

The results of the XRD analysis show the effects of nano SiO$_2$ addition on WPG, 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 untreated 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 the degree 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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