White and Brown Rot Fungal Decay Resistance of Epoxy Composite Modified with Nanocellulose and Tetraethoxysilane

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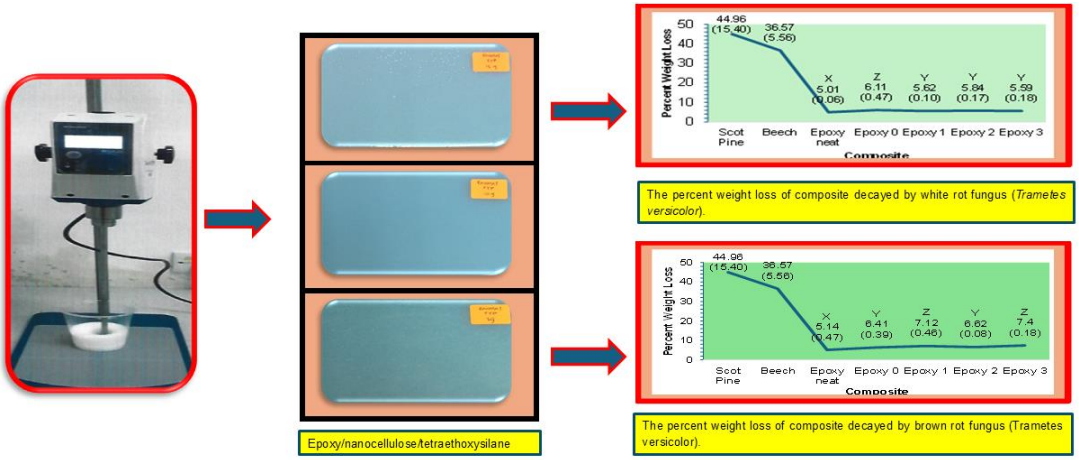
GRAPHICAL ABSTRACT



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The resistance of epoxy composite modified with nanocellulose and tetraethoxysilane (TEOS) to decay by white rot (*Trametes versicolor*) and brown rot (*Coniophora puteana*) fungi was investigated using EN 113 (1996) as the guideline. The objective of this study was to investigate the effect of TEOS as a cross-linked agent in epoxy/nanocellulose composite, and its resistance against white rot and brown rot fungi. The epoxy resin was mixed with 10 wt% nanocellulose. The other three sets were prepared the same, but with the addition of 1%, 2%, and 3% TEOS for each set. All types of epoxy composites were air dried in a mold at ambient temperature for seven days followed by oven drying at 80 °C for 30 min. The composites were oven dried at 103 °C, sterilized, and exposed to the fungi at 22 °C for 16 weeks. It was found that the use of 1% to 3% TEOS in the composite reduced the percent weight loss following decay by *T. versicolor*, but not in the case of *C. puteana*. Overall, all types of the composite in this study were classified as highly durable and durable against the *T. versicolor* and *C. puteana* respectively. The surface and structure of all types of composites were still intact after 16 weeks of exposure period.

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Keywords: Composite; Nanocellulose; TEOS; Decay; Resistance; Class

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INTRODUCTION

The fourth generation of composites, known as the "natural fiber-reinforced composite", was invented to address the dimensional stability flaws in the preceding generation. The principal requirements utilize a blend of rubber, elastomer, resin, and synthetic polymer as the matrix to enhance the mechanical capabilities and dimensional stability (Salit *et al.* 2015). Meanwhile, the fifth generation, called "functional composite", was invented to meet a specific purpose in mind for biomaterial and biomedical applications. This new generation of composite combines with a specific and selective additive material such as hydroxyapatite, titanium oxide, calcium orthophosphate,

halloysite nanotube, and others. The purpose of the functional composite is to serve as a vehicle to support the release of drugs and proteins, and to be used in tissue implants, enamel, and artificial bone (Dorozhkin 2009; Lvov *et al.* 2014; Pan *et al.* 2016). Today, the application of functional composites is diverse. They are used as electromagnetic shielding materials for aerospace, electronics, and wearable fields, in which the selected substrates are polymers, such as polyurethane (Jiang *et al.* 2019; Lu *et al.* 2021; Sang *et al.* 2021; Wang *et al.* 2022), nanofibers (Ji *et al.* 2018; Zhu *et al.* 2022) and aerogels (Ling *et al.* 2013; Zeng *et al.* 2020), nanofillers, including metal-based materials (Zhang *et al.* 2015; Gao *et al.* 2021; Liao *et al.* 2021; Liu *et al.* 2021b), MXene materials (He *et al.* 2020; Song *et al.* 2020, 2021; Zhu *et al.* 2021), and carbon-based materials (Gao *et al.* 2020; Xiang *et al.* 2020; Han *et al.* 2021; Liu *et al.* 2021a; Song *et al.* 2021).

Structural power composite, also known as structural battery composite, is another type of functional composite design to offer mass-less energy storage for structural systems that are powered by electricity. Carbon fibers are used in a structural electrolyte matrix material to create structural battery composites. Utilizing their superior mechanical qualities, high electrical conductivity, and great lithium insertion capacity, neat carbon fibers are employed as a structural negative electrode. The structural positive electrode is made of carbon fibers covered with lithium iron phosphate. The electrochemically active material in this case is lithium iron phosphate, and the fibers both conduct electrons and support mechanical stresses. Mechanical loads are transferred between the fibers by the lithium-ion conductive structural electrolyte surrounding them (Asp *et al.* 2019).

The agriculture-based biocomposite is receiving increased attention in developing the fourth and fifth generation of composites because it is inexpensive, widely available, and meets some of the sustainable development goals, such as SDG 9 (industry, innovation, and infrastructure) and SDG 12 (responsible consumption and production). This included the epoxy resin reinforced with amorphous and crystalline silica from rice husk (Hamid *et al.* 2019), ramin/flax hybrid natural fibre composite (Sumesh *et al.* 2023), *Aloe vera*/ramie fibre-reinforced epoxy hybrid composite (Arivendan *et al.* 2024), as well as pineapple fiber and potato waste polyethylene composites (Sumesh *et al.* 2024). The use of silica aimed to completely remove the hydrophobic nature of the lignocellulosic material in biomass material. The epoxy nanocrystalline silica composite shows a lower thickness swelling as compared to the epoxy nano-amorphous silica composite. However, the epoxy nanocrystalline composite has a higher mechanical property. Overall nano-amorphous silica and nanocrystalline silica at 10% dose gave optimum properties (Hamid *et al.* 2019).

The tetraethoxysilane (TEOS) is a functional cross linking silica agent that has been reported to improve the interfacial, mechanical interlocking, optical, thermal stability and conductivity of the fibre and polymer mixed composite (Wu *et al.* 2017; Abd Elnaiem *et al.* 2021). In a further study, tetraethoxysilane (TEOS) was used as a cross linker for epoxy reinforced with 10% dose of nanocellulose (Hamid *et al.* 2021). The water absorption in epoxy mixed with a 10% dose of nanocellulose was significantly higher in the 1% dose of TEOS (2.06%) than the nanocellulose composite reacted with a 2% dose (1.76%) and a 3% dose (1.46%) of TEOS. The modulus of elasticity (MOE) was significantly higher in epoxy mixed with a 10% dose of nanocellulose reacted with a 1% dose (2389.7 MPa) of TEOS, but it was not significantly different ($P < 0.05$) with a 2% dose (3356.5 MPa). Additionally, a 3% dose of TEOS significantly reduced the MOE (1378.1 MPa) compared to the composite without TEOS (2159.7 MPa). However, until recently, no study has been reported on its decay resistance to white rot and brown rot fungi.

Despite possessing excellent functional properties depending on the type of additives used, both fourth and fifth composites could be subjected to accelerated weather ageing and microorganism attack, such as fungi, especially when used in direct contact with air and soils. Therefore, the objective of this study was to investigate the resistance of epoxy, nanocellulose, and TEOS mixture composites to white rot and brown rot fungi.

This study is important to explore the uses of epoxy/nanocellulose composite in harsh environment condition as substitution to plastic, wood, wood-based composites, and natural fibre reinforce composite as building materials, vehicle and drone components.

MATERIALS AND METHODS

The commercial nanocellulose was supplied by Zaopnano Sdn. Bhd, Serdang, Malaysia. Epoxy resin (Asasin 8205A), hardener (Asahard 8205B), and tetraethoxysilane (TEOS) were supplied by Asachem (M) Sdn. Bhd and Merck (M) Sdn. Bhd, Petaling Jaya, Malaysia.

Fabrication

The fabrication of different types of composites such as the neat epoxy, epoxy mixed with 10% nanocellulose, and additional of 10 % nanocellulose/epoxy composite with 1, 2, and 3% TEOS, separately were prepared according to Hamid *et al*. (2021). The 10% nanocellulose was chosen in this study because it gave a maximum modulus of rupture (55.7 MPa) and a minimum thickness swelling (1.29%). After adding the hardener, the mixture was mechanically agitated for an additional 3 min at 300 rpm while 1%, 2%, and 3% (weight-based) dosages of tetraethoxysilane were added to the beaker. After being left in the mold for seven days at room temperature to dry, the composite was further cured in an oven for 30 min at 80 °C.

Decay Test

The wood deterioration fungi were chosen in this study, as the cellulose was one of the main structural chemical constituents being degraded by the Basidiomycetes. Same as wood, the composite used in this study also contained cellulose and its application for outdoor industrial and building materials are directly contact with water, moisture, surrounding air and weather. All types of mixture were leached in water for 14 days according to EN 84 (1997), and then dried and weighed. They were γ irradiated and exposed to white rot (*Trametes versicolor*) or brown rot (*Coniophora puteana*) over 4% Malt extract agar in vented 500-mL squat jars in accordance with the procedures in EN 113 (1996). For these purposes, 60 mL of 4% Malt agar (40 g L21 Oxoid powdered malt extract, 20 g L21 Oxoid No. 3 agar, deionized water) was dispensed into 500-mL squat jars. These were sealed with vented lids that had non-absorbent cotton wool plugs, and the jars were sterilized in an autoclave.

The *T. versicolor* and *C. puteana* were allowed to grow on the medium at 22 °C and 65% relative humidity for 2 weeks before exposure to the blocks. The neat block epoxy, block epoxy mixed with 10% nanocellulose and its mixture with 1%, 2%, and 3% TEOS composites were exposed over a sterilized polypropylene mesh in each jar. Six replicates were used for each type of composite. In addition, similarly sized Scots pine (*P. sylvestris*) sapwood and European beech (*F. sylvatica*) blocks were exposed as reference wood specimens. There were six jars with two blocks in each for the reference wood

specimens. All of the blocks were incubated for 16 weeks in the manner as stated above. At the end of the test, the excessive mycelium was removed, and the moisture content and mass loss due to decay and operational weight loss were determined.

Determination of Durability Classes

Subsequently, the durability classification was ascertained by referring to the guidelines provided in EN 350-1 (1994), wherein a ratio, represented by the symbol *x*, was ascertained by comparing the test specimens' average mass loss to that of the reference wood species, namely Scots pine or beech. The *x* values in this investigation were computed using two distinct wood reference species. The *x* values for the following classes were found to be less than or equal to 0.15 for class 1 (very durable), greater than 0.15 and less than or equal to 0.30 for class 2 (durable), greater than 0.30 and less than or equal to 0.60 for class 3 (moderately durable), greater than 0.60 and less than or equal to 0.90 for class 4 (slightly durable), and greater than 0.90 for class 5 (not durable). The durability classes were allocated in accordance with EN 350-1 (1994).

The durability classes used for wood were still relevant in this study, as nanocellulose was also prepared from any lignocellulosic materials such as wood, bamboo, oil palm trunk, banana, and others. The nanocellulose and epoxy composite could be used as a substitution material for plastic, solid wood, wood based composite, and natural fibre reinforce composite in many applications.

RESULTS AND DISCUSSION

Decay Resistance to White Rot

The percent weight loss following the decay by white and brown rot fungi was significantly different with the composite mixture (Table 1). In both fungi (Fig. 1 and Fig. 2), the neat epoxy without nanocellulose and TEOS was significantly lowest after decay by the white rot (5.01%) and brown rot (5.14%). This indicated that the addition of nanocellulose in epoxy reduced its resistance to attack by both fungi. This could be explained as the brown rot and white rot fungi possess greater enzymatic diversity supporting lignocellulose attack (Hori *et al*. 2013).

Table 1. ANOVA on Percent Weight Loss of Composite Following Decay by White and Brown Rot Fungi

Source	Fungi	DF	F	Significance
Composite Type	White rot	4	16.46	0.000
	Brown rot	4	36.81	0.000

From Fig. 1, the percent weight loss of Scots pine (45.0%) and beech (36.6%) woods were the highest compared to the composites. In the composite, the percent weight loss of epoxy reinforced with 10% nanocellulose without TEOS had a higher percent weight loss (6.1%) compared to the epoxy/nanocellulose reacted with 1% (5.6%), 2% (5.8%), and 3% (5.6%) TEOS. The percent weight loss epoxy/nanocellulose composite was not significantly different when treated with either 1%, 2% and 3% TEOS. This indicated that the use of TEOS could increase the resistance of epoxy/nanocellulose composite to white rot attacked.

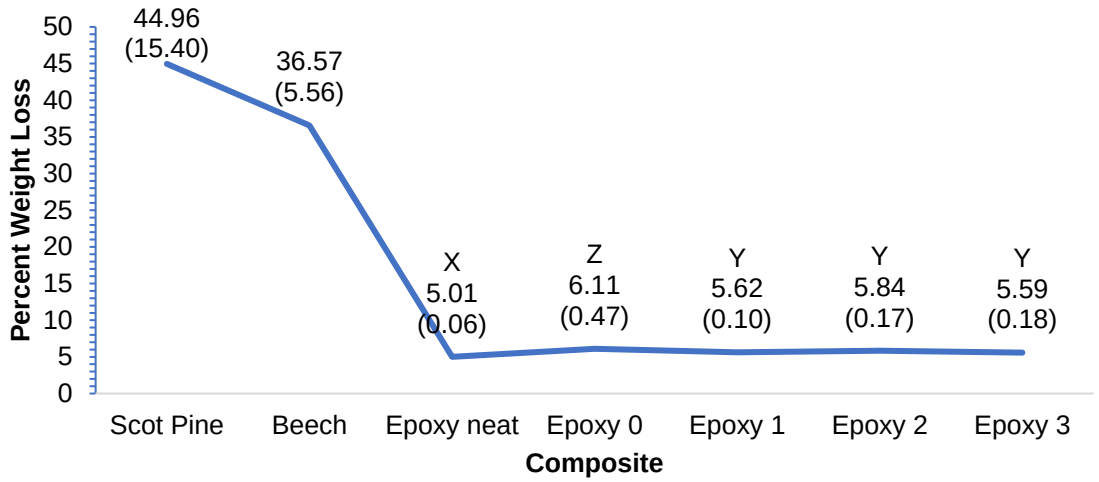

Fig. 1. The percent weight loss of composite decayed by white rot fungus (*Trametes versicolor*). Figures in the parentheses are standard deviations. Mean values followed by the same letter (s) in the same bar were not significantly different at the 0.05 probability level according to the Duncan test.

The weight loss of the composite made in this study was comparable to other types of products/processes. For comparison, the weight loss of wood-plastic composite (WPC) made from bamboo, temperate, and tropical woods ranged from 2.86% to 5.01%, except for poplar and rubber wood, which was 6.50% and 8.06%, respectively (Feng *et al.* 2021). The WPC made of rice husk mixed with 6% carbon nanotube exhibited a 6% weight loss, WPC made of hybrid poplar (Fabiyi *et al.* 2011), acetylated, propionylated, and bytyrylated rubberwood exhibited reaction at the highest weight percent gain, in which the weight losses ranged from 4.31% to 5.01% (Hamid *et al.* 2018). The weight loss of the composite in this study was lower compared to the heat-treated spruce (7.9%) and ash (6.9%) at 210 °C but tended to be higher than heat-treated Scots pine (0.4%), iroko (2.2%), and tali (0%), as reported by Sivrikaya *et al.* (2015). The weight loss of composite following decay in this study was lower than the heat-treated rose gum (23.8% to 34.3% weight loss) at temperature ranging from 140 to 220 °C, and poplar wood polymer composite impregnated with chitosan and nano silver, which ranged from 30.2 to 45.4%.

Table 2. The Weight Loss of Wood Plastic Composite, Heat and Chemical Modification of Solid Wood as Reported by Many Literatures

Type	Material	Polymer	Fungi	Additive	WL (%)	Duration (Weeks)	Reference
WPoC	Poplar	Chitosan	TV	Nanosilver (0.25%)	45.44	8	Spavento *et al.* (2023)
				Nanosilver (0.5%)	40.69		
				Nanosilver (1%)	41.49		
				Nanosilver (2%)	36.76		
				Nanosilver (4%)	30.18		

WPC	Rice husk	PP	CV	Ca. nanotube (0%)	14.0	16	Kord *et al.* (2021)
				Ca. nanotube (2%)	11.0		
				Ca. nanotube (4%)	8.0		
				Ca. nanotube (6%)	6.0		
WPC	Poplar	PP	CV	Maleic anhydride Stearic acid	6.50	12	Feng *et al.* (2021)
	Bamboo				4.90		
	Fir				2.86		
	Rubber				8.06		
	Ramin				3.94		
	Pine				5.00		
HT	Scots pine	210 °C	TV		0.4	8	Sivrikaya *et al.* (2015)
	Spruce	210 °C			7.9		
	Ash	210 °C			6.9		
	Iroko	210 °C			2.1		
	Tali	210 °C			0		
WPC	Black locust	HDPE	TV	Maleic anhydride Stearic acid	2.04	12-14	Fabiyi *et al.* (2011)
	Hybrid poplar				6.70		
	White oak				2.20		
	Douglas fir				1.07		
	Ponderosa pine				3.36		
HT	Rose gum	140 °C	PS		34.32	12	Calonego *et al.* (2010)
		160 °C			33.19		
		180 °C			32.57		
		200 °C			28.95		
		220 °C			23.81		
AC	Rubber wood	6.8 WG	TV		16.13	16	Hamid *et al.* (2018)
		11.4 WG			9.68		
		13.1 WG			6.44		
		14.6 WG			4.31		
PC	Rubber wood	8.3 WG	TV		18.88	16	Hamid *et al.* (2018)
		12.4 WG			6.53		
		14.0 WG			5.93		
		15.1 WG			4.41		
BC	Rubber wood	4.7 WG	TV		25.41	16	Hamid *et al.* (2018)
		7.2 WG			15.68		
		10.2 WG			7.48		
		13.3 WG			5.01		

WPoC- Wood polymer composite, WPC- Wood-plastic composite, HT- heat treatment, AC- acetylation, PC- propionylation, BC- butyrylation, CV- *Coriolus versicolor*, TV- *Trametes versicolor*, PS- *Pycnoporus sanguineus* , Ca- carbon

Decay Resistance to Brown Rot

From Fig. 2, the epoxy mixed with nanocellulose without TEOS gave a lower percent weight loss (6.4%) following decay by brown rot, but it was not significantly different with epoxy/nanocellulose composite reacted with 2% TEOS (6.6%). The epoxy reinforced with nanocellulose and reacted with 3% TEOS gave the highest percent weight

loss (7.4%) following decay, but it was not significantly different with epoxy/nanocellulose reacted with 1% TEOS (7.12%).

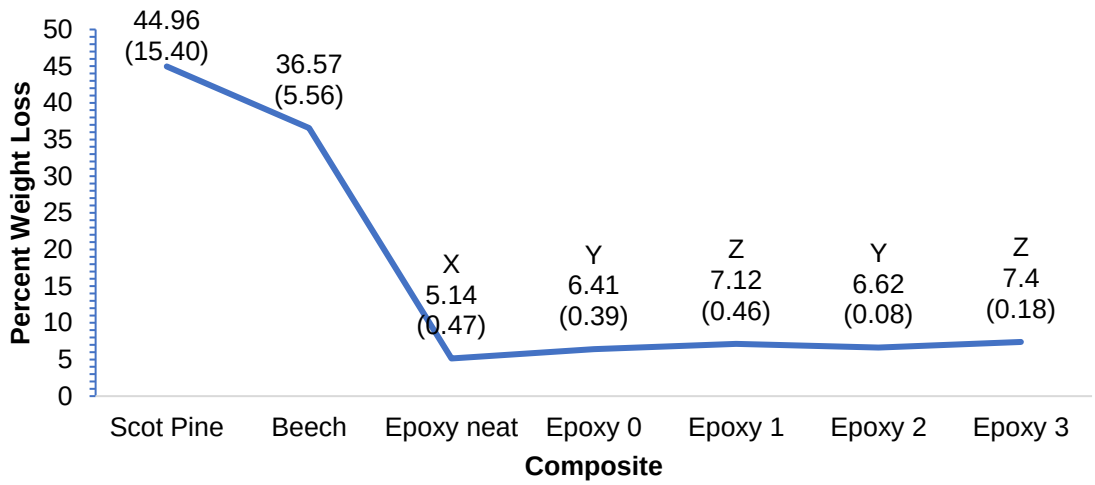

Fig. 2. The percent weight loss of composite decayed by brown rot fungus (*Coniophora puteana*). Figures in the parentheses are standard deviations. Mean values followed by the same letter (s) in the same bar were not significantly different at the 0.05 probability level according to the Duncan test.

The weight loss of composite decayed by brown rot in this study, which ranged from 6.4% to 7.4%, was comparable with WPC made of poplar (7.6%) and rubber wood (8.7%), but slightly higher than WPC made of bamboo (5.1%), fir (4.0%), ramin (4.7%), and pine (5.5%) woods. The weight loss of composite made in this study was also comparable with WPC made of ponderosa pine (7.4), but higher than WPC made of black locust (3.1%) and white oak (3.2%). The weight loss of WPC made of Douglas fir (8.1%) and hybrid poplar (9.5%) were higher than the composite made in this study.

In comparison to the wood heat treatment, the weight loss of composite made in this study was higher than the heat-treated Scot pine, spruce, ash, iroko, and tali against the *Coniophora puteana* and *Poria placenta*. The average weight loss of composite made in this study was also higher than acetylated (2.4%), propionylated (6.1%), and butyrylated (4.5%) rubberwood reacted at the highest weight percent gain.

In the perspective of decay, this indicated that a composite made of epoxy mixed with nanocellulose and TEOS could be used as alternative material to wood plastic composite, heat, and chemical modification of wood to resist white and brown rot attacks.

Decay Resistance Class

The resistance classes of the different types of composites to white and brown rot fungi are displayed in Table 4 and Table 5. For both types of fungus, the neat epoxy without any mixture was classified as very durable to white and brown rot fungi when using both Scots pine and beech woods as reference specimens. The composite mixture of epoxy/nanocellulose reacted with 1% and 3% TEOS were classified as very durable to white rot fungus, as both Scots pine and beech were used as the reference wood specimens.

Table 3. Weight Loss of WPC, Heat, and Chemical Modification of Solid Wood to Brown Rot as Reported by the Literature Review

Type	Material	Polymer	Fungi	Additive	WL (g)	Duration (Weeks)	Reference
WPoC	Poplar	Chitosan	CP	Nanosilver (0.25%)	48.99	8	Spavento *et al.* (2023)
				Nanosilver (0.5%)	50.34		
				Nanosilver (1%)	47.69		
				Nanosilver (2%)	49.90		
				Nanosilver (4%)	27.22		
WPC	Poplar	PP	P.Pla	Maleic anhydride	7.64	12	Feng *et al.* (2021)
	Bamboo			Stearic acid	5.12		
	Fir				4.02		
	Rubber				8.68		
	Ramin				4.70		
	Pine				5.54		
HT	Scots pine	210 °C	CP		0.5	8	Sivrikaya *et al.* (2015)
	Spruce	210 °C			0		
	Ash	210 °C			0.5		
	Iroko	210 °C			0.7		
	Tali	210 °C			0		
HT	Scots pine	210 °C	P.Pla		0.2	8	Sivrikaya *et al.* (2015)
	Spruce	210 °C			5.9		
	Ash	210 °C			0		
	Iroko	210 °C			0.6		
	Tali	210 °C			00		
WPC	Black locust	HDPE	CP	Maleic anhydride	3.13	12-14	Fabiyi *et al.* (2011)
	Hybrid poplar			Stearic acid	9.46		
	White oak				3.19		
	Douglas fir				8.11		
	Ponderosa pine				7.38		
AC	Rubber wood	6.8 WG	CP		19.51	16	Hisham *et al.* (2018)
		11.4 WG			9.29		
		13.1 WG			5.11		
		14.6 WG			2.38		
PC	Rubber wood	8.3 WG	CP		20.36	16	Hisham *et al.* (2018)
		12.4 WG			14.57		
		14.0 WG			6.99		
		15.1 WG			6.07		
BC	Rubberwood	4.7 WG	CP		13.06	16	Hisham *et al.* (2018)
		7.2 WG			15.71		
		10.2 WG			5.66		
		13.3 WG			4.49		

WPC- Wood plastic composite, HT-heat treatment, AC-Acetylation, PC-Propionylation, BC-Butyrylation, CP-*Coniophora puteana*, P.Pla-*Poria placenta*

In contrast, the mixture of epoxy/nanocellulose composite without TEOS and epoxy/nanocellulose reacted with 2% TEOS were classified as highly durable to brown rot fungus when using Scots pine as a reference wood specimen. However, this was not the case when beech wood was used as a reference wood specimen, for which all types of composites in this study were classified as only durable to brown rot fungus. The durability classes of the epoxy mixed with nanocellulose with and without TEOS in this study were comparable to the acetylated, propionylated, and butyrylated rubber wood at the maximum weight percent gain (Hassan *et al*. 2017).

The different percentage weight loss and resistance class obtained by both white rot and brown rot fungi could be explained by the mechanism of attacked in wood. White rot fungi simultaneously degrade all components of the plant cell wall and in some cases, more extensive degradation of lignin and hemicellulose than cellulose occurs (Kirk and Farrell 1987). In contrast to white rot fungi, brown rot fungi are able to circumvent the lignin barrier, removing the hemicellulose and cellulose with only minor modification to the lignin (Cowling 1961; Winandy and Morell 1993; Green and Highley 1997).

Table 4. X-value and Decay Resistance Classes of the Composite Decayed by White Rot (*T. versicolor*) Using Both Scots Pine and Beech Woods as Reference Specimen

Composite	X-value (S. Pine)	Class	X-value (Beech)	Class
Neat epoxy	0.11	Very durable	0.14	Very durable
Epoxy/cellulose (ES)	0.14	Very durable	0.17	Durable
ES and 1% TEOS	0.13	Very durable	0.15	Very Durable
ES and 2% TEOS	0.13	Very durable	0.16	Durable
ES and 3% TEOS	0.12	Very durable	0.15	Very Durable

Table 5. X-value and Decay Resistance Classes of the Composite Decayed by Brown Rot (*C. puteana*) Using Both Scots Pine and Beech Woods as Reference Specimen

Composite	X-value (S. Pine)	Class	X-value (Beech)	Class
Neat epoxy	0.11	Very durable	0.14	Very durable
Epoxy/cellulose (ES)	0.14	Very durable	0.18	Durable
ES and 1% TEOS	0.16	Durable	0.19	Durable
ES and 2% TEOS	0.15	Very durable	0.18	Durable
ES and 3% TEOS	0.16	Durable	0.20	Durable

Surface Appearance

As shown in Figs. 3 and 4, all types of composites in this study either neat epoxy, epoxy mixed with nanocellulose, or mixture composite modified with 1%, 2%, and 3% TEOS were still intact and showed a good structural shape after 16 weeks exposure to white and brown rot fungi. However, the most remarkable change was only the surface color that turned from white to red.

Neat epoxy			
Epoxy/nanocellulose			
Epoxy/nanocellulose/ 1% TEOS			
Epoxy/nanocellulose/2% TEOS			
Epoxy/nanocellulose/3% TEOS			

Fig. 3. Surface appearance and structure of composites after 16 weeks exposure to *T. versicolor*

Neat epoxy			
Epoxy/nanocellulose			
Epoxy/nanocellulose/ 1% TEOS			
Epoxy/nanocellulose/2% TEOS			
Epoxy/nanocellulose/3% TEOS			

Fig. 4. Surface appearance and structure of composites after 16 weeks exposure to *C. puteana*

CONCLUSIONS

1. The percent weight loss of epoxy and nanocellulose mixed with or without TEOS was lower following decay by *T. versicolor* than those of *C. puteana*. This indicates that these types of composites are more resistant to *T. versicolor* than the *C. puteana*. The use of 1 to 3% TEOS in the composite reduces the percent weight loss following decay by *T. versicolor* but not in the case of *C. puteana*.
2. Overall, all types of the composite in this study were classified as highly durable and durable against the *C. versicolor* and *C. puteana*. The surface appearance and structure of all types of composites were still intact after the exposure period.

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