Density and Dimensional Stability of a Wood-polymer Nano-Composite from Fast-growing Wood

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The characteristics of ganitri wood can be improved *via* wood impregnation. The objective of this research was to analyze the density and dimensional stability of a wood polymer nano-composite, *i.e.*, impregnated ganitri wood with a mixture of melamine formaldehyde furfuryl alcohol and nano-SiO_2. The results showed that the impregnation process improved the physical properties of the impregnated ganitri wood. Impregnation with 0.5% melamine-formaldehyde furfuryl alcohol (MFFA)-nano SiO_2 had a significant effect on the density, weight percent gain, anti-swelling efficiency, bulking effect (BE), and water uptake (WU). Increased percentage of density and BE values after being treated by MFFA with 0.5% nano-SiO_2 were 51.2% and 311.7%, respectively. The percentage decrease in WU was 47.5% (MFFA with 0.5% nano-SiO_2). X-ray diffraction analysis verified a decrease in the crystallinity of the wood cellulose. The melamine formaldehyde furfuryl alcohol and nano-SiO_2 polymers were found to cover the wood cell walls and lumens (based on scanning electron microscopy images). The formaldehyde emissions of the wood polymer nano-composite decreased. Therefore, it is possible to produce more environmentally friendly materials through wood polymer nano-composites.

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INTRODUCTION

Community forests are widely planted with fast-growing trees. The advantages of fast-growing trees include having a short cutting cycle and large-diameter timber. *Elaeocarpus sphaericus* Schum. (ganitri/rudraksha wood) is classified as a fast-growing tree species with a large diameter (Rahman 2012). The ganitri tree can be used as a seed/fruit producer (jewelry product), carpentry wood, or as a road protection tree (urban forest). Ganitri trees grow in Manila, Philippines, through Myanmar, and throughout northeastern India, Bangladesh, Nepal, and Bhutan. The ganitri tree is rather light and soft (Siarudin and Widiyanto 2013). This wood can be used for building materials and is classified as durability class IV (Heyne 1987). For example, ganitri wood aged 6 years from Sukabumi, Indonesia has an air-dry specific gravity of 0.35, and it is included in the durability class IV (Prihatini *et al.* 2020). However, according to Laksono (2019), ganitri wood aged 6 years has 100% juvenile wood. Due to the large portion of juvenile wood, wood modification is an effort to improve its quality.

The most commonly used modification technique for wood is impregnation with furfuryl alcohol (FA). Furfurylation has been successful in increasing the dimensional stability of wood (Lande *et al.* 2004; Esteves *et al.* 2011). Nanotechnology-based treatments in wood modification have been used to improve the quality of fast-growing woods. The results of a study by Hazarika and Maji (2014), who added nano-SiO_2 to melamine formaldehyde-furfuryl alcohol (MFFA), showed that nano-SiO_2 can also reduce water absorption and improve mechanical properties, fire resistance, and thermal stability of *Ficus hispida* wood. Nano-SiO_2 was treated with FA, which resulted in the increased hardness, water uptake, and anti-swelling efficiency of poplar wood (Dong *et al.* 2014). Based on the research of Rahayu *et al.* (2020), nano silica can effectively improve the dimensional stability and density of sengon wood. Melamine formaldehyde modified with furfuryl alcohol was also able to improve the dimensional stabilization and mechanical properties of Chinese spruce wood. Melamine formaldehyde (MF) resin has been widely used in wood modification because it can react easily in acidic, alkaline, or neutral media (Yao *et al.* 2017). The formaldehyde content was also a consideration in this study. Formaldehyde emissions from wood products are very important because they have a negative impact on human health. According to Candan and Akbulut (2013), the addition of nano silica materials drastically affects the emission properties of formaldehyde from particleboard and plywood panels.

Therefore, the objective of this research was to analyze the density and dimensional stability of wood polymer nanocomposites originated from impregnated ganitri wood with a mixture of melamine formaldehyde furfuryl alcohol and nano-SiO_2.

EXPERIMENTAL

Materials and Tools

The ganitri wood samples came from community forests in Sukabumi, West Java, Indonesia. The ganitri tree used was 6 years old and defect-free, with a tree height of 15 meters and a diameter of 36 cm. The chemicals used were melamine, formaldehyde, furfuryl alcohol, NaOH, nano-SiO_2 (particle diameter 15 nm ± 5 nm), maleic anhydride, universal pH paper, and demineralized water.

The tools used were an analytical balance, caliper, oven, fan, sonicator, impregnation device, moisture meter, aluminum foil, Oswald viscometer, pycnometer, thermometer, Fourier-transform infrared (FTIR) spectrometer, scanning electron microscope (SEM), X-ray diffractometer (XRD), desiccator, and spectrophotometer.

Preparation of the Test Samples

The ganitri wood was sawed without distinguishing between the heartwood and sapwood. The test sample size used was 2 cm x 2 cm x 2 cm for the density, weight percent gain (WPG), bulking effect, water uptake (WU), and anti-swelling efficiency (ASE) analyses. Each sample had 5 replications.

Preparation of the Impregnant Solution

Preparation of the melamine formaldehyde-furfuryl alcohol (MFFA) copolymers

The method utilized referred to Hazarika and Maji (2014), Yao *et al.* (2017), and Prihatini *et al.* (2020) for the procedure. Melamine, formaldehyde, and furfuryl alcohol (at a 1 to 3 to 5 mole ratio) were placed in a flask. The pre-reaction started when the pH of the medium was adjusted *via* adding a 10% NaOH solution until the pH was between 9.4 and

9.6, and the temperature was slowly raised to 98 °C. When the solution was clear, furfuryl alcohol was added. After the reaction had run for approximately 10 min, 1.5% maleic anhydrous catalyst was added, and the reaction was continued for approximately 10 min. For impregnation, this MFFA copolymer was prepared at a concentration of 50%.

Dispersion of the nano-SiO_2 in the melamine formaldehyde-furfuryl alcohol (MFFA) copolymers

This method referred to Hazarika and Maji (2014) for the procedure. Nano SiO_2 was immersed in FA-water (at a 1 to 1 mol ratio) solution for 24 h with mechanical stirring, then sonicated for 30 min. After that, the MFFA copolymer was slowly added to the nano SiO_2 dispersion while being stirred. Afterward, the MFFA copolymer was slowly added to the nano-SiO_2 dispersion with stirring. Furthermore, this mixture was sonicated for 15 min. This mixture was made with three nano-SiO_2 compositions (0.5%, 1%, and 1.5%); this was done to find the optimal composition for wood impregnation. After that, the copolymer quality was tested.

Viscosity of copolymer solutions

The evaluation was performed by Ostwald viscometer (SNI 06-4567 (1998)). Some copolymer solution was put inside the Ostwald viscometer. Then, the time duration was measured for the surface of the copolymer solution to reach from the upper to the lower limit. Viscosity measurements were performed on three duplicates and on demineralized water of 27 °C. The following equation was used for viscosity,

$$\eta_s = \frac{\eta_0 \cdot t_s \cdot \rho_s}{t_0 \cdot \rho_0} \quad (1)$$

where η is the viscosity (poise), η_0 is the water viscosity (cP), and η_s is the copolymer viscosity (cP). The parameter ρ_0 is water density (g/cm^3), t_0 is water flow time (seconds), and t_s is copolymer flow time (seconds).

Preparation of the Wood Polymer Nano-Composite (WPnC)

A furfurylation process was carried out. The furfurylation process consists of three steps, *i.e.*, impregnation, polymerization, and drying. Impregnation was carried out by adapting the method outlined in Hazarika and Maji (2014). In this study, the ganitri wood samples that were previously oven dried were measured and weighed. The wood samples were either untreated or treated. These were put into different containers, followed by the impregnant solution being poured out and put in an impregnation tube. Impregnation was performed *via* a vacuum treatment of 0.5 bar for 1 h, followed by a pressure of 2 bar for 2 h. The polymerization process was carried out by cleaning the wood samples of the impregnant residue that was still attached to the outside; the samples were then wrapped in aluminum foil and placed in an oven at 90 °C for 24 h. For the drying process, after the aluminum foil was opened, the samples were reheated at a temperature of 103 °C ± 2 °C for 24 h. The samples that were oven dried had their dimensions measured and were weighted to determine the density, WPG, BE, WU, and ASE.

Characterization of the Impregnated Ganitri Wood

Determination of the presence of a mixture of MFFA and nano-SiO_2 particles in ganitri wood was carried out using FTIR, XRD, SEM, and energy-dispersive X-ray spectroscopy (EDX).

Formaldehyde Emission Testing

For this formaldehyde emission test, a slightly modified version of the method outlined in JIS A standard 1460 (2001) was employed. The wood was cut into several pieces with a total area of 1800 cm^2, with as many as 2 sets. Before being tested, pre-conditioning was carried out at a temperature of 20 °C ± 2 °C with an air humidity of 65% ± 5% to reach a constant weight for each sample that was a minimum length of 25 mm. After that, the conditioning test was carried out at a temperature of 20 °C ± 0.5 °C for 24 h ± 5 min, where the sample was put in a desiccator with 300 mL of distilled water. The formaldehyde concentrations in the distilled water were determined using a spectrophotometer.

RESULTS AND DISCUSSION

Copolymer Manufacturing

The quality of the copolymer test results can be seen in Table 1, *i.e.*, the density, viscosity, solids content, and curing time. The results of the research by Yao *et al.* (2017) showed that a viscosity value of 13 cP was still easily absorbed into wood. The drying time should be in the range of 4 to 8 h for easy and fast manufacturing. From the quality test results (Table 1), it was found that both the MFFA copolymers without nano-SiO_2 and with nano-SiO_2 had the same curing time (6 h), except for the MFFA copolymers with a 1.5% nano-SiO_2, which took 7 h. The copolymer density increased with the addition of nano-SiO_2 and increased as the nano-SiO_2 concentration increased. The viscosity also increased as the nano-SiO_2 concentration increased. The viscosity of the liquid experienced a significant increase in the MFFA copolymer with 1.5% nano-SiO_2 added. From the visual appearance of the MFFA copolymer with 1.5% nano-SiO_2, gelation began to occur after sonication.

Table 1. Copolymer Quality Test Results

Impregnant Solutions	Density (g/cm^3)	Viscosity (cP)	Solid Content (%)	Curing Time (h)
MFFA	1.15±0.02	3.70±0.15	19.79±0.13	6
MFFA with 0.5% nano-SiO_2	1.16±0.02	7.31±0.03	21.68±0.04	6
MFFA with 1% nano-SiO_2	1.17±0.03	8.34±0.02	22.70±0.84	6
MFFA with 1.5% nano-SiO_2	1.20±0.02	63.85±3.32	39.98±1.25	7

Effect of the Melamine Formaldehyde-Furfuryl Alcohol (MFFA) Nano-SiO_2 Impregnation Treatment on Density and Dimensional Stability of Ganitri Wood

Impregnation using MFFA copolymers succeeded in increasing the parameters of its physical properties, which included its density, weight percent gain (WPG), bulking effect (BE), water uptake (WU), and anti-swelling efficiency (ASE) (Table 2). Hazarika and Maji (2014) stated that when wood is filled with MFFA, the polymer will fill the lumens and empty cell walls, which results in an increase in density, WPG, ASE, and BE, and a decrease in WU.

Density and WPG of ganitri wood

Ganitri wood density values of MFFA and MFFA nano-SiO_2 treated (Table 2) tended to increase compared to untreated ganitri wood. Density values of ganitri wood were 0.41 g/cm^3 (untreated), 0.53 g/cm^3 (MFFA), 0.62 g/cm^3 (MFFA with 0.5% nano-SiO_2), 0.61 g/cm^3 (MFFA with 1% nano-SiO_2), and 0.60 g/cm^3 (MFFA with 1.5% nano-SiO_2). This is in line with the research by Yao *et al.* (2017), who showed that impregnation with melamine formaldehyde modified with furfuryl alcohol succeeded in increasing the density and WPG. The density of the ganitri wood increased, which was in line with the WPG increasing (Table 2). WPG values of ganitri wood were 45.4% (MFFA), 65.7% (MFFA with 0.5% nano-SiO_2), 64.2% (MFFA with 1% nano-SiO_2), and 59.3% (MFFA with 1.5% nano-SiO_2). The higher the WPG value, the higher the ganitri wood density. Increased percentage of density values after being treated by MFFA dan MFFA with 0.5% nano-SiO_2 were 29.3% and 51.2%, respectively.

Dimensional stability of ganitri wood

An increase in the WPG value was also directly proportional to the BE value (Table 2). The addition of nano-SiO_2 in the MFFA was also shown to be able to increase the parameters of its physical properties (when compared to the wood without the addition of nano-SiO_2). According to Hazarika and Maji (2014), nano-SiO_2 will fill the empty space of wood, thereby reducing its water absorption capacity. In ganitri wood with the addition of 0.5% nano-SiO_2, there was an increase in its physical properties; however, an increase in the nano-SiO_2 concentration was not directly proportional to an increase in the physical properties.

The increase in the WPG, BE, density, and ASE values were inversely proportional to the WU. This is presumably because the impregnant layer will reduce the ability of the wood to absorb water. The MFFA copolymer and the addition of nano-SiO_2 successfully coated the surface of the cell so that the ability of the wood to absorb water was considerably reduced, which could cause less expansion of the wood. The addition of nano-SiO_2 is also able to reduce the wood's ability to absorb water; however, an increased concentration of nano-SiO_2 was not directly proportional to the decrease in the ability of the wood to absorb water, although it was still lower compared to wood that was impregnated by only MFFA. Generally, increased percentage of BE values after being treated by MFFA dan MFFA with 0.5% nano-SiO_2 were 231.9% and 311.7%, respectively. Meanwhile the decreased percentages for WU were 21.1 % (MFFA) and 47.5% (MFFA with 0.5% nano-SiO_2).

MFFA copolymer concentration

In the current study, the physical values increased with nano-SiO_2 at a concentration of 0.5%, but all values, including the ASE, decreased at concentrations of 1% and 1.5%. Although the physical parameters were still higher at 1.0% and 1.5% nano-SiO_2, they were not significantly different from wood impregnated with MFFA. This was different from the research results of Prihatini *et al.* (2020), who conducted research on jabon wood and found that there was an increase in the physical properties parameters as the nano-SiO_2 concentration increased from 0.5% to 1%.

This difference is probably due to the size of the ganitri cells, which are smaller than in jabon wood, making it more difficult for fluids to enter the cells. According to Martawijaya *et al.* (2005), the pore diameter of jabon wood is 130 to 220 µm, and the number of pores is 2 to 5 per mm^2, while Ganitri has a pore diameter of 84.1 to 117.9 µm with a pore number of 5 to 7 per mm^2 (Prihatini *et al.* 2020). This was in accordance with Xu (2020), who stated that wood impregnation is essentially the process of removing water or air in macropores and replacing it with resin, so the cell size will have an effect.

However, at the same 1.5% nano-SiO_2 concentration, the physical properties still decreased. The decrease in the value of the physical properties of the mixture was probably due to the change in the form of the copolymer mixture into a gel, which could not as easily penetrate the wood cells. Based on the results of the quality test, the increase in viscosity was also high. This high viscosity was thought to cause reduced liquid absorption by the wood, corresponded to the results of the study by Gavrilovic-Grmusa *et al.* (2012), who stated that the increase in the viscosity of UF resin caused a decrease in the absorption of adhesive by wood. Xu (2020) stated that large molecules have a high viscosity, which also inhibits resin penetration. According to Milea *et al.* (2011), with changes in the pH and a close distance between particles due to solvent evaporation, the surface charge will decrease, and gelation will take place.

Table 2. Ganitri Wood Dimensional Stability Testing

Treatments	WPG (%)	BE (%)	Density (g/cm^3)	WU (%)	ASE (%)
Untreated Ganitri	-	1.88 (± 1.01)	0.41 (± 0.02)	86.34 (± 5.30)	-
Ganitri MFFA	45.42[a] (± 2.78)	6.24[a] (± 0.88)	0.53[a] (± 0.01)	68.14[b] (± 5.11)	53.03[a] (± 6.34)
Ganitri MFFA with 0.5% nano-SiO_2	65.73[b] (± 4.56)	7.74[b] (± 1.28)	0.62[b] (± 0.03)	45.32[a] (± 4,88)	70,19[b] (± 5.79)
Ganitri MFFA with 1% nano-SiO_2	64.20[b] (± 5.26)	7.72[b] (± 0.31)	0.61[b] (± 0.01)	45.58[a] (± 1.49)	67.35[b] (± 6.46)
Ganitri MFFA with 1.5% nano-SiO_2	59.30[b] (± 3.84)	7.38[ab] (± 0.72)	0.60[b] (± 0.02)	48.71[a] (± 6.08)	56.58[a] (± 5.89)

Note: *The value in parentheses () indicates the standard deviation value; [a-c] Values followed by the different letters show real difference according to the Duncan test (the value is the average of 5 replications)

The results of the statistical analysis showed that ganitri wood treated with MFFA with the adddition of nano-SiO_2 was significantly different from ganitri wood without the nano-SiO_2 treatment. Ganitri wood had the highest increase in physical parameters with the addition of 0.5% nano SiO_2.

Fourier-Transform Infrared (FTIR) Analysis

The FTIR spectrum of nano-SiO_2 is shown in Fig. 1 (graph a). From Fig. 1, it can be seen that the nano-SiO_2 spectrum exhibited 3 peaks, *i.e.*, 800 cm^{-1}, which is a siloxane (Si-O-Si) functional group, 1034 cm^{-1}, which is a Si-OC group, and a silanol (Si-OH) group at the range of 3063 cm^{-1} to 3673 cm^{-1}. The presence of this silanol (Si-OH) group agreed with Launer and Arkles (2013), who stated that silanol has an absorption peak at a wave number range of 3200 to 3400 cm^{-1} and the sharpest peak at wave number 3690 cm^{-1}. The siloxane group (Si-O-Si), which has a peak at a wavelength of 810 cm^{-1}, agreed with the research by Raabe *et al.* (2018).

The FTIR test results of untreated ganitri wood (as shown in Fig 1. graph b) are characterized by the absorption bands at 3425 cm^{-1} (–OH stretching), 2916 cm^{-1} and 2843 cm^{-1} (–CH_2 asymmetric stretching), 1744 cm^{-1} (C=O stretching), 1666 cm^{-1} for (–OH bending), and 1026 cm^{-1} (C–O stretching). The presence of nano-SiO_2 (as shown in Fig. 1 graph d through f) in the composite caused a decrease in the intensity of the hydroxyl group (-OH) and the carbonyl group. The peak intensity of the -CH group of the treated wood was lower the -CH peak intensity of untreated. The intensity of the carbonyl group also decreased due to the formation of hydrogen bonds with the hydroxyl groups on the silica surface (Motaung and Luyt 2010). For the Si-O-Si group, peaks are formed at different wave numbers. In wood, the Si-O-Si group has a peak at the wave number of 1018 cm^{-1} (asymmetric stretching) (Coates 2006). The shift of the peak to a higher wave number indicates an interaction between the nano-SiO_2 and the wood. According to Li *et al.* (2015), a shift towards larger numbers occurs when the particle size increases. Hydrogen bonding with the siloxane (Si-O-Si) group causes a reduction in the amount of hydroxyl group. The shift in the wave number to a larger direction indicates that hydroxyl groups are being replaced by Si-O-Si groups. This causes a reduction in the ability of wood to absorb water. This corresponds with the findings of Hazarika and Maji (2014), who reported a decrease of hydroxyl group intensity with the increase of nano-SiO_2 amount in the composite. The inclusion of the Si-O-Si groups also increased the weight of the wood, so its WPG and density will also increase. From the solid content test results, it was also seen that the addition of nano-SiO_2 increased the solids content.

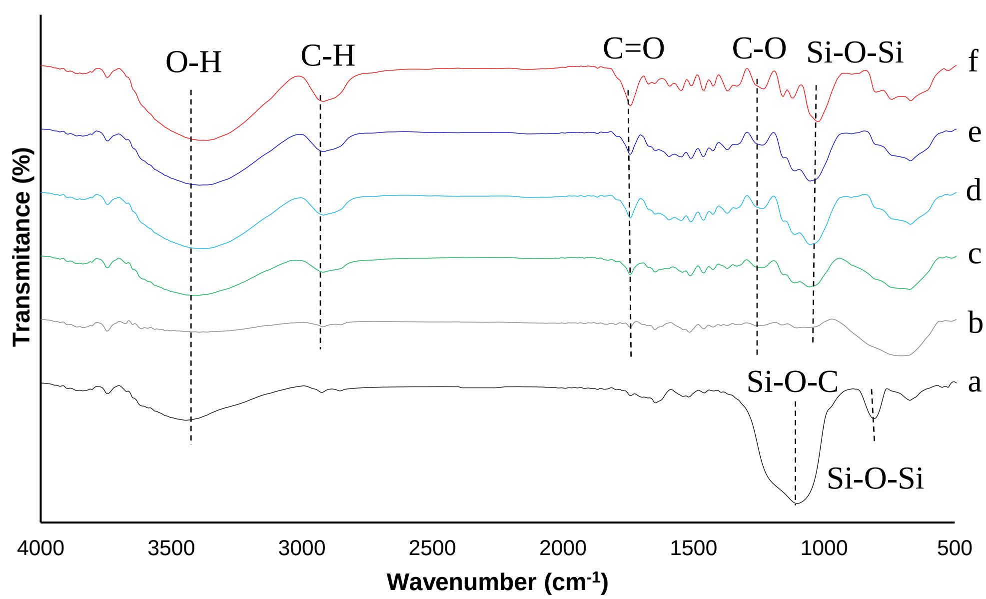

Fig. 1. The ganitri wood FTIR spectrum: (a) nano SiO_2; (b) untreated; (c) MFFA; (d) MFFA nano-SiO_2 0.5%; (e) MFFA nano-SiO_2 1%; and (f) MFFA nano-SiO_2 1.5%

X-ray Diffraction (XRD) Analysis

From the XRD curve shown in Fig. 2, the untreated wood had sharper peaks, so its crystallite value was high (as shown in Fig 2. graph a). The untreated wood showed broad diffraction peaks at 2θ 22.84° with 012 cellulose crystalline fields. Two additional small peaks appeared at 2θ 16° and 44.54°, with crystal fields of 020 and 050, respectively. After MFFA and MFFA nano-SiO_2 treatment occurred, the peak intensity decreased (Fig 2.

graph b through e). For ganitri wood with a 0.5% MFFA-nano-SiO_2 treatment, at an angle of 2θ by 16°, the graph tends to be flat. This indicates a decrease in the crystallinity of wood cellulose. Figure 2 shows that the nano-SiO_2 and MFFA had wide diffraction peaks, which shows that they were both amorphous (Fig. 2 graph f and g).

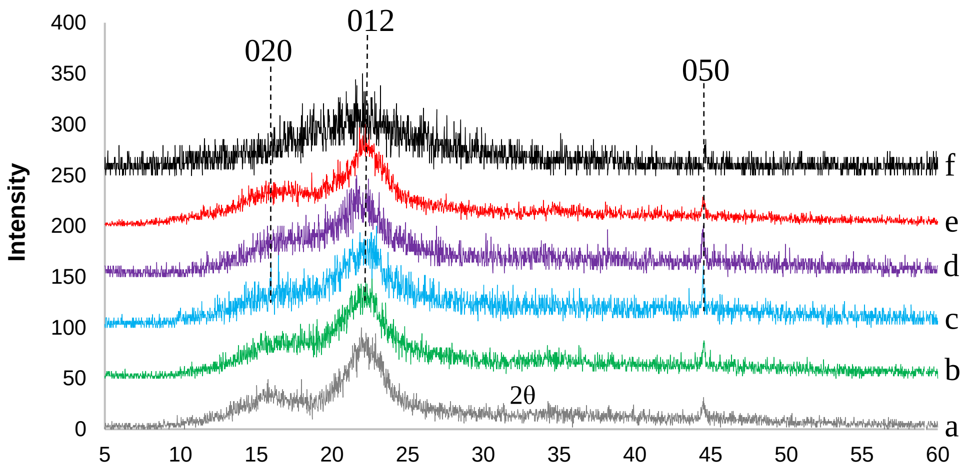

Fig. 2. The ganitri wood XRD results: (a) untreated; (b) MFFA; (c) MFFA nano-SiO_2 (0.5%); (d) MFFA nano-SiO_2 (1%); (e) MFFA nano-SiO_2 (1.5%); and (f) nano-SiO_2

Results of Testing the Degree of Crystallinity

The degrees of crystallinity associated with the treatments are shown in Table 3. The untreated ganitri wood cellulose showed the highest degree of crystallinity. The lowest degree of crystallinity for both of the two wood samples was the same treatment, *i.e.*, the wood treated with 0.5% MFFA nano-SiO_2. This was in agreement with the results by Hazarika and Maji (2014), who stated that nano silica impregnation with MFFA copolymers can reduce the degree of crystallinity of cellulose. The results of research by Raabe *et al.* (2018) also stated that coating the fiber surface with nanosilica reduced the degree of crystallinity by 15%. The results of research by Tang *et al.* (2017) showed that the addition of SiO_2 increased the formation of interfacial hydrogen bonds with fibers. The increase in the formation of hydrogen bonds with the Si-O-Si group caused the number of hydroxyl groups to decrease. Shiraishi *et al.* (1979) reported that chemical grafting reactions occurred in the amorphous regions of wood cellulose. The polymer reacts on the surface of the crystallite to open some of the hydrogen-bound cellulose chains, so that more amorphous cellulose is produced. A decrease in the degree of crystallinity indicates the formation of hydrogen bonds between silica and wood, so that the number of hydroxyl groups will decrease. This was in line with the FTIR results. Decreasing the crystallinity index will increase the physical properties of ganitri wood. In addition, ganitri wood treated with MFFA nano-SiO_2 1.5% showed a greater degree of crystallinity than ganitri wood treated with MFFA and MFFA nano-SiO_2 (at 0.5% and 1% concentrations). This was probably due to the gelation process, which makes the copolymer more difficult to penetrate the wood. It could lead to less cellulose polymer being broken down in order to bond with MFFA copolymer. However, the degree of crystallinity was still smaller when compared to the untreated wood. This corresponded to Hazarika and Maji (2014), who found the same trend in *Ficus hispida*.

Table 3. Results of the Wood Crystallinity Index Test

Treatment	Degree of Crystanility (%)
	Ganitri wood
Untreated Ganitri	22.57
Ganitri MFFA	16.83
Ganitri MFFA with 0.5% nano-SiO_2	13.91
Ganitri MFFA with 1% nano-SiO_2	15.51
Ganitri MFFA with 1.5% nano-SiO_2	20.17
Nano silica	21.17

Scanning Electron Microscopy (SEM) Test Results

Figure 3 shows the electron micrograph of the untreated and treated ganitri wood samples. It can be seen that wood that was not treated had empty pits and looked hollow, which means that it had empty space and empty cell walls (Fig. 3a and 3b). The impregnation process caused the entry of impregnant material into the wood cells. The presence of polymer deposits on the cell walls and pits can be seen in the wood samples that underwent the MFFA treatment (Fig. 3b) and in the wood treated with MFFA nano-SiO_2 (Figs. 3c through 3e). After receiving the MFFA treatment, the pits that previously had empty space became evenly closed. Indications of the presence of nano-SiO_2 can be detected by several white deposits located on the cell walls and pits between the ganitri wood cells after being impregnated with the MFFA nano-SiO_2 (Figs. 3d through 3e). Nano-SiO_2 was able to get into the pits, stick to, and even cover almost all the pore walls in the wood. In addition, Fig. 3 shows the amount of silica adhering to the surface of the wood cell, but it did not cover all the surface of the pits. Figure 3e shows the pits of the ganitri wood cells were still visible.

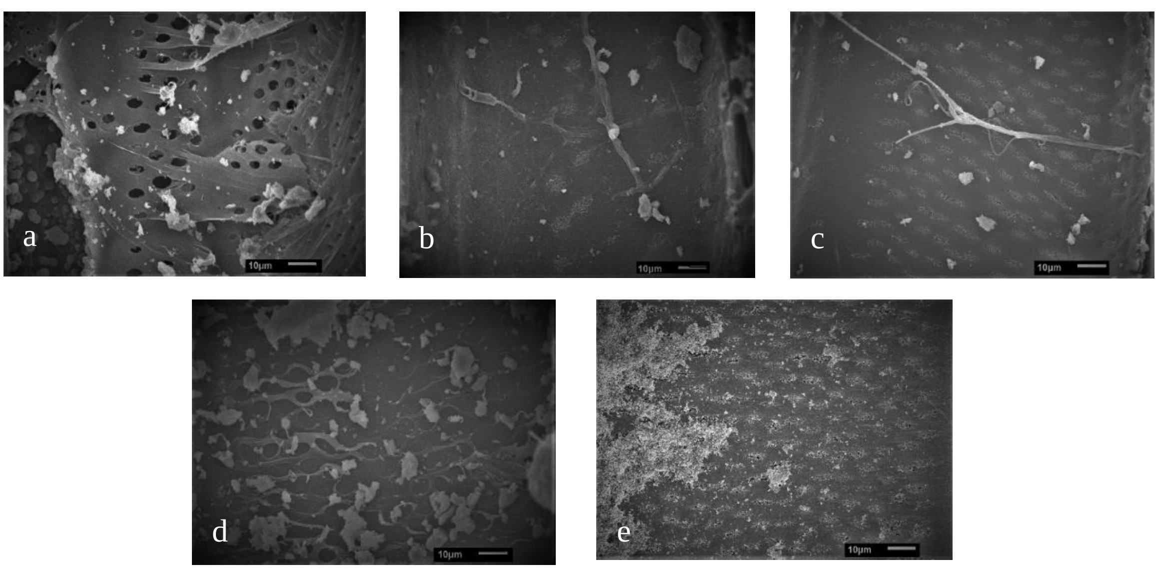

Fig. 3. SEM results of the tangential cross section of ganitri wood (1000x): (a) untreated; (b) MFFA; (c) MFFA nano-SiO_2 0.5%; (d) MFFA nano-SiO_2 1%; and (e) MFFA nano- SiO_2 1.5%

The EDX analysis results of the MFFA impregnated ganitri wood with nano SiO_2 added showed the presence of silica content. The silica content in the wood impregnated with MFFA nano-SiO_2 increased as the percentage of nano-SiO_2 increased.

Table 4. EDX Test Results

Treatment	Silica Content (% mass)
	Ganitri wood
Ganitri MFFA with 0.5% nano-SiO_2	0.23
Ganitri MFFA with 1% nano-SiO_2	0.32
Ganitri MFFA with 1.5% nano-SiO_2	0.89

Formaldehyde Emission Test

The results of the formaldehyde emission test for wood impregnated with MFFA and MFFA nanoSiO_2 are shown in Table 5. The test results show that using both MFFA and MFFA with nano silica added resulted in formaldehyde emissions that still met the JIS A standard 1460 (2001), because the formaldehyde levels were below 0.5 mg/L. This means that the wood products produced were safe for the environment.

Table 5. Formaldehyde Emission Levels

Treatments	Formaldehyde Emission Levels (mg/L)
Ganitri MFFA	0.3144
Ganitri MFFA with 0.5% nano-SiO_2	0.1962

From the table, it can be seen that the formaldehyde emission from wood with the addition of nano SiO_2 had lower emission results than the wood without nano SiO_2. This was in accordance with Lin *et al.* (2006), who stated that when the nano-SiO_2 content was below 1.5%, the amount of free formaldehyde decreased. Salari *et al.* (2013) reported that the addition of SiO_2 nanoparticles at concentrations between 1% and 3% was able to reduce formaldehyde emissions in oriented strand board (OSB) panels. This was reinforced by the results of the research by Candan and Akbulut (2013), who added nano-SiO_2 to plywood boards. This reduction in formaldehyde emissions is believed to occur because hydrogen bonding occurs between the nano-SiO_2 and formaldehyde (Lin *et al.* 2006; Roumeli *et al.* 2012).

CONCLUSIONS

Based on the present results, it is concluded that ganitri wood samples can be impregnated with melamine-formaldehyde furfuryl alcohol (MFFA) and nano-SiO_2 copolymers under vacuum and compressed conditions. The conclusions in detail were as follows:

1. Impregnation of wood with MFFA and nano-SiO_2 copolymers can increase the physical properties of ganitri wood, *i.e.*, an increased density, weight percentage gain (WPG), bulking effect (BE), anti-swelling efficiency (ASE), and a decreased water uptake (WU). Increased percentage of density and BE values after being treated by MFFA with 0.5% nano-SiO_2 were 51.2% and 311.7%, respectively, while the decreased percentage for WU was 47.5% (MFFA with 0.5% nano-SiO_2). The MFFA and nano-

SiO_2 copolymer impregnation was influenced by the wood building cells and the cell size.

2. The characterization of the results of the MFFA and nano-SiO_2 impregnation *via* FTIR showed a shift in the peak of Si-O-Si toward a higher wave number, which indicated an interaction between the nano-SiO_2 and the wood.
3. The results of the X-ray diffraction (XRD) analysis showed that there was a decrease in the crystallinity of wood cellulose after impregnating with MFFA and nano-SiO_2 copolymers.
4. The presence of MFFA and nano-SiO_2 polymers in the wood cell walls and cell lumens was confirmed *via* scanning electron microscopy (SEM) testing.
5. Based on the test results of the physical properties and the characterization *via* Fourier transform infrared (FTIR), XRD, and SEM, they showed that the addition of 0.5% nano-SiO_2 in the MFFA copolymer was the optimum concentration for the impregnation of ganitri wood.
6. The results of the formaldehyde emission testing on wood impregnated with MFFA-nano-SiO_2 at 0.5% showed that the addition of nano SiO_2 was able to reduce the formaldehyde emissions. Impregnated wood produced low formaldehyde emissions and met JIS A standard 1460 (2001), so that it can be considered environmentally friendly.

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