

## Algal Decay Resistance of Conventional and Novel Wood-Based Composites

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Measures of the resistance to algal decay of conventional (medium density fiberboard [MDF] and plywood) and novel wood-based composites (WPC) were investigated in the same or varying wood species by using an artificial accelerated test with four mixed algal suspensions (*Chlorella vulgaris*, *Ulothrix* sp., *Scenedesmus quadricauda*, and *Oscillatoria* sp.). The morphology characterization of the surface and fracture of the specimens was analyzed using scanning electron microscopy (SEM) and a digital instrument. The pH value and the mass loss rate of the different wood species were also tested. The results showed that the algal resistance of the MDF and plywood were superior to that of the WPC of the same wood species. Furthermore, the algal resistance capacity of WPC made from various wood species were ranked as: *Liquidambar formosana* > *Cunninghamia lanceolata* and *Melaleuca leucadendra* > *Ricinus communis* > *Eucalyptus grandis* × *E. urophylla* and *Pinus massoniana*. There was a close relationship between the pH value and the algal resistance level; as the pH value increased, the alga resistance of the WPC also increased. The algal colonization only had a negative effect on the appearance of the samples.

*Keywords:* Algal resistance; Algae; Decay; MDF; Plywood; WPC

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### INTRODUCTION

Natural wood fibers originating from wood, bamboo, hemp, sisal, and straw have been combined with thermoplastic resins, such as polyethylene (PE), polypropylene (PP), and polyvinyl chloride (PVC), to make novel wood-based composites, namely wood plastic composites (WPC) (Kaewkuk *et al.* 2013; Cavdar *et al.* 2014; Xue *et al.* 2014; Malakani *et al.* 2015; Yong *et al.* 2015). Wood plastic composites are commonly substituted for traditional pressure- or dip-treated solid wood and conventional wood-based composites, such as plywood, particle board, and medium density fiberboard (MDF) for a wide array of applications (Verhey *et al.* 2001; Fabiyi *et al.* 2009; Ayrilmis 2013; Peng *et al.* 2014). Most recently, composites with high wood flour content have led to the further development of WPC, with the advantage of lower cost (Hamzeh *et al.* 2011).

Their excellent biodegradation resistance of the WPC was an advantage at one time. Scientists speculated that the encapsulation of the hydrophobic plastic matrix of wood fiber could provide sufficient resistance to biodegradation (decay from fungi, moulds, algae, termites, and marine borers), without requiring chemical preservative agents. This idea was gradually demolished by the results of laboratory testing

(Gnatowski 2009; Segerholm *et al.* 2012; Tascioglu *et al.* 2013). The current methods are presumed reasonable because of the varied number of formulations employed, *i.e.*, different types and amounts of wood fiber, resin matrix, and other additives, as well as processing technologies (Schirp *et al.* 2008; Naumann *et al.* 2012; Xu *et al.* 2013, 2015).

Among these variables, wood species has played a significant role in the WPC's biodegradation performance. Different wood species have resulted in either a passive or negative effect on the susceptibility and resistance to harmful living organisms (Vazquez *et al.* 2008; Xu *et al.* 2015). The colonization of mold on maple flour/PVC composites was reported to be more severe than on pine flour/PVC composites by Dawson-Ahdoh *et al.* (2004). Lomeli-Ramirez *et al.* (2009) concluded that the fungal resistance of PP-based WPC manufactured using either maple flour or oak flour was poor, while pine flour performed quite well. Fabiyi *et al.* (2011) demonstrated that Douglas fir wood flour was more suitable for PE-based WPC products than polar wood flour, with a minimal chance of attack by white rot. The PE-based WPC, with a high content of wood (50 to 70 wt%), experienced 18% and 7% more weight loss when exposed to a white rot for 16 weeks and a brown rot for 12 weeks, respectively (Mankowski and Morrell 2000). In addition, PVC-based WPC exhibited excellent resistance ability to harmful biological species, when compared to PE-based WPC (Xu *et al.* 2013). Up until now, most scientific investigations on WPC biodegradation were mainly focused on mould and fungal resistances; few scientific studies address the biodegradation of WPC by algal decay.

In this study, a comparison of algal resistance for conventional wood-based composites (MDF and plywood) and novel WPC using the same wood raw material species was investigated using the artificial accelerated test method. Furthermore, the algae resistance differences of the WPC specimens made from six wood species were also estimated. The pH value for the different wood species was tested to reveal the differences in various treatment groups. Lastly, scanning electron microscopy (SEM) and a digital instrument was used to observe the algal microspore growth or mycelium concentration on the WPC specimens at the surface or in the interfacial gaps of the wood and thermoplastic components.

## EXPERIMENTAL

### Raw Materials

Six wood species were used as wood flour: *Cunninghamia lanceolata*, *Pinus massoniana*, *Eucalyptus grandis* × *E. urophylla*, *Melaleuca leucadendra*, *Liquidambar formosana*, and *Ricinus communis*. The woods were obtained from Zengcheng Baigao Manufacturing Co. Ltd., China. In addition, *Eucalyptus grandis* × *E. urophylla* wood was selected to prepare the MDF and plywood. Four algal species, *Chlorella vulgaris* (ATCC 11468), *Ulothrix* sp. (ATCC 30443), *Scenedesmus quadricauda* (ATCC 11460), and *Oscillatoria* sp. (ATCC 29135), were provided by the Guangdong Institute of Microbiology, Guangzhou, China. The thermoplastic resin was PVC (DG-800), with an average degree of polymerization (800) and a density of 1.35 to 1.45 g/cm<sup>3</sup> (Tianjin Dagu Co. Ltd., China). Urea-formaldehyde (UF) resin adhesive with a solid content of 60% and a viscosity of 0.19 Pa·s for the preparation of MDF and plywood was purchased from a local chemical company. Other additives for the WPC preparation included processing aids, calcium zinc stabilizer, and paraffin wax, which were purchased from the Guangzhou Yantai Chemical Co. Ltd., China.

## Methods

### *MDF preparation*

*Eucalyptus grandis* × *E. urophylla* wood fiber was selected to prepare the MDF samples. Wood fiber was dried to a moisture content of less than or equal to 5.0 wt%, and was submerged into the UF adhesives. Resinated fiber materials with additional additives were pre-pressed and final-pressed for approximately 6 min at the temperature range of 130 to 180 °C and a pressure range of 1.5 to 4 MPa, forming the MDF boards with the dimensions of 500 mm × 500 mm × 6 mm and a mean density of 0.9 g/cm<sup>3</sup>.

### *Plywood preparation*

*Eucalyptus grandis* × *E. urophylla* veneers were used to prepare the 3-layer plywood panels with the dimensions of 320 mm × 320 mm × 2.2 mm. First, a 5.0 wt% ammonium chloride solution was added to the UF resins for mixing. Subsequently, the compound resin was applied to both sides of the veneer at a spread rate of 350 g/m<sup>2</sup>. The adhesive-coated veneers were then stacked between two uncoated veneers with the grain directions of two adjacent veneers perpendicular to one another. Finally, the assembled veneers were pre-pressed using 0.5 MPa of pressure at room temperature for 30 min, and hot-pressed at the temperature and pressure of 120 °C and 1.0 MPa, respectively. The thickness-dependent pressing time was applied for this process at 1 min/mm.

### *PVC-based WPC preparation*

Forty to sixty mesh particle size wood flour (*Cunninghamia lanceolata*, *Pinus massoniana*, *Eucalyptus grandis* × *E. urophylla*, *Melaleuca leucadendra*, *Liquidambar formosana*, and *Ricinus communis*) was oven-dried at 105 ± 2 °C to ensure the moisture content was less than 1.0%. Wood flour (100 phr) and PVC resin were pre-mixed in a high-speed mixer (SHR-10A, Zhangjiagang, China) operating at 1800 rpm at a temperature of 80 °C for 8 min. Then, 4.0 phr of calcium zinc stabilizer, 6.0 phr of compound processing aids, and 0.8 phr of paraffin wax were also added into the compound system and mixed for 10 min at 105 °C. Finally, the solution was extruded as a sheet using a conical twin-screw extruder (LSE-35, Guangzhou, China) at 130 to 185 °C from the hopper to die zones with a rotational speed of 20 to 25 rpm.

### *Artificial accelerated algal resistance test*

The artificial accelerated algal resistance tests of the specimens were conducted according to the GB/T 21353 (2008) standard. First, the liquid culture medium was prepared in a solution by adding 1.25 g/L of potassium nitrate, 1.25 g/L of potassium dihydrogen phosphate, 0.49 g/L of magnesium sulfate, 0.5 g/L of ethylenediamine-tetraacetic acid disodium salt, 0.1 g/L of boric acid, and 0.21 g/L trace elements. The solution was poured into a 250 mL Erlenmeyer flask after mixing, and then sterilized at 120 °C for 30 min. Finally, the solid medium was formed by adding a 1.5% agar gel into the prepared liquid culture medium.

Four algal species (*Chlorella vulgaris*, *Ulothrix* sp., *Scenedesmus quadricauda*, and *Oscillatoria* sp.) were selected for use in this study. Each algal sample (1.0 mL) was pumped from the four algal suspensions using a sterile pipette and pipetted into the prepared liquid culture medium. Subsequently, the liquid culture medium was maintained at a temperature of 25 ± 2 °C, 1000 to 3000 lx 14:10 h light/dark cycles, and 80% relative humidity for 2 weeks. The content of each algal sample was adjusted to 1×10<sup>6</sup> cfu/mL. Finally, the mixing suspensions with four algal species (*Chlorella vulgaris*, *Ulothrix* sp.,

*Scenedesmus quadricauda*, and *Oscillatoria* sp.) were prepared by adding the same amount of each algal sample to the suspension media.

The sterilized samples for exposure to algae with the dimensions of 30 mm (length) × 30 mm (width) were placed into petri plates. Then, the melted solid medium was added into the petri plates over 2/3 of the plate. The surface of each sample was sprayed with an equal amount of algal suspension using a spray bottle with a pressure pipe by pressing the same times. Petri plates were stored at 25 ± 2 °C for 1000 to 3000 lx 14:10 h light/dark cycles, and 80% relative humidity for 4 weeks. Resistance to the algae was estimated according to the spread of growth, as shown in Table 1. It should be noted that the surface of the medium should have exhibited algal growth by the seventh day; otherwise the test would have been considered invalid and would have been repeated.

**Table 1.** The Level Description of Algal Growth

The level description of algal growth	Level
None	0
Slight (growth area: < 10%)	1
Gentle (growth area: 10-30%)	2
Middle (growth area: 30-60%)	3
Harsh (growth area: > 60%)	4

#### *Change in mass evaluation of samples*

The evaluation of change in mass by the samples was determined by Eq. 1,

$$R = \frac{m_0 - m_1}{m_0} \times 100\% \quad (1)$$

where  $R$  is the change rate of mass (%),  $m_0$  is the mass before testing (g), and  $m_1$  is the mass after testing (g).

#### *Micro-morphology observation of samples*

The surface and fracture morphology of the specimens were observed using a SEM (Hitachi S-3000N, Philips, Japan) with an acceleration voltage of 10 kV. The specimens were dried and sputter coated with a thin layer of gold before observation.

#### *The pH value of different wood species*

Wood samples (200 g) were air-dried at room temperature and adequate ventilation. Then, the samples were ground into sawdust with the particle size of 40 to 60 mesh, oven-dried (105 °C), and sealed for use. Three grams of wood particles were immediately added into a 50 mL beaker, and 30 mL de-ionized water was subsequently added. Then, the solution was stirred for 10 min and let stand in the beaker for 20 min. Finally, the pH value of the solution was measured using an acidometer (PHD-200A, Shanghai, China). The experiment was performed in three parallel measurements.

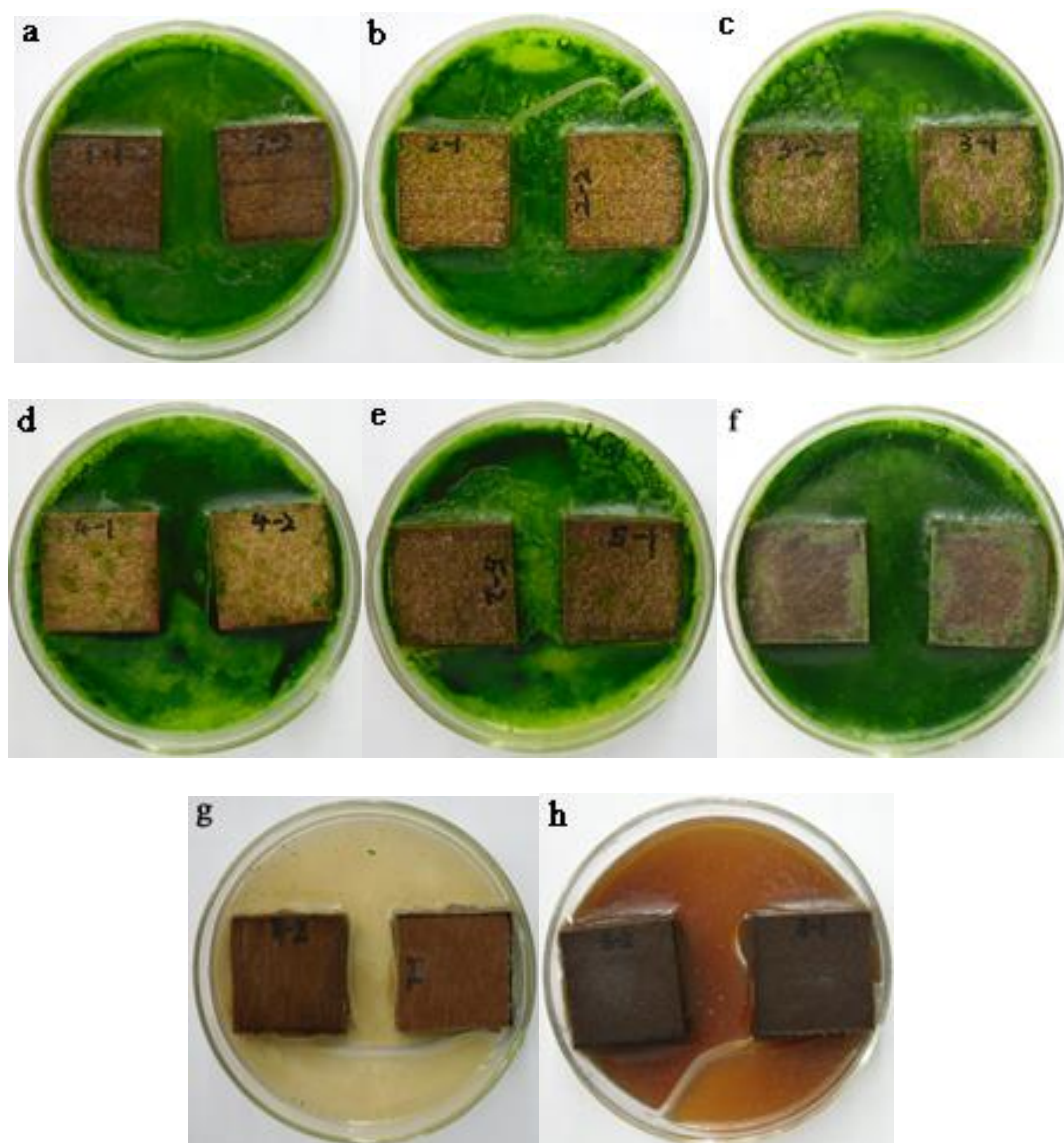
## RESULTS AND DISCUSSION

### The Visual Appearance and Mass Loss Analysis

Figure 1 illustrates the algal colonization of the different WPC specimens made from six wood species, as well as the MDF and plywood after an incubation period of 28

days. It was clearly observed that there were marked differences on the algal resistance (based on appearance) and the resistance levels for the different wood groups.

Specifically, it was found that the algal resistance ability was greater in the conventional wood-based composites (MDF and plywood) than in the novel wood-based composites (WPC), when the same wood raw material was used (Table 2). The resistance level for MDF, plywood, and WPC made with the same material (*Eucalyptus grandis* × *E. urophylla*) corresponded to level 0, 0, and 4, respectively. The conventional wood-based composites with the highest resistance ability can be explained by the existence of highly toxic formaldehyde, which effectively inhibited the natural growth of algae. Therefore, the WPC also proved to be an environmental-friendly novel material.



**Fig. 1.** The visual observations of algal resistance of different WPC-based wood groups: (a) *Liquidambar formosana*, (b) *Cunninghamia lanceolata*, (c) *Eucalyptus grandis* × *E. urophylla*, (d) *Pinus massoniana*, (e) *Melaleuca leucadendra*, (f) *Ricinus communis*, (g) *Eucalyptus grandis* × *E. urophylla*, and (h) *Eucalyptus grandis* × *E. urophylla*

As for the WPC groups, it was observed that the surfaces of *Eucalyptus grandis* × *E. urophylla*, *Pinus massoniana*, and *Ricinus communis* were harshly covered by algae (Fig. 1c, d, f). This corresponded to the resistance levels of 4, 4, and 3, respectively, which meant that they exhibited poor abilities in resisting the growth of algae. On the contrary, the samples of *Cunninghamia lanceolata*-based WPC (Fig. 1b) and *Melaleuca leucadendra*-based WPC (Fig. 1e) were found to be relatively low in algal growth, with the resistance levels of 2 and 2, respectively. The WPC, *Liquidambar formosana* (Fig. 1a) exhibited the optimum algal resistance, since this group displayed the strongest resistance ability (level 1).

The change rates of mass for the different samples are shown in Table 2. It was observed that negative values for mass resulted for all groups, which indicated that the decay from algae would not decrease the quality of the sample. However, the mass of the samples gradually increased because of the water absorption over time. Consequently, the change rates of mass with negative values could be referred to as the water absorption rates. The water absorption rates of the plywood (102.73%) and MDF (93.43%) were much greater than other WPC specimens. Nevertheless, there was no correlation between the change rate of mass and the resistance level. Overall, it can be concluded that the algal colonization only had a negative effect on appearance and aesthetic appeal of the WPC specimens.

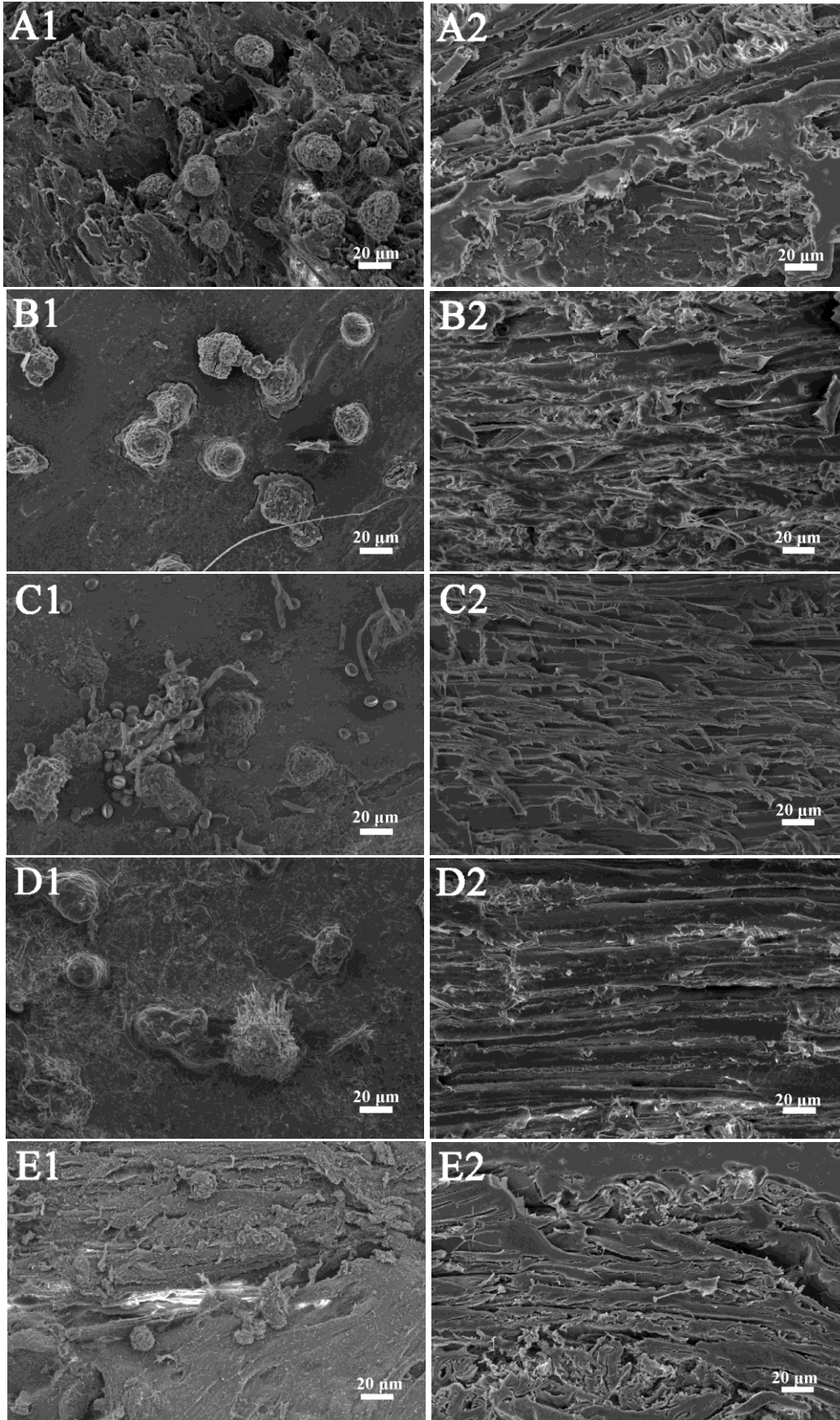
**Table 2.** The Algal Resistance Level of the Different WPC Species

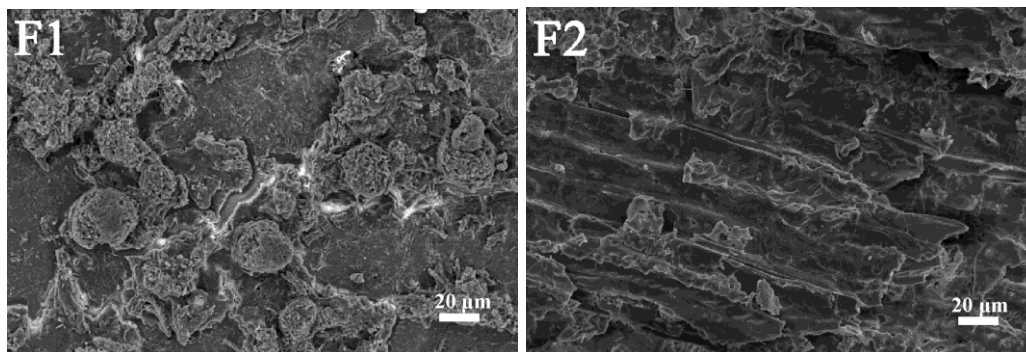
WPC Group	Resistance Level	Change rate of mass (%)
<i>Liquidambar formosana</i>	1	-13.64 ± 0.73
<i>Cunninghamia lanceolata</i>	2	-7.17 ± 0.20
<i>Eucalyptus grandis</i> × <i>E. urophylla</i>	4	-18.80 ± 0.66
<i>Pinus massoniana</i>	4	-11.43 ± 0.98
<i>Melaleuca leucadendra</i>	2	-15.47 ± 1.14
<i>Ricinus communis</i>	3	-14.41 ± 0.38
<i>Eucalyptus grandis</i> × <i>E. urophylla</i>	0	-102.73 ± 3.17
<i>Eucalyptus grandis</i> × <i>E. urophylla</i>	0	-93.43 ± 6.15

### The Surface and Fracture Micro-Morphology Analysis

The surface and fracture micro-morphology of the different WPC samples (six wood species) are shown in Fig. 2. As shown, the algal colonization on the sample surfaces was clearly shown, and the algal spores appeared in the aggregation morphology (Fig. 2. (A1, B1, C1, D1, E1, and F1)). However, it was noticed that minimal hyphae and spores existed on the inner fracture layers for all of the groups in Fig. 2. (A2, B2, C2, D2, E2, and F2).

Although there were only a few micro-voids between the wood particles and thermoplastic resins, channels for the movement of spores and hyphae into the samples were invalid because of the formation and aggregation during the growing process. This also provided evidence that the algae mainly affected the appearance of the WPC samples, not the inner structure and quality.



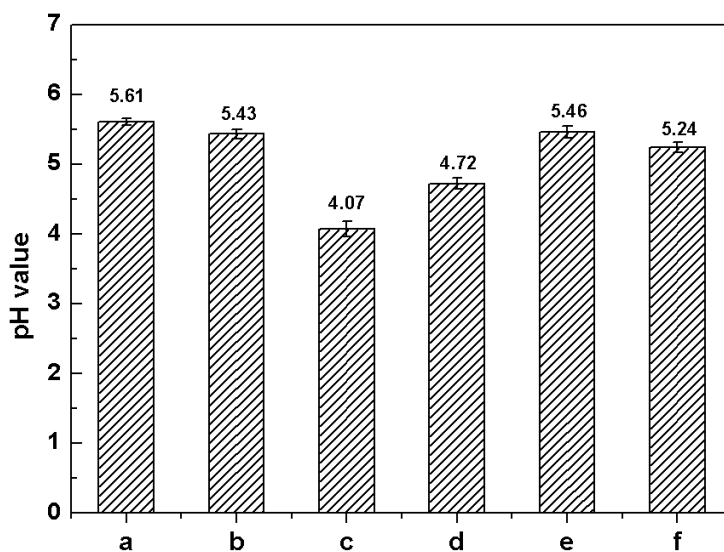


**Fig. 2.** The micro-morphology of the algal resistance test for the WPC species: (A) *Liquidambar formosana*, (B) *Cunninghamia lanceolata*, (C) *Eucalyptus grandis* × *E. urophylla* (D) *Pinus massoniana*, (E) *Melaleuca leucadendra*, and (F) *Ricinus communis*. For each group, 1 represents the surface morphology and 2 represents the fracture morphology

### The pH Value Analysis on the Different Wood Species

Figure 3 shows the pH values for the six wood species that were selected for the WPC preparation. It is known that most wood species were acidic, which is consistent with the results from this study. It was found that there was a close relationship between the pH value and the algal resistance level, whereas, the higher the pH value, the greater the algal resistance level in the WPC samples.

Specifically speaking, the wood sample with the highest pH value (5.61) was *Liquidambar formosana*. Its algal resistance ability was also the strongest (level 1) of the wood groups. Conversely, the *Eucalyptus grandis* × *E. urophylla* and *Pinus massoniana* woods exhibited relatively lower pH values of 4.07 and 4.72, respectively, corresponding to weaker resistance ability (level 4). The other groups demonstrated similar relationships. Therefore, it can be concluded that algal growth and propagation were more suitable for relatively acid conditions at the same temperature, humidity, and intensity of illumination. It was speculated that the optimum range of pH value of wood species for rapid growth of algae was between 4 and 5 in accordance with the experimental results.



**Fig. 3.** The pH values of the six wood species: (a) *Liquidambar formosana*, (b) *Cunninghamia lanceolata*, (c) *Eucalyptus grandis* × *E. urophylla*, (d) *Pinus massoniana*, (e) *Melaleuca leucadendra*, and (f) *Ricinus communis*



Additionally, previous literature also indicated that the phenolic hydroxyl group engenders an inhibition effect on algal growth. Algal cells die because of the change in and destruction of the cell membrane structure by phenolic hydroxyl groups (Einhellig 1995; Everall and Lees 1997; Li *et al.* 2010). This study demonstrated that there were chemically volatile substances in the different wood species. The phenolic hydroxyl groups mentioned may have been present in the volatile chemical substances of the *Liquidambar formosana* wood, *i.e.*, 3-hydroxy-4-methoxy-benzaldehyde, 3,4,5-trimethoxy-phenol, and 4-hydroxy-3,5-dimethoxy-benzaldehyde (Xu *et al.* 2015). Such a mechanism is consistent with the highest algal resistance level being found for the *Liquidambar formosana*-based WPC.

## CONCLUSIONS

1. The algal resistance levels of different wood-based composites were ranked as: conventional wood-based composites (MDF and plywood) > novel wood-based composites (WPC) of the same wood material.
2. The algal resistance levels of WPC groups made from various wood species were ranked as: *Liquidambar formosana* > *Cunninghamia lanceolata* and *Melaleuca leucadendra* > *Ricinus communis* > *Eucalyptus grandis* × *E. urophylla* and *Pinus massoniana*. The algal colonization had a negative effect only on the appearance and aesthetic appeal of the samples.
3. It was found that there was a correlation relationship between the pH value and the algal resistance level. Therefore, the higher the pH value, the greater the algal resistance level of the WPC samples. Moreover, some volatile chemical substances were deduced to have an inhibitory effect on the algal resistance of WPC.

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