Water Absorption, Dimensional Stability, and Mold Susceptibility of Organically-modified-Montmorillonite Modified Wood Flour/Polypropylene Composites

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Wood flour (WF) was modified by sodium-montmorillonite (Na-MMT) and didecyl dimethyl ammonium chloride (DDAC) in a two-step process to form organically-modified-montmorillonite (OMMT) inside the WF with varied MMT concentration (0.25, 0.5, 0.75, and 1%, respectively). Then, the modified WF was mixed with polypropylene (PP) to produce WF/PP composites. The WF and WF/PP composites were characterized, and the water absorption, dimensional stabilities, and the mold susceptibility of the composites against *Aspergillus niger*, *Penicillam citrinum*, and *Trichoderma viride* were investigated. The results showed that Na-MMT was successfully transformed to OMMT inside WF. Owing to the hydrophobic nature and barrier effect of OMMT on water permeability, the composites showed some improvements in water resistance, dimensional stabilities and antibiotic performance. MMT concentration was also an important factor. The water repellency and dimensional stability were improved with increasing MMT concentration at first and then dropped after the MMT concentration exceeded 0.5%. However, the mold resistance of the composites increased along with increasing MMT concentration. With 1% MMT treated, the mold growth rating decreased to 1 (mold covering of 0-25%). These results suggested that OMMT modified WF had a positive effect on restricting water absorption, swelling, and mold susceptibility for the WF/PP composites.

Keywords: Wood flour/polypropylene composites; Organically-modified-montmorillonite (OMMT); Water absorption; Dimensional stability; Mold susceptibility

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INTRODUCTION

Wood plastic composites (WPC) usually refer to the composites containing wood (or natural) flour/fibers (WF) and thermoplastics. Owing to their low cost, high specific strength, and renewability, WPC products have gained market share in various applications such as decks, railings, and automotive parts (Smith and Wolcott 2006). Rapid growth in the WPC market has fueled the development of new applications in construction and for building materials. In exterior applications, especially under humid conditions, these materials require high degrees of water repellency, which benefits both dimensional stability and fungal resistance. Although the hydrophobic thermoplastic matrix slows the uptake of water, moisture absorption and fungal attack remain important concerns for WPC (Lomelí-Ramírez *et al.* 2009). The hydrophilic character of WF itself and gaps at poor interfaces between WF and polymer matrix are pathways for water and microorganisms into WPC. Moisture infiltration into WPC causes swelling, which stresses the interfacial bond between WF and polymer matrix. Repeated swelling and shrinking over time eventually lead to microcracks in the composites, promoting the growth of decay fungi and mold. Thus, the strength of the composites as a whole is weakened (Schirp and Wolcott 2005). Accordingly, to reduce the effect of water, there are two essential strategies available: modifications of WF that change its chemical structure (Pelaez-Samaniego *et al.* 2013) or addition of coupling agents such as silane or maleic anhydride grafted polymers (Wang *et al.* 2011; 2012). Up to the present, various treatments have been tried as ways of decreasing the hygroscopicity of WF, such as acetylation (Segerholm *et al.* 2012), benzylation (Dominkovics *et al.* 2007), hydrophobation treatment (Xie *et al.* 2013), hemicelluloses extraction (Hosseinaei *et al.* 2012), and heat treatment (Luo *et al.* 2013). However, most of these methods are not satisfying because of the relatively complicated processing, high cost, or unfavorable damages to WF.

Nanocomposite technology with layered silicate nanoclays as *in situ* reinforcement has been intensively investigated in recent years (Messersmith and Giannelis 1995; LeBaron *et al.* 1999). It offers new opportunities for the modification of wood products and polymers. Among these silicates, montmorillonite (MMT) is the most widely used types of nanoclay because of its natural occurrences and beneficial properties (high cationic exchange capacity, high surface area, and large aspect ratio) (Utracki *et al.* 2007). It is a layered silicate with a mean layer thickness of 0.96 nm. The simple chemical component of MMT is $Al_2O_3 \cdot 4SiO_2 \cdot 3H_2O$ with two layers of a tetrahedral layer of Si-O sandwiching one central layer of octahedral Al-O. However, the quality of dispersion of nanoparticles depends on the extent of compatibility between the nanoparticles and the wood or polymer matrix and can result in a microphase-separated, an intercalated or an exfoliated morphology (Krikorian and Pochan 2003). To form a true nanocomposite, an exfoliated structure is expected. Cai *et al.* (2007; 2008) ground MMT with a ball-mill and then mixed the nanoclay with water-soluble melamine-urea-formaldehyde resin and impregnated the mixture into the wood. They found significant improvements in physical and mechanical properties of the composites compared with untreated wood, such as density, surface hardness, modulus of elasticity, and especially the dimensional stability and water repellency. MMT is also an efficient and promising enhancer for polymers even when the content is less than 10% (*wt.*) (Cheng *et al.* 2003; Lewitus *et al.* 2006). Du *et al.* (2013) studied the water permeability of MMT/polylactide composites and found that the water permeation was 34% less than in the neat polymer at an MMT content of 10 wt%. According to Drozdov *et al.* (2003), the decrease in moisture diffusion was associated with confinements of molecular mobility caused by the clay filler. In recent years, MMT has been applied into WPC by direct ternary melt blending (Zhong *et al.* 2007; Sui *et al.* 2009). According to their results, MMT dispersed and intercalated with the polymer fraction rather than WF. Therefore, the enhanced mechanism was similar to that in function with polymers alone. Compared with sodium-montmorillonite (Na-MMT), organically-modified-montmorillonite (OMMT) had a better effect on the physical and mechanical properties of WPC owing to the hydrophobic effect and exfoliated structure of OMMT (Liu *et al.* 2013a). In addition, OMMT had a synergic effect with coupling agents to further improve the properties of WPC (Liu *et al.* 2013b). Consequently, OMMT serves as a better choice than natural MMT to form high-performance composites.

In a previous study (Liu *et al.* 2013c), the authors used Na-MMT and didecyl dimethyl ammonium chloride (DDAC) to modify WF in a two-step process to form

OMMT inside the WF, which made WF as a “reinforcement” rather than a “filler”. After that, the modified WF was mixed with poly(lactic acid) (PLA) to prepare WF/PLA composites. Significant improvements were observed in physical and mechanical properties of the composites at a suitable OMMT concentration (lower than 1%). Meanwhile, the enhancements were more obvious in coarse WF than fine WF. WF/polypropylene (PP) composite is one of the most widely used types of WPC on the market. However, there are some drawbacks of WF/PP composites such as their low dimensional stability and mold resistance in the presence of water. In this study, WFs were modified by the same process and then combined with PP to make WF/PP composites. The goal was to restrict the water penetration and fungal attack, two issues that are very important for broadening the applications of WF/PP composites. Therefore, the water absorption, dimensional stability, and mold susceptibility of the composites were investigated. X-ray diffraction (XRD) and Fourier transform infrared spectrum (FTIR) analyses were carried out to confirm the successful transformation of OMMT. After mold test, the surface of the composites were observed by using scanning electron microscope (SEM) for further explanation.

EXPERIMENTAL

Materials

The wood flour of poplar (*Populus tomentosa* Carr.) with size 10 to 60 mesh was supplied by Xingda Wood Flour Company, Gaocheng, China. It has an average length of 1.5 mm and average diameter of 0.2 mm. Polypropylene (PP) (K8303; Sinopec Chemical Products Sales Company, China) with a density of 0.9 g/cm^3 was purchased from Beijing Yanshan Petrochemical *Co. Ltd*., China. It had a melt point around 165 °C and a melt flow index of 1.5 to 2.0 g/10 min at 230 °C. Na-montmorillonite (Na-MMT) was purchased from Zhejiang Fenghong Clay Chemical *Co. Ltd.*, Huzhou, China. It is a hydrophilic clay powder with volatiles below 4% at 105 °C for 2 h. The viscosity of 3% w/w Na-MMT in distilled water is 3000 mPa·s with pH of 8 to 10. The mean interlayer distance of Na-MMT is 1.459 nm with particle sizes of 76 μm. The cation exchange capacity of Na-MMT is 90 mmol/100 g. The modifier was didecyl dimethyl ammonium chloride (DDAC, 70%), which was purchased from Shanghai 3D, Bio-chem Co. Ltd., Shanghai, China. Polyfluotetraethylene (PTFE) films were used as demoulding materials to avoid sticking board during hot-pressing.

Modification of Wood Flour

Prior to use, the WF was dried in an oven at 103±2 ^{o}C for 5 h to 1-2% moisture content. The first step was to impregnate WF with Na-MMT suspensions at four concentrations (0.25, 0.5, 0.75, and 1%, respectively) through a vacuum-pressure process. First, WF was put into a beaker in a treating tank, and then it was vacuum-treated at 0.01 MPa for 30 min. Afterwards, Na-MMT suspension, which was dissolved in the distilled water, was let in to completely submerge WF with pressure treated at 0.6 MPa for 1 h. The average particle size of Na-MMT suspension was 1.215 μm, as tested by Laser Particle Analyzer (Delsa^{TM} Nano C, Beckman coulter, USA). The impregnated WF was taken out from Na-MMT suspension with a 100-mesh sieve and dried in an oven at 103±2 ^{o}C to constant weight. The weight percent gains of treated WF were 0.18, 0.62, 3.84, and 6.25%, respectively. The treated WF was then put into a beaker and submerged in DDAC solution in the second step. The concentration of DDAC was calculated according to the concentration of Na-MMT at a ratio of the cation exchange capacity of Na-MMT of 0.7:1 according to formulations given in Table 1. The beaker was placed in a water bath at 60 °C for 2 h with a mechanical stirring at speed of 80 r/min. Then, WF was taken out and dried at 103±2 °C to constant weight. The weight percent gains of modified WF added another 3 to 3.5% compared to the first step. Besides, the "unmodified" control WF was treated with distilled water by using the same process.

Table 1. Compositions and Labeling of the Studied Formulations

Label	MMT concentration (%)	DDAC concentration (%)	WF (wt%)	PP (WT%)
0 (control)	0	0	50	50
0.25	0.25	0.08	50	50
0.5	0.5	0.16	50	50
0.75	0.75	0.24	50	50
1	1	0.32	50	50

Preparation of Composites

The raw materials for WF/PP composites contained 50 wt% of unmodified or modified WF and another 50 wt% PP. They were mixed in a high speed blender at about 2900 rpm for 4 min. The mixture was then dried at 103±2 °C for 2 h. After drying, the mixture was extruded via a co-rotating twin-screw extruder (KESUN KS-20, Kunshan, China) with a screw diameter of 20 mm and a length-to-diameter ratio of 36/1. The corresponding temperature profile along the extruder barrel was 165/170/175/180/175 °C, and the screw speed was 167 rpm. The extruded rods were cut into small particles about 5 mm, and then the blends were dried again at 103±2 °C for 2 h and taken out for hand matting. A hot press (SYSMEN-II, China Academy of Forestry, Beijing, China) was used to produce the composites by compressing the mat at 180 °C with a pressure of 4 MPa for 6 min. The target density of the composites was 1.0 g/cm^3 at size of 270 mm × 270 mm × 3 mm. Prior to demoulding, the formed mat was cooled down at 4 MPa for another 6 min at room temperature. After that, all the mats were cut into required dimensions for further tests.

Characterization of WF and WF/PP Composites

The X-ray diffraction (XRD) analyses of Na-MMT, neat PP, unmodified and 1% MMT modified WF, and their PP composites samples were carried out on an X-ray 6000 (Shimadzu, Japan) machine. All the samples were first ground to pass an 80-mesh sieve and air-dried before analysis. The X-ray beam was Cu-Kα (λ=0.1540 nm) radiation, operated at 40 kV and 30 mA. The scanning rate was 2°/s, and 2θ ranged from 2° to 40° with the rotational speed of 30 rpm.

The chemical groups of unmodified and 1% MMT modified WF and their PP composites samples were examined by Fourier transform infrared spectrum analysis spectrometer (FTIR, BRUKER Vertex 70v, Germany), and potassium bromide (KBr) was used to collect the background. Air-dried powder, passed through a 100-mesh sieve, was mixed with KBr in a weight ratio of 1:100 before spectrum collection. All spectra were displayed in wavelengths ranging from 400 to 4000 cm^{-1}.

Water Absorption and Dimensional Stability

The determination of water absorption was carried out according to the Chinese standard GB/T 17657-1999 (1999). Composites samples with dimensions of 50 mm × 50 mm × 3 mm (thickness) were completely immersed into distilled water at 20±2 ^{o}C for 8 days. After immersions for 6, 24, and 48 h and thereafter at 48 h intervals, the samples were taken out with removal of excess water on the surface and weighed. The water uptakes were calculated based on the weight percent gains at each stage. The thickness swelling rates were calculated based on the mid-span thickness changes until the final. Four replicates of the samples were used for each group, based on which the standard deviations were calculated.

The dimensional stability test was carried out according to the Chinese standard GB/T 17657-1999 (1999). The dimension of the composite samples was 140 mm × 13 mm × 3 mm (thickness). Prior to the test, the samples were kept in a conditioning chamber with a set relative humidity of 65% and a temperature of 20 ^{o}C for 4 days. The original dimensions of the samples were recorded. The dimensional stability was indicated by both shrinkage and swelling rates. To test the shrinkage rate, the samples were dried at 70±2 ^{o}C in an oven for 24 h. After cooling down in a desiccator for 1 h, the samples were weighed and their dimensions were measured. For swelling tests, the samples were placed in a hermetic jar filled with sodium tartrate ($(CHOHCOO)_2Na_2{\cdot}2H_2O$) saturated solution for 48 h at 40 ^{o}C, which corresponded to a high relative humidity of 90 to 95%. Afterwards, they were taken out and cooled down in a desiccator for 1 h for weighing and dimension measuring. The moisture desorption and adsorption was calculated based on the weight change. The shrinkage and swelling rates were calculated based on the change of length. Six replicates were used for each group, based on which the mean values and standard deviations were calculated.

Mold Susceptibility Test

The mold susceptibility of the WPC samples was evaluated according to the Chinese standard GB/T 18261-2000 (2000). The mold fungi were separately inoculated on a culture medium, a mixture of tomato juice and glucose, in petri dishes and incubated at 25 ^{o}C for 7 days. The fungi tested involved *Aspergillus niger*, *Penicillam citrinum*, and *Trichoderma viride*. After the fungi in petri dishes had developed spores, a sterilized U-shape glass rod (3 mm in diameter) was placed over the substrate. Two samples were placed on the glass rod in each petri dish. The petri dishes were placed in an incubator with the temperature maintained at 25 ^{o}C and the relative humidity close to 100%. After 4 weeks of exposure, the samples were visually rated on a scale of 0 to 4 according to the extent and intensity of mold growth on the surface. The ratings of 0 to 4 indicated the mold coverings of 0%, 0-25%, 25-50%, 50-75%, 75-100% of the samples, respectively. Six replicates were used for each mold test group.

SEM Analysis

The morphologies of unmodified WF/PP composites samples after mold test and 1% MMT modified WF/PP composite sample against *Penicillam citrinum* were observed with a Hitachi (S-3400, Japan) scanning electron microscope (SEM) with an acceleration voltage of 10 kV. The samples were sputter-coated with gold prior to observation.

RESULTS AND DISCUSSION

XRD Analysis

The X-ray diffraction (XRD) scans for sodium-montmorillonite (Na-MMT), neat PP, and unmodified and 1% montmorillonite (MMT) modified WF and WF/PP composites are shown in Fig. 1. In Fig. 1a, the positions of the diffraction peaks for crystal planes (101), (002), and (040) in cellulose for WF at $2\theta=17^o$, 22.5^o, and 35^o did not change after modification with organically-modified-montmorillonite (OMMT), suggesting that the modification did not influence the main crystalline structures of WF. Similarly, the modified WF showed little effect on crystalline structures of WF/PP composites by showing the same position of the diffraction peaks of (110), (040), (130), and (111) for α-PP at $2\theta=14^o$, 17^o, 18.5^o, and 21^o (Fig. 1b). However, a new peak was found at $2\theta=3.26^o$ after OMMT modification in both WF and WF/PP composite, which was related to the crystalline structure of OMMT with the d-spacing of 2.707 nm. The d-spacing of OMMT modified with Na-MMT and DDAC was 2.323 nm. Therefore, a conclusion could be drawn that Na-MMT was transformed to OMMT after the two-step treatment process. Meanwhile, the enlarged d-spacing of OMMT indicated that WF might be intercalated into OMMT layers. However, there was some Na-MMT residue left, as evidenced by a peak at $2\theta=6.14^o$ both in the modified WF and WF/PP composite.

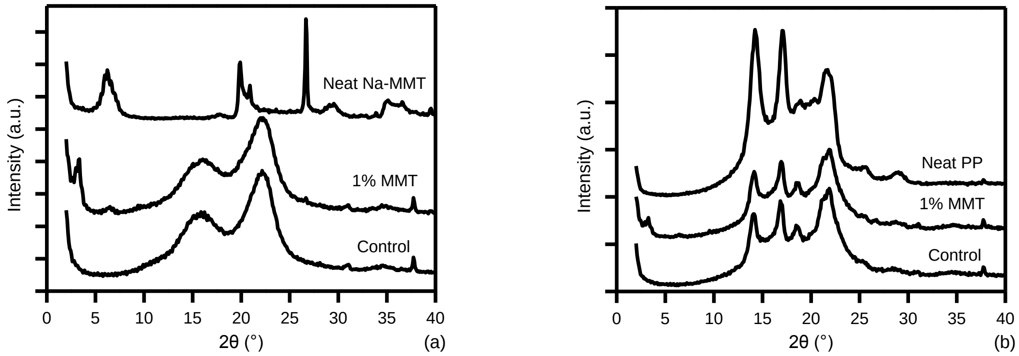


Fig. 1. XRD patterns of neat Na-MMT, WF (a), neat PP and WF/PP composites (b)

FTIR Analysis

FTIR results of unmodified and 1% MMT modified WF and their composites with PP are shown in Fig. 2. The peaks of WF with OMMT were almost the same as pure WF except for two new peaks showing up at 519 cm^{-1} and 461 cm^{-1}, which should be assigned to the absorption of Si-O-M (metal cation) and M-O group of OMMT. Besides, the shoulder peaks at 2900 cm^{-1} and 2850 cm^{-1} assigned to the symmetrical and anti-symmetrical stretching of C-H were more obvious in OMMT modified WF than WF control, which provided the evidence of the presence of DDAC long organic hydrocarbon chain. The peak in OMMT modified WF at 1035 cm^{-1} was sharper than for pure WF. It should be attributed to the stretching of Si-O-Si group, which overlapped the stretching of C-O-C groups in WF. These results showed that OMMT had been introduced into WF matrix but did not change the main components of WF. However, for WF/PP composites, these differences were insignificant except for peaks at 519 cm^{-1} and 461 cm^{-1}. This could be explained by the limited amount of OMMT on the whole. The FTIR results were consistent with the XRD results.

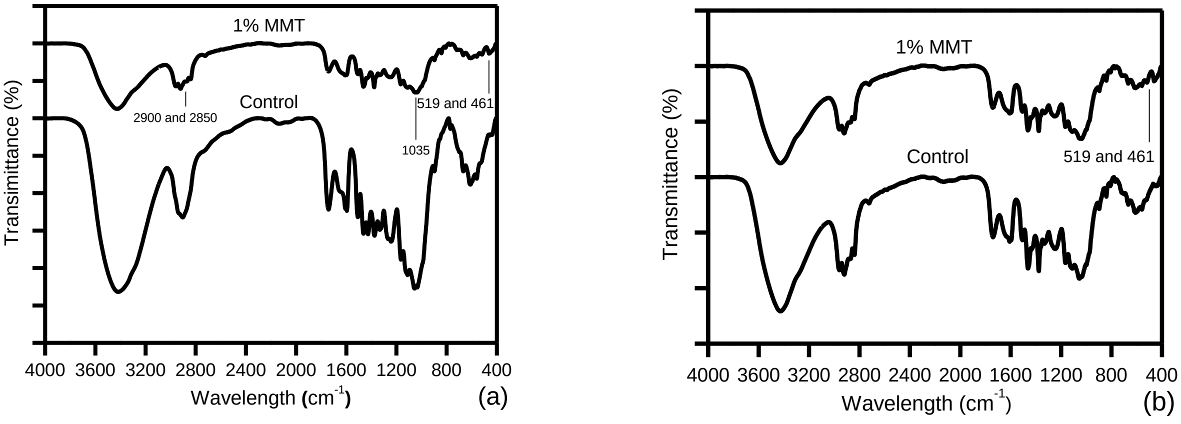


Fig. 2. FTIR curves of WF (a) and WF/PP composites (b)

Water Uptake and Thickness Swelling

The results of water absorption tests are shown in Fig. 3. The water uptakes of WF/PP composites prepared with OMMT modified WF were significantly less than that of the WF/PP control (Fig. 3a). Irrespective of OMMT concentration, OMMT modified WF/PP composites absorbed about 3.5%, while the WF/PP control absorbed 4.6% after 196 h of immersion, with a reduction in water uptakes of over 20% in all cases. This might be due to the barrier effects of the silicate fillers inhibiting the water absorption (Drozdov *et al.* 2003) or the reduction of accessible OH groups in modified WF as proved in our previous study (Liu *et al.* 2013c).

Water in WPC can exist in three regions, namely, the WF lumen, WF cell walls, and gaps between filler and matrix in the interface region (Karmaker 1997). The reduction of OH groups in WF could lead to a decrease in hygroscopicity of WF. Meanwhile, the combination of PP with modified WF could be stronger than pure WF. The improved compatibility of matrix and filler could reduce the gaps between them and limit the penetration of water into these gaps. These all contributed to the water repellency of WF/PP composites. The objective of reducing OH groups in WF and improving interfacial adhesion between WF and polymer matrix have been considered by other researches (Dominkovics *et al.* 2007; Luo *et al.* 2013; Xie *et al.* 2013). For different MMT concentration groups, it seemed that the 0.5% MMT modified group absorbed the least water. The increase in water uptake at high OMMT loadings might be caused by the agglomeration of OMMT on WF surface, which might cause weak interface bonding to the composites.

Similar to the results of water uptake tests, OMMT-modified groups exhibited lower thickness swelling values than the control group, suggesting that the introduction of silicate fillers was a positive factor to restrict the hydro-expansion of WF/PP composites. The thickness swelling rates decreased with the increase of MMT concentration until reaching the minimum value of 2.6% at 0.5% MMT concentration, which is nearly one-fourth of that of the WF/PP composite control. The increase in thickness swelling rate after 0.5% MMT concentration could be due to the poor interfacial adhesion at excessive OMMT loading on WF surface.

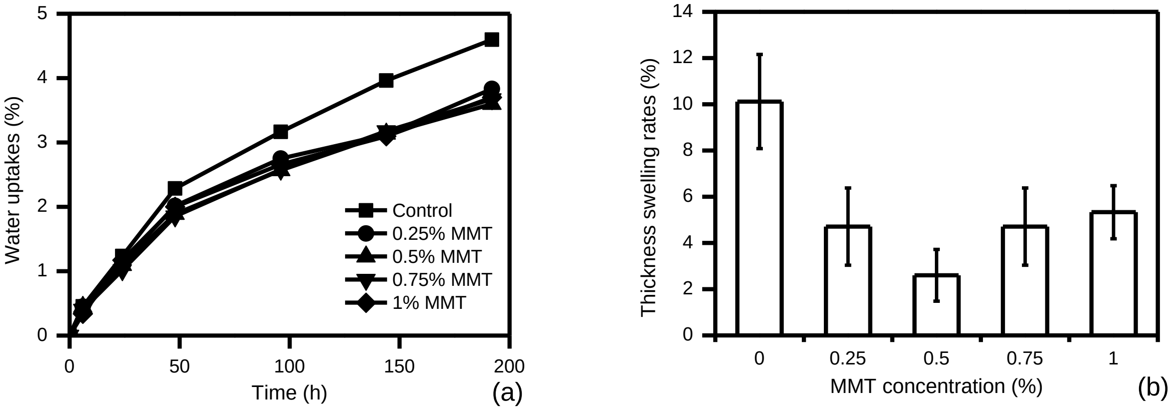


Fig. 3. Water uptakes (a) and thickness swelling rates (b) of WF/PP composites

Moisture Sorption and Dimensional Stability

The dimensional stability results are similar to the water uptake results, as listed in Table 2. Since the moisture sorption of WF/PP composites only occurred in the WF cell wall but not in the cell cavity, the desorption and adsorption values of moisture for WF/PP composites were very low, *i.e.* 0.52 to 0.63% and 1.88 to 2.16%, respectively. These values were much lower than those of solid wood (around 12% and 25%). As a result, these composites showed very good dimensional stabilities including both anti-shrinkage and anti-swelling performance. This could be explained by the envelopment of WFs by the hydrophobic polymer matrix. OMMT modification further hindered the moisture sorption and dimensional changes of WF/PP composites. Among all groups, 0.5% MMT modified WF/PP composite showed the best dimensional stability.

Table 2. Moisture Sorption and Dimensional Stability of WF/PP Composites

MMT concentration (%)	Desorption Process		Adsorption Process	
	Weight percent losses (%)	Length shrinkage rates (%)	Weight percent gains (%)	Length swelling rates (%)
0 (control)	0.63 (0.03)*	0.18 (0.04)	2.16 (0.04)	0.31 (0.10)
0.25	0.60 (0.02)	0.16 (0.03)	2.12 (0.06)	0.26 (0.10)
0.5	0.52 (0.02)	0.11 (0.03)	1.88 (0.04)	0.12 (0.06)
0.75	0.58 (0.04)	0.13 (0.07)	1.98 (0.04)	0.20 (0.07)
1	0.58 (0.02)	0.14 (0.01)	2.00 (0.11)	0.19 (0.08)
* The values in parentheses represent the standard deviations of six replicates.				

Mold Susceptibility Test

The results of the mold susceptibility test are shown in Fig. 4. For all mold species, the WF/PP composites were infected on their surfaces after 4 weeks of mold exposure. Low OMMT loadings showed little effect on the mold resistance of WF/PP composites by showing a mold growth rating of 4, which was the same as the WF/PP control, suggesting these composites were easily attacked by mold. However, when the MMT concentration was increased to 0.5%, the mold growth ratings of WF/PP composites were reduced. The decrease in mold growth rating on OMMT-modified WF/PP composites could be associated with two factors: (1) OMMT inhibited the moisture penetration into WF/PP composites because of its hydrophobicity and barrier effect; (2) the modifier DDAC is also a kind of antiseptic used in many preservative systems such as ammoniacal copper quat (ACQ), which could improve the fungicidal properties of wood products (Kartal *et al*. 2005). At 0.5% or 0.75% MMT concentrations, it seemed that OMMT was more effective against *Aspergillus niger* and *Trichoderma viride* than *Penicillam citrinum*. However, when MMT concentration reached 1%, the mold growth ratings for varying mold species were all around 1, showing an acceptable level of prevention.

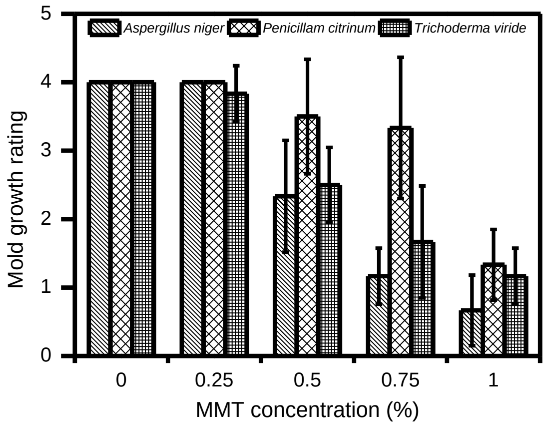
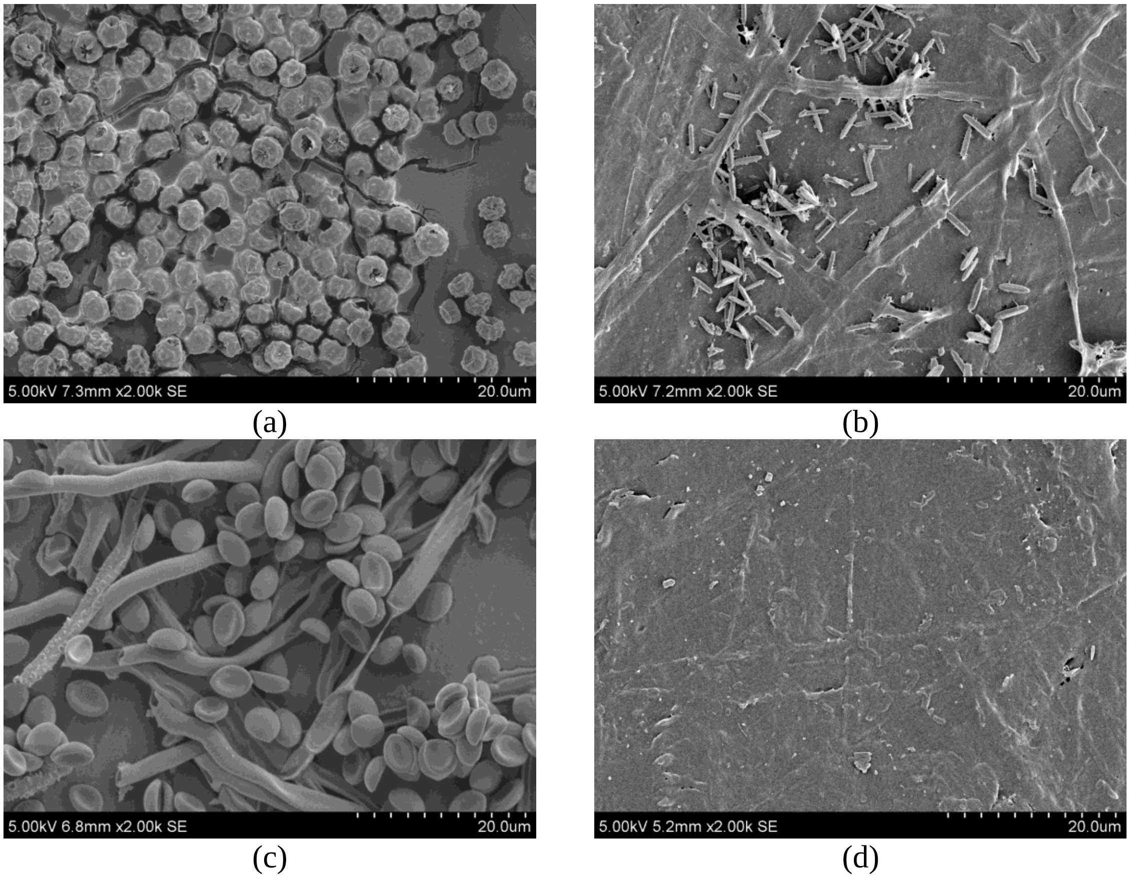


Fig. 4. Mold growth rating of WF/PP composites



(a) (b)

(c) (d)

Fig. 5. Surface of WF/PP composites after mold susceptibility test; (a)-(c) WF/PP composite controls infected with *Aspergillus niger, Penicillam citrinum, and Trichoderma viride*, respectively; (d) 1% MMT modified WF/PP composite infected with *Penicillam citrinum.*
The scale bars show 20 µm lengths.

SEM Analysis

The surfaces of WF/PP composites after mold susceptibility test are shown in Fig. 5. It was obvious that WF/PP composites without OMMT treatment were susceptible to attack by fungi mold. The surfaces exhibited the mold spores of spherical *Aspergillus niger* or flat-ellipsoidal *Trichoderma viride* or rod-like *Penicillam citrinum.* Besides, there were some cracks visible on the surfaces, which might be due to the expansion between the surface and interior layers. The surface of composite with 1% MMT treated was very clear. No obvious fungi were found after the mold susceptibility test.

CONCLUSIONS

1. The two-step organically-modified-montmorillonite (OMMT) modified wood flours (WFs) were successfully applied in polypropylene (PP) to produce modified WF/PP composites with improved water repellency, dimensional stability, and mold susceptibility at suitable MMT concentrations. At MMT concentrations below 0.5%, the moisture resistance and dimensional stabilities of WF/PP composites were improved significantly. After introducing more OMMT into WF/PP composites, these enhancements were weakened because of the gathering of OMMT on WF surface. However, it still performed better than the WF/PP control.
2. OMMT modified WF inhibited the mold growth on composite surfaces, which might be due to the reduction of moisture content and the presence of didecyl dimethyl ammonium chloride (DDAC) antiseptic. At higher OMMT loading, this effect was more obvious according to this study.

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