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IMPROVED PERFORMANCE OF WOOD POLYMER NANOCOMPOSITE IMPREGNATED WITH METAL OXIDE NANOPARTICLE-REINFORCED PHENOL FORMALDEHYDE RESIN

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Low molecular weight phenol formaldehyde (LmwPF) resin (30%) was reinforced with three types of metal oxide nanoparticles, namely, nano silicon dioxide (SiO_2), nano aluminium oxide (Al_2O_3) and nano zinc oxide (ZnO) at loading levels of 1, 3 and 5%. Following the X-ray diffraction results, nanofillers were dispersed into LmwPF resin by sonification for 60 min. Pure LmwPF resin and nanofiller-reinforced LwmPF resin were impregnated into sesenduk (*Endospermum diadenum*) wood producing impreg wood and wood polymer nanocomposite (WPNC) respectively. Physical and biological properties as well as formaldehyde emission of the samples were evaluated. The results revealed that the LmwPF resin reinforced with nanofillers had lower viscosity, higher solids content and faster gelation time than pure LmwPF. Dimensional stability was significantly improved in impreg wood. WPNC impregnated with nanofillers-reinforced LmwPF resin showed further improvement in dimensional stability. All treated samples were highly resistant against white rot fungus particular those reinforced with nano ZnO. However, higher formaldehyde emission was observed in WPNC compared with that of the impreg wood. Generally, nano Al_2O_3 gave the best results in terms of physical and biological properties of the WPNC, irrespective of loading levels.

Keywords: Sesenduk, phenolic resin, impregnation, wood polymer nanocomposite

INTRODUCTION

Sesenduk (*Endospermum diadenum*) is a tropical light hardwood and has an air-dry density between 305 and 655 kg m^{-3}. It grows in the forests of South-East Asian countries such as Malaysia, Thailand, Indonesia and Philippines. Sesenduk wood is classified as non-durable and, in tropical climate it is prone to attack by blue-stain fungi. Its high amenability to preservative treatment improved its natural durability (MTC Wood Wizard 2020). Over the last few years, due to the diminishing supply of high quality woods from natural forest in Malaysia, and due to its abundancy in the local secondary forest, sesenduk has become a popular substitute material for the overexploited tropical woods (Lee & Zaidon 2015). However, due to its non-durable nature, treatment is needed for sesenduk wood to expand its uses in the wood-based industries.

Bulking treatment with phenolic resins has shown success in improving the dimensional stability, strength properties and natural durability of low density wood (Lee & Zaidon 2015). The concentration of the phenol formaldehyde (PF) resin used should be 25–35% to attain maximum bulking effect (Nur Izreen et al. 2011, Rabi'atol Adawiah et al. 2012). If the concentration of phenolic resin as bulking agent is too high, then the emission of formaldehyde will also be undesirably high (Nur Izreen et al. 2011). Addition of nanofillers could be a solution in resolving the issue of high formaldehyde emission as it could lead to a satisfactory performance of PF resin even at lower concentration. In the recent years, there have been extensive studies on nanotechnological modification of low-quality wood. Impregnation method using nanotechnology is one of the modification

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techniques. Solid wood impregnated with nanofiller-reinforced phenolic resin and cured under heat is called wood polymer nanocomposites (WPNC). The ability of nanofillers in improving properties of wood-based materials, even at extremely low loading level, has drawn substantial interest among researchers (e.g. Cai et al. 2007, Rezaur et al. 2012, Islam et al. 2012, Nabil et al. 2015).

Various types of nanofillers have been used as reinforcing agent in enhancing the properties of PF resin. Rubberwood impregnated with low molecular weight phenol formaldehyde (LmwPF) resin reinforced with nano zinc oxide (ZnO) showed higher resistance against white rot fungi, *Pycnoporus sanguineus* (Anwar 2019b). Nanoclay has also been known as an effective reinforcing agent (Anwar et al. 2019a). Nabil et al. (2016) impregnated sesenduk wood with admixture of PF resin and nanoclay and recorded an increment of 93% in resistance against white rot fungi when 1.5% nanoclay was incorporated into 20% PF resin. In addition, significant improvement against subterranean termite was also observed in the impregnated sesenduk wood (Nabil et al. 2016). Better storage modulus was recorded in novel hybrid nanofillers composed of graphene oxide and alkali lignin reinforced PF resin (Zhang et al. 2019). Nano aluminum oxide (Al_2O_3) incorporated into urea formaldehyde (UF) resin and used as binding agent for fabrication of medium density fiberboard (MDF) produced boards with improved strength properties and decreased formaldehyde emission (Kumar et al. 2013a).

However, application of nanofillers such as nano silicon dioxide (SiO_2), nano aluminium oxide (Al_2O_3) and nano zinc oxide (ZnO) into solid wood is rather limited. Most research focused on using nanofillers for developing wood-based composite such MDF and particleboard or thermoplastic polymer. The reports on the properties of low-density wood impregnated with nanofiller-reinforced phenolic resin are relatively scarce. Therefore, the objective of this study was to study the effects of incorporating metal oxide nanofillers (nano SiO_2, nano Al_2O_3 and nano ZnO) at different loading levels on the properties of LmwPF. Physcial, mechanical and biological properties as well as formaldehyde emission of the resultant WPNC were also evaluated.

MATERIALS AND METHODS

Materials preparation

LmwPF resin with molecular weight of 600 was used in this study. The resin was obtained from Malaysian Adhesives & Chemicals Sdn Bhd located in Shah Alam. The initial concentration of the LmwPF resin was 45%. Prior to the experiment, the resin was diluted with distilled water to attain 30% concentration. Three types of single-element metal oxide nanofillers, namely, nano SiO_2, nano Al_2O_3 and nano ZnO were used to reinforce the LmwPF resin in this study.

Experimental design

The experiment was divided into two parts. The first part was the preparation and characterisation of nanofiller-reinforced LmwPF resin. The second part involved impregnation of sesenduk wood with pure LmwPF resin (denoted as impreg wood) and with nanofiller-reinforced LmwPF resin (denoted as WPNC). Physical and biological properties as well as dimensional stability of both impreg wood and WPNC were evaluated in this part.

Part 1: Preparation and characterisation of nanofiller-reinforced LmwPF resin

Dispersion of nanofiller in LmwPF matrix using sonication technique

The loading levels of nanofillers used were 0, 1, 3 and 5% based on the solids content of 30% LmwPF. The nanofillers were then incorporated into LmwPF resin and the mixtures were shaken well to ensure an even distribution of the nanofiller into the resin system. The dispersion of nanofiller-reinforced LmwPF resin was conducted according to the study by Nabil et al. (2015). A high intensity ultrasonic machine was immersed into the mixture of nanofillers and resin and the mixture were sonicated for 60 min at 50 kHz amplitude, pausing for 5 s at every 60 s to avoid heat generation.

Physical properties of nanofiller-reinforced LmwPF resins

The pH value, viscosity, solids content and gelation time of the LmwPF resin and LmwPF resin reinforced with nanofillers were evaluated. pH values of the resins were determined using digital pH meter while viscosity was evaluated using a viscometer. To determine gelation time, 6.5 g resins were poured into a test tube and immersed in a hot bath at a 100 °C. The time required for the resins to cure was recorded. To determine solids content, 1.5 g resins were poured on an aluminum foil and heated in an oven at 105 ± 2 °C for 3 hours. The samples were cooled to room temperature and reweighed and the solids content was determined.

X-ray diffraction analysis of nanofiller-reinforced LmwPF resins

The admixtures of LmwPF resin and nanofiller were cured in an oven at 103 ± 2 °C for 3 hours. The cured resins were then grounded into powder and subjected to X-ray diffraction (XRD) analysis to examine the dispersion of nanofillers in LmwPF resin. The samples were examined separately using Cu Kα radiation ($\lambda = 1.5406$A) at 40 kV and 40 mA.

Part 2: Preparation of impreg wood and WPNC

Preparation of wood samples and impregnation solutions

Air-dried sesenduk wood was obtained from the Forest Research Institute Malaysia located in Kepong, Selangor. Only defect-free samples were selected. The selected wood samples were flat sawn into strips having dimensions of 200 mm long × 50 mm wide × 5 mm thick. Prior to impregnation treatment, the strips were conditioned in a conditioning room having a temperature of 25 ± 2 °C and relative humidity of 65 ± 2% until constant weight was attained. For the preparation of impregnated wood, LmwPF resin with initial concentration of 45% was diluted to 30% using distilled water. Three types of nanofillers, namely nano SiO_2, nano Al_2O_3 and nano ZnO was incorporated into LmwPF at loading levels of 0, 1, 3 and 5%. In this study, sesenduk impregnated with pure LmwPF resin was called impreg wood while the wood impregnated with nanofiller-reinforced LmwPF resin was called WPNC.

Preparation of impreg wood and WPNC

The conditioned wood strips were impregnated using pure LmwPF resin and nanofiller-reinforced LmwPF resin, i.e. impreg wood and WPNC respectively. The wood strips were completely submerged into a vacuum-pressure cylinder filled with resins. Initial vacuum of 60 mm Hg was applied for 15 min to suck out the air from the wood strips. Next, compressed air with a pressure of 690 kPa was applied for 1 hour. When the impregnation process was finished, the pressure was released slowly and the samples were taken out. Cloth was used to wipe off excess solution on the surface of the samples before they were weighed. The treated samples were then air-dried for 24 hours. For WPNC, the treated samples were then polymerised in an oven set at 150 °C for 90–110 min. Cured samples were conditioned until constant weight was achieved and the sample properties were evaluated.

Physical properties of impreg wood and WPNC

Physical properties, namely, density, density increment, bulking coefficient, and weight percent gain were evaluated based on the procedures stipulated in BS EN 325:1993 (BS 1993) with some modifications in sample size, i.e. reduced.

Dimensional stability of impreg wood and WPNC

Dimensional stability test was performed following Ashaari et al. (1990). The tested properties including volumetric swelling coefficient, anti-swelling efficiency, water absorption and thickness swelling. For every treatment, five samples (20 mm long × 20 mm wide × 5 mm thick) were prepared. The samples were immersed in distilled water and taken out after 24 hours. The thickness, weight and volume of the samples before and after water immersion were recorded. The values of volumetric swelling coefficient (S), anti-swelling efficiency (ASE), water absorption (WA) and thickness swelling (TS) were determined using equations 1–4 (Rowell & Youngs 1981, Deka et al. 2000).

$$S\ (\%) = 100\ [(V_f - V_i)/V_i] \qquad (1)$$

where, V_f and V_i = volume of wood after and before water soaking in mm^3 respectively.

$$\text{ASE } (\%) = 100\ [(S_u - S_t)/S_t] \quad (2)$$

where, S_u = volumetric swelling coefficient of the untreated samples, S_t = volumetric swelling coefficient of treated samples.

$$\text{WA } (\%) = 100\ [(W_f - W_i)/W_i] \quad (3)$$

where, W_f = weight of samples after water immersion in g, W_i = initial weight of sample before water immersion in g.

$$\text{TS } (\%) = 100\ [(T_f - T_i)/T_i] \quad (4)$$

where, T_f = thickness of sample after water immersion in mm, T_i = initial thickness of sample before water immersion in mm

Resistant against white rot fungus of impreg wood and WPNC

The natural durability test against white rot fungus, *Pycnoporous sanguineus*, was conducted according to the procedures specified in the ASTM D 1413 (1999). For each treatment, three sample replicates having dimensions of 20 mm × 20 mm × 5 mm were used. Weight of samples before and after 16-weeks exposure to white rot fungi was recorded. Weight loss caused by the white rot fungus was calculated according to equation 5.

$$\text{Weight loss } (\%) = ((Wa - Wb)/Wa) \times 100 \quad (5)$$

where, Wa = condition weight before exposure to fungus in g, and Wb = condition weight after exposure to fungus in g.

Evaluation of formaldehyde emission on impreg wood and WPNC

Formaldehyde emission test was conducted in accordance to the procedures specified in the Malaysian Standard 1787: Part 15 (MS 2005).

Data analysis

Analysis of variance (ANOVA) was performed at 95% confident level ($p \leq 0.05$) using SPSS procedure to determine the significant levels of the studied variables. Turkey's honest significant difference tests were then used to further evaluate the effects of nanofiller types and nanofiller loading levels on the properties of the impreg wood and WPNC.

RESULTS AND DISCUSSION

Physical properties of nanofiller-reinforced LMWPF resin

The average value of pH, viscosity, solids content and gelation time of pure LmwPF and nanofiller-reinforced LmwPF are listed in Table 1. Pure LmwPF resin had a pH value of 8.5. However, after the addition of nanofillers, there were only slight changes in the pH value. Generally, the pH value of the nanofiller-reinforced LmwPF

Table 1 Physical properties of impregnation solutions

Sample	pH	Viscosity (cP)	Solids content (%)	Gelation time (min)
LmwPF	8.5	11.70	37	118
LmwPF + 1% nano SiO_2	8.5	9.00	40	113
LmwPF + 3% nano SiO_2	8.5	10.95	41	110
LmwPF + 5% nano SiO_2	8.5	11.55	40	111
LmwPF + 1% nano Al_2O_3	8.3	10.00	40	113
LmwPF + 3% nano Al_2O_3	8.3	11.55	41	113
LmwPF + 5% nano Al_2O_3	8.3	13.95	40	110
LmwPF + 1% nano ZnO	8.6	12.75	39	114
LmwPF + 3% nano ZnO	8.6	13.80	40	110
LmwPF + 5% nano ZnO	8.6	13.95	41	107

LmwPF = low molecular weight phenol formaldehyde; concentration of LmwPF was 30%

resin ranged from 8.3 to 8.6, maintaining the alkaline state of the LmwPF resin. The findings are in agreement with the studies by Hafizah et al. (2012) and Nabil et al. (2015).

Pure LmwPF had a viscosity of 11.7 cP. After the addition of 1% nano SiO_2, the viscosity of the LmwPF decreased to 9.0 cP and increased with increasing loading of nano SiO_2. At 5% nano SiO_2 addition, the viscosity of the mixture (11.55 cPa) was almost comparable with that of pure LmwPF resin. Similar trend was also observed for nano Al_2O_3. LmwPF resin admixed with nano ZnO had higher viscosity than pure LmwPF resin, even at 1% loading level. Addition of nanofillers also increased solids content of the LmwPF resin from 37% to up to 41%. Nanofillers facilitated the gelation time of LmwPF resin at 100 °C, reducing it from 118 min for pure LmwPF resin to 107–113 min for nanofiller-reinforced LmwPF resins. Higher viscosity is undesirable for nanofiller-reinforced LmwPF resins as impregnation agent. Resins with higher viscosity has poor flowability which can restrict its penetration into wood during impregnation process (Shahid et al. 2014). However, the increment in viscosity of the resins in this study was still acceptable. Meanwhile, increased solids content and shortened gelation time are favourable for nanofiller-reinforced LmwPF resins. Resins with higher solids content can ensure higher polymer retention in wood during impregnation process (Xu 2020). On the other hand, resin with shorter gelation time requires shorter curing time and saves more production cost.

Effect of sonication time on dispersion of nanofillers

The dispersion of 5% nanofiller in the LmwPF resin at sonication times of 0, 15, 30 and 60 min are shown in Figure 1. Without sonication, the peaks displayed in the figure indicated that the nanofillers were still in their agglomeration form and therefore are still not well dispersed in the LmwPF resin (Nabil et al. 2015). No significant different in intensity of the crystalline was seen after 15 min sonification. However, when the sonification times was prolonged to 30 min, the intensity of the peak increased slightly but still not fully dispersed. A flatter peak near 22° θ was observed when 60 min sonification time was applied, indicating good dispersion of nanofillers in the LmwPF resin. Nabil et al. (2015) also reported the same finding where at least 60 min sonification time was required to yield an optimum dispersion of nanoclay in LmwPF resin. Therefore, 60 min sonification time was applied in this study to disperse nanofillers in LmwPF resin and was subsequently used for wood impregnation treatment.

Evaluation of physical properties of impreg wood and WPNC

Density, bulking coefficient and weight percent gain of the untreated and treated wood are shown in Table 2. The density of untreated sesenduk wood was 437 kg m^{-3}. After impregnation with pure 30% LmwPF, the density of sesenduk wood increased as much as 42% to 622 kg m^{-3}.

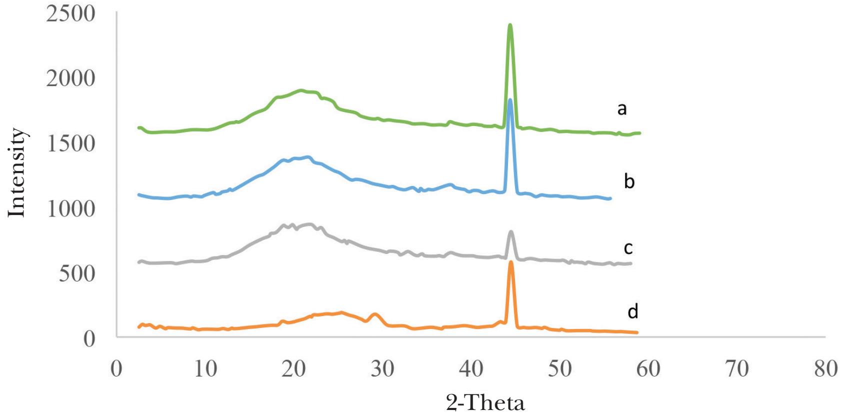

Figure 1 Effect of sonication time on the dispersion of nanofillers at different sonification times, (a) 0, (b) 15 min, (c) 30 min and (d) 60 min

Table 2 Physical properties of untreated wood, impreg wood and WPNC

Treatment combination		Density	BC	WPG
Nanofiller	Nanofiller loading (%)	(kg m^{-3})	(%)	(%)
Untreated wood	-	437^{f}	-	-
Impreg wood	-	622^{a}	8.73^{ab}	29.21^{b}
LmwPF + nano SiO_2 WPNC	1	485^{f}	9.46^{ab}	34.06^{ab}
	3	498^{f}	9.64^{ab}	33.22^{ab}
	5	506^{f}	8.75^{ab}	27.34^{b}
LmwPF + nano Al_2O_3 WPNC	1	616^{a}	11.08^{ab}	32.65^{ab}
	3	556^{bcd}	13.11^{a}	40.57^{a}
	5	547^{cde}	9.6^{ab}	40.82^{a}
LmwPF + nano ZnO WPNC	1	607^{ab}	9.92^{ab}	27.95^{b}
	3	573^{abc}	7.94^{b}	26.17^{b}
	5	617^{a}	8.65^{ab}	29.32^{b}

Mean values in the same column with the same letter are not significantly difference at $p \leq 0.05$; BC = bulking coefficient, WPG = weight percent gain; LmwPF = low molecular weight phenol formaldehyde; concentration of LmwPF was 30%

Sesenduk wood impregnated with LmwPF resin reinforced with 1, 3 and 5% nanofillers recorded density values ranging from 485 to 617 kg m^{-3}, an increment of 11 to 41% compared with that of the untreated sesenduk wood. Similar observation was also reported by Cai et al. (2007) who stated that the density of aspen wood increased after impregnated with nanoclay-reinforced melamine urea formaldehyde resin. The highest density was recorded in sesenduk impregnated with 5% nano ZnO-reinforced LmwPF resin. It is noted that sesenduk wood impregnated with pure LmwPF had higher density than sesenduk wood impregnated with nanofiller-reinforced LmwPF resin, which was in agreement with the findings reported by Cai et al. (2007).

Bulking coefficient of the of the impreg wood was 8.73%. Bulking coefficient values increased when LmwPF were reinforced with nanofillers, with exception of 3 and 5% nano ZnO. Resins with nanofillers will fill empty lumen, pits and parenchyma present in the wood resulting favourably increment in bulking coefficient (Ang et al. 2014). Weight percent gain of sesenduk wood impregnated with pure LmwPF resin and nanofiller-reinforced LmwPF resin ranged from 29.21 to 40.82%. Significant differences were found within the types of nanofillers used and weight percent gain.

Evaluation of dimensional stability of impreg wood and WPNC

The mean value of anti-swelling efficiency, water absorption and thickness swelling are shown in Table 3. The water absorption and thickness swelling values for sesenduk impregnated with pure 30% LmwPF were 27.36 and 2.09% respectively. Anti-swelling efficiency values ranged from 38.54 to 66.72%, and this indicated the effectiveness of the treatment. After being reinforced with nanofillers, water absorption and thickness swelling values reduced significantly by almost half. Significantly lower water absorption was observed in samples impregnated with 5% nano Al_2O_3, 1% nano ZnO and 5% nano ZnO compared with the rest of the nanofillers at different loading levels. Reduction in water uptake is due to the fact that nano Al_2O_3 and nano ZnO layers create a tortuous path that limit the transportation of water into the wood (Clausen et al. 2010). There was no significant difference in thickness swelling between the nanofiller types and loading levels used. However, sesenduk samples impregnated with LmwPF reinforced with 3 and 5% nano Al_2O_3 had the lowest thickness swelling values of 1.54 and 1.64% respectively. The findings are in agreement with Cai et al. (2007) who reported

Table 3 Dimensional stability of untreated wood, impreg wood and WPNC

Treatment combination		ASE	WA	TS
Nanofiller type	Nanofiller loading (%)	(%)	(%)	(%)
Untreated wood	-	-	69.42^e	4.78^a
Impreg wood	-	56.25^{ab}	27.36^b	2.09^b
LmwPF + nano SiO_2 WPNC	1	54.1^{ab}	31.45^{cd}	2.23^b
	3	49.94^{ab}	36.55^{cd}	2.29^b
	5	46.89^{ab}	44.07^d	2.41^b
LmwPF + nano Al_2O_3 WPNC	1	48.79^{ab}	30.52^{cd}	2.62^b
	3	59.71^{ab}	34.06^{cd}	1.54^b
	5	66.72^a	25.8^a	1.64^b
LmwPF + nano ZnO WPNC	1	45.46^{ab}	26.86^a	3.01^b
	3	38.54^b	29.73^c	2.6^b
	5	53.33^{ab}	26.63^a	2.42^b

Mean values in the same column with the same letter are not significantly difference at $p \leq 0.05$; LmwPF = low molecular weight phenol formaldehyde; concentration of LmwPF was 30%; WPNC = wood polymer nanocomposite, ASE = anti-swelling efficiency, WA = water absorption, TS = thickness swelling

that wood impregnated with melamine urea formaldehyde reinforced with different types of layered aluminosilicate nanofillers had lower water absorption compared with untreated wood. Accessibility of water is inhibited by strong interaction between nano silica particles, wood and polymer matrix (Deka & Maji 2013). Medium-density fibreboard made from nano Al_2O_3-reinforced resin also showed reduced swelling of thickness, which increased with increasing nano Al_2O_3 concentration (Kumar et al. 2013a, b)

Resistance against *P. sanguineus*

Table 4 summarises the weight loss of samples and increment in resistance after four weeks of exposure to *P. sanguineus.* Untreated sesenduk wood experienced the most severe attack by the fungus as indicated by the weight loss of 10.17%, and therefore was regarded as non-durable. After impregnation with LmwPF, the resistance against fungal decay increased significantly. For impreg wood, the weight loss caused by fungal decay was merely 0.22%, with an increment of 97.8% in resistance. Reinforcing LmwPF resin with metal oxide nanofillers did not give significant effect on improving fungal resistance of the WPNC. Incorporation of nano ZnO at any loading level provided total protection to WPNC against white rot fungi.

The visual observation of the test blocks after 16 weeks exposure to *P. sanguineus* are shown in Figure 2. From the figure, it can be seen that white rot fungi colonised and grew abundantly on the untreated sesenduk wood. All untreated sesenduk samples were covered by the white rot fungi. On the contrary, the surfaces of the samples impregnated with pure LmwPF resin and nanofiller-reinforced LmwPF resin were free of fungi colonisation.

The effectiveness of nano ZnO in improving the fungal resistance of treated wood could be attributed to the antifungal properties of nano ZnO itself (Clausen et al. 2010, Clausen et al. 2011, Kartal et al. 2009). Nano ZnO is also toxic towards fungi (Kartal et al. 2009, Farahani & Banikarim 2013). The physical barrier formed by LmwPF and nano ZnO protects treated wood from the digestive enzyme secreted by fungus (Zanatta et al. 2017). Apart from that, decreased hygroscopicity as indicated by lower water absorption and high anti-swelling efficiency values are also one of the main reasons for the improved fungal resistance (Nabil et al. 2015).

Evaluation of formaldehyde emission

Formaldehyde emission value from the samples impregnated with LmwPF and nanofiller-reinforced LmwPF resin are summarised in Table 5. Formaldehyde emission of the impreg

Table 4 Mean weight loss and increment in resistance of untreated wood, impreg wood and WPNC after four weeks exposure to white-rot fungus

Treatment	Nanofiller loading (%)	Weight loss (%)	Increment in resistance (%)	Durability
Untreated wood	-	10.17^a	-	Non-durable
Impreg wood	-	0.22^b	97.8	Very durable
LmwPF + nano SiO_2 WPNC	1	0.26^b	97.4	Very durable
	3	0.23^b	97.7	Very durable
	5	0.29^b	97.1	Very durable
LmwPF + nano Al_2O_3 WPNC	1	0.00^b	100	Very durable
	3	0.73^b	92.8	Very durable
	5	0.72^b	93	Very durable
LmwPF + nano ZnO WPNC	1	0.00^b	100	Very durable
	3	0.00^b	100	Very durable
	5	0.00^b	100	Very durable

Mean values in the same column with the same letter are not significantly difference at $p \leq 0.05$; LmwPF = low molecular weight phenol formaldehyde; concentration of LmwPF was 30%, WPNC = wood polymer nanocomposite

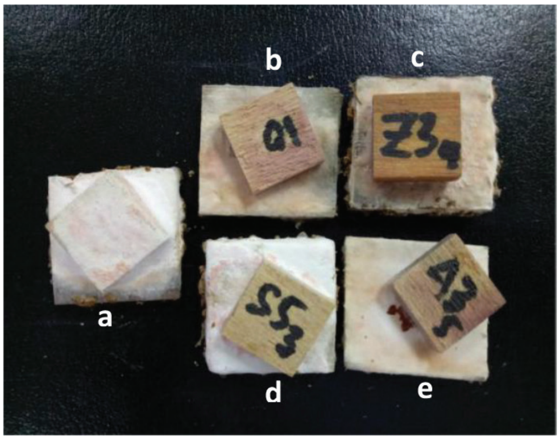

Figure 2 Test block of (a) untreated samples, (b) impreg wood, and wood polymer nanocomposite reinforced with (c) nano ZnO, (d) nano SiO_2 and (e) nano Al_2O_3 after 16 weeks exposure to *Pycnoporus sanguineus*

wood impregnated with pure LmwPF resin was 22.11 mg L^{-1}. Incorporation of nanofillers into LmwPF resin did not affect the formaldehyde emission of the WPNC significantly. Formaldehyde emission of the WPNC impregnated with 1, 3 and 5% nano SiO_2 reinforced LmwPF resin ranged from 24.69 to 26.30 mg L^{-1}. WPNC impregnated with nano Al_2O_3 and nano ZnO at the same loading level recorded formaldehyde emission values ranging from 22.54 to 26.71 mg L^{-1} and 21.65 to 25.72 mg L^{-1} respectively.

Based on the results obtained, it was observed that the addition of nanofillers did not reduce the formaldehyde emission level of sesenduk wood. In fact, formaldehyde emission level increased in some of the cases compared with impreg wood. The results are in agreement with Candan and Akbulut (2012) who reported that the formaldehyde emission level of plywood bonded with urea formaldehyde resin increased when reinforced with metal oxide nanofillers. On the contrary, melamine urea formaldehyde

Table 5 Formaldehyde emission test for impreg wood and WPNC

Treatment combination		Formaldehyde emission (mg L^{-1})
Nanofiller type	Nanofiller loading (%)	
Impreg wood	-	22.11^a
LmwPF + nano SiO_2 WPNC	1	24.79^a
	3	26.30^a
	5	24.69^a
LmwPF + nano Al_2O_3 WPNC	1	22.54^a
	3	26.71^a
	5	25.72^a
LmwPF + nano ZnO WPNC	1	21.65^a
	3	23.95^a
	5	25.72^a

Mean values in the same column with the same letter are not significantly different at $p \leq 0.05$; LmwPF = low molecular weight phenol formaldehyde; concentration of LmwPF was 30%, WPNC = wood polymer nanocomposite

showed reduced formaldehyde emission (up to 82%) level after being reinforced with metal oxide nanofillers (Candan & Akbulut 2012). The effectiveness of metal oxide nanofillers is highly dependent on the types of resin used (Candan & Akbulut 2012). Therefore, in this case, incompatibility between metal oxide nanofillers, nano SiO_2, nano Al_2O_3 and nano ZnO, and LmwPF resin might be the reason that caused the inability in reducing formaldehyde emission.

For the case of 1% nano ZnO, the formaldehyde emission of the treated sesenduk was found lower than the impreg wood. Incorporation of 1 and 3% SiO_2 reduced the formaldehyde emission form UF-bonded oriental strand board (OSB) (Salari et al. 2013). However, when the loading of SiO_2 increased to 5%, the formaldehyde emission of OSB increased. This might be caused by resin aggregation resulting from the higher loading level of nanofiller used, which negatively affected the formaldehyde emission value (Salari et al. 2013).

CONCLUSIONS

Impreg wood and WPNC were produced in this study using sesenduk wood impregnated with 30% LmwPF resin and nanofiller-reinforced LmwPF resin respectively. Generally, in comparison with pure LmwPF resin, nanofiller-reinforced LmwPF resin had lower viscosity, higher solids content and faster gelation time. XRD analysis revealed that 60 min of sonification time was required to yield an optimum dispersion of nanoclay in the LmwPF resin. Densities of impreg wood and WPNC increased mainly due to increase in weight percent gain. Water absorption and thickness swelling values of impreg wood and WPNC reduced significantly compared with that of the untreated sesenduk wood. Significant improvement in resistance against white rot fungi was also observed. However, addition of nanofillers did not significantly affect the formaldehyde emission of WPNC. Of the nanofillers tested, nano Al_2O_3, irrespective of loading level, displayed superior physical properties and dimensional stability of WPNC compared with nano SiO_2 and nano ZnO.

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