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Effects of Two Types of Clay on Physical and Mechanical Properties of Poly(lactic acid)/Wood Flour Composites at Various Wood Flour Contents

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ABSTRACT: To investigate the effects of two types of clay, namely, Na-montmorillonite (Na-MMT) and organic-montmorillonite (OMMT), on poly(lactic acid) (PLA)/wood flour (WF) composites, some physical and mechanical properties including the water sorption, thickness swelling, flexural modulus of rupture (MOR), and modulus of elasticity (MOE) of PLA/WF composites at different WF contents of 0, 20, 40, and 60 wt% were tested in this study. The results showed that: (1) the 24 h water uptake and thickness swelling increased and the flexural MOR and crystallinity decreased with the increasing WF content, whereas the flexural MOE of the composites increased with WF content up to 40 wt% but decreased sharply at WF content of 60 wt%; (2) the addition of Na-MMT slightly increased the 24 h water uptake as well as the thickness swelling rate below 40 wt%, whereas OMMT reduced the thickness swelling at higher WF contents (40, 60 wt%) although it showed little effect on 24 h water uptake; (3) both Na-MMT and OMMT could improve the flexural MOR and MOE of PLA/WF composite at WF contents below 40 wt%, and OMMT resulted in more obvious improvement than Na-MMT. However, they both showed negative effect at WF content of 60 wt%; (4) XRD and FT-Raman analysis suggested that clays would be attached more on the surface of the WF rather than diffused in the PLA matrix at a higher WF content (60 wt%); (5) SEM analysis proved that the interfacial adhesion of PLA and WF became poorer at WF content above 40 wt%, whereas it could be improved by OMMT modified. © 2012 Wiley Periodicals, Inc. J. Appl. Polym. Sci. 000: 000–000, 2012

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INTRODUCTION

Because of the increasing environmental concerns and restricted availability of petrochemical resources, biopolymers became a strong interest in the public research during the recent decades. Poly(lactic acid) (PLA), as one of this type, is produced by its monomers, lactic acids which are obtained from the fermentation of sugar feed stock.[1] The synthesis of PLA from lactic acid is followed by a ring-opening polymerization of the cyclic lactide dimmer. However, due to its complicated process of production, the price of PLA is much higher than other polymers such as polyethylene (PE), polypropylene (PP), poly(vinyl chloride) (PVC), and polystyrene (PS). Furthermore, its brightness, low deformation-at-break, and limited processing approaches restrict its widespread applicability.[2] Numerous studies have been focused on improving the properties as well as reducing its price by blending PLA with some natural fiber materials,[3–6] synthetic monomers and polymers like polyethylene glycol (PEG)[7], glycerol[1] maleic anhydride (MA)[8] and many kinds of inorganic fillers.[9–12]

Wood flour (WF) usually made by conventional grinding is gaining increasing acceptance as a kind of fillers for polymers since it offers many advantages such as low density, high stiffness, biodegradation, easy availability, renewability, and relatively low cost. It has been used as raw material filling some polymers as reinforcement of wood-plastic composites for a long time. The addition of WF enhances some physical, mechanical and thermal properties, such as rigidity, stiffness, strength, hardness, and heat resistance.[13–15]

Clay as another kind of filler reinforcement has attracted a great deal of interest both for wood products and polymers. Among all the clays, montmorillonite (MMT) is one of the most widely used type. It is a layered silicate with a mean layer thickness of 0.96 nm. The simple chemical components of MMT was $Al_2O_3{\cdot}4SiO_2{\cdot}3H_2O$ with two layers of tetrahedron of Si–O and one layer of octahedron of Al–O in between. There are three types of structures when the layered silicate is introduced into wood or in polymer matrix: (1) a phase-separated structure; (2) an intercalated structure; and (3) an exfoliated or delaminated structure.[16] An exfoliated structure is always expected to form a true nanocomposite. Zhao and Lv[17] proposed a possibility to prepare wood/clay composites by introducing clay into the nanoscale wood cavities. Then, they successfully prepared MMT/wood composite by using phenol-formaldehyde resin as the intermediate and found that some exfoliated clay entered the amorphous region of wood cell wall and the strength of wood after treatment increased.[18] Wang et al.[19] used organic-montmorillonite (OMMT) modified by didecyl dimethyl ammonium chloride to treat wood and found that the compression strength of treated wood increased up to 27% compared with untreated control and the decay resistance of the composite was also improved. Jiang et al.[20] studied the micronized clay suspension by combining OMMT with polyvinyl alcohol (PVA) to treat wood, and the results showed that the water sorption, dimension stability, and surface hardness of treated wood were greatly improved. While for the polymers, the clay also played a significant role in property enhancements even at contents lower than 10 wt% by acting as a nucleating agent, increasing the crystallinity and strength.[21–23] In recent years, clay has been used in wood-plastic composites for modification, and also got a lot of achievements. Zhong et al.[24] studied the organoclay in wood/PE composite and found that the incorporation of organoclay in the composite reduced the thermal expansion and increased the heat deflection temperature without affecting the processability. Matuana[25] studied the effects of two types of clays on wood/PVC composites. He proved that the intercalated structure of clay in PVC matrix and PVC tensile strength of the onset diffusion of unmodified clay in PLA composites was improved about 10 MPa and the elongation-at-break was rising up to 128% by using X-ray diffraction (XRD) analysis and transmission electron microscopy (TEM). He also found that the unmodified inorganic clay performed better than the organo-modified clay when the same processing method was used. Lee and Kim[26] prepared wood/PP/clay composites by melt blending and found that the PP matrix of the WPC were intercalated by organoclay and the addition 1% of organoclay improved the tensile properties, impact strength, thermal stability, glass transition temperature, and crystallinity of the WPC system slightly while the storage modulus increased considerably. Hemmasi et al.[27] obtained the results that the higher content of clay [>6 phc (per hundred compounds)] may resulted in decreasing of mechanical properties, such as flexural strength, tensile modulus, and elongation at break.

As mentioned above, a lot of investigations have been performed on clay reinforced wood flour/polymer composites; however, the effects of clay, especially different types of clay (hydrophilic and hydrophobic clays), on PLA/WF system at different WF contents have not been studied yet. Therefore, this study proposed to use both WF and clay on PLA to make a cost-effective and high-performance PLA/WF composite by comparing the physical properties (water absorption and thickness swelling), mechanical properties (flexural MOR and MOE) of the PLA/WF composites modified by two types of clay at different WF contents. X-ray diffraction (XRD), scanning electron microscope (SEM), and Fourier transform Raman spectrum (FT-Raman) analysis were also performed for further explanation.

MATERIALS AND METHODS

Materials

PLA (AI-1001) (D content) was obtained from Esun, Shenzhen BrightChina Industry (China). It had a density of about 1.25 g/cm^3 and a melt flow index about 10–20 g/10 min at 190°C. The Mn of PLA was about 80,000. The wood flour (WF) of poplar (*Populus tomentasa* Carr.) with a size passed 100-mesh sieve was supplied by Xingda Wood Flour Company, China. The clays used in this study were natural Na-montmorillonite (Na-MMT), and organic-montmorillonite (OMMT) by modifying MMT with a quaternary ammonium salt, which were both purchased from Zhejiang Hongfeng Clay Chemical (China). The mean interlayer distances of Na-MMT and OMMT are 14.59Å and 22.17Å, respectively. The particle sizes of the two types of clays were both 76 μm. Polyfluotetraethylene (PTFE) membranes were used in this study as demolding materials to avoid sticking board during hot-pressing.

Preparation of Samples

Before use, the WF was dried in an oven at 105°C for 5 h to 1–2% moisture content. The size of PLA/WF/clay composites with target density of 1.28 g/cm^3 was 270 mm × 270 mm × 3 mm. Then, PLA, WF, and 2 phc (per hundred compounds) of clays were weighed and mixed according to formulations given in Table I in a high speed blender at about 2900 rpm for 4 min. The mixture was then dried at 105°C for 2 h and taken out for hand matting. A hot press (SYSMEN-II, made by Chinese Academy of Forestry) was used to produce the composites by compressing the mat at 180°C with a pressure of 4 MPa for 6 min. Before demolding, the formed mat was cooled down at 4 MPa for another 6 min at room temperature. Same method was used to prepare the pure PLA, PLA/clay, and PLA/WF controls. After that, all the mats were cut into required dimensions for further tests.

Determination of Physical and Mechanical Properties

The 24 h water uptake and thickness swelling tests were carried out according to the procedure described in the Chinese standard GB/T 24137-2009 (2009). Samples with dimensions of 50 mm × 50 mm × 3 mm (thickness) were completely immersed into water at 20°C ± 2°C for 24 h, and then the water absorption and thickness swelling tests were calculated based on the weight and mid-span thickness changes.

The flexural tests were carried out according to the Chinese standard GB/T 9341-2000 (2000), which involves a three-point bending test at a crosshead speed of 1mm/min. The size of the samples was 60 mm × 25 mm × 3 mm (thickness).

Characterization by XRD, SEM, and FT-Raman

The X-ray diffraction (XRD) analysis was carried out on an X-ray 6000 (Shimadzu, Japan) to measure the crystallinity of the composites and the controls. The samples were first grounded into powder to pass an 80-mesh sieve and air-dried before analysis.

Table I. Compositions and Labeling of the Studied Formulations

			Clay loadings (phc)	
Labels	PLA (wt%)	WF (wt%)	Na-MMT	OMMT
C	100	0	0	0
M	100	0	2	0
O	100	0	0	2
2-C	80	20	0	0
2-M	80	20	2	0
2-O	80	20	0	2
4-C	60	40	0	0
4-M	60	40	2	0
4-O	60	40	0	2
6-C	40	60	0	0
6-M	40	60	2	0
6-O	40	60	0	2

phc, per hundred compound.

The X-ray beam was Cu-Ka ($\lambda = 0.1518$ nm) radiation, operated at 40 kV and 30 mA. The scanning rate was 0.09°/s and 2θ ranged from 2° to 30° with the rotation speed of 30 rpm.

The powder of the two types of clays and the internal surfaces of samples were examined by using a Philips (Hitachi S-3400, Japan) scanning electron microscope (SEM) with an acceleration voltage of 10 kV. The samples were sputter-coated with gold before observation.

Fourier transform Raman spectrum (FT-Raman) analysis was carried out on an inVia-Reflex (Renishaw, Britain) spectrometer. A polarized Nd: YAG laser source with the wavelength of 785 nm was used. For each spectrum, five scans were accumulated for Fourier transformation to give a resolution of 2 cm^{-1}. All spectra were displayed in wavelengths raging from 200 to 2000 cm^{-1}.

RESULTS AND DISCUSSION

Water Uptake and Thickness Swelling

The results of 24 h water uptake and thickness swelling rates of the samples were shown in Figure 1. It is obvious from Figure 1(a) that the 24 h water uptake of the PLA/WF composites increased greatly with the increasing WF content. Compared with 0.21% for pure PLA, the 24 h water uptake increased to 16.83% while adding 60 wt% content of WF. This is reasonable since the natural WF is porous and hydrophilic. The effects of two types of clay on 24 h water uptake were different. Na-MMT showed a negative effect on the water absorption, namely, the 24 h water uptake further increased after the addition of Na-MMT. It is because that Na-MMT is also a kind of hydrophilic materials. Compared with pure PLA/WF composites at WF content of 60 wt% (16.83%), the 24 h water uptake of Na-MMT modified composites was 18.41%, which was about 2% higher. While for OMMT, it showed little effect on water absorption at lower WF content (≤40 wt%), but when the WF content increased to 60 wt%, the water absorption was reduced. It may be caused by the barrier effect of filler, which inhibits the water penetration in the PLA/WF composites. The result is consistent with the previous investigations suggesting that OMMT could reduce water absorption and improve the dimensional stability of wood-based composites.[28–30] They assumed that the impermeable nanolayers of OMMT introduced into the composite would generate a tortuous fluid-flow pathway, which contributed to the reduction of water uptake. According to their explanation, OMMT could be more easily combined with WF rather than PLA matrix at higher WF contents. This might explain the obvious reduction in water absorption at higher wood contents.

The thickness swelling rates of unmodified and modified PLA/WF composites were shown in Figure 1(b). At lower WF contents (≤40 wt%), Na-MMT showed a negative effect by further increasing the thickness swelling of the composite; however, while the WF content increased to 60 wt%, the thickness swelling rates of Na-MMT modified composites became lower than unmodified controls. The reason may be related with the poor interfacial adhesion between WF and PLA matrix at high WF contents such like 60 wt%, which leads to some microvoids or cracks in PLA matrix with WF surrounded.[31] In that case, some Na-MMT might fill in the voids or cracks and would not be responded to the thickness changes. There is another possibility that the Na-MMT could attach to WF, which may also restrict the hydroexpansion of WF. As regard to OMMT, there was no significant difference for PLA/WF composites with or without OMMT when the WF contents were low (≤20 wt%). while at

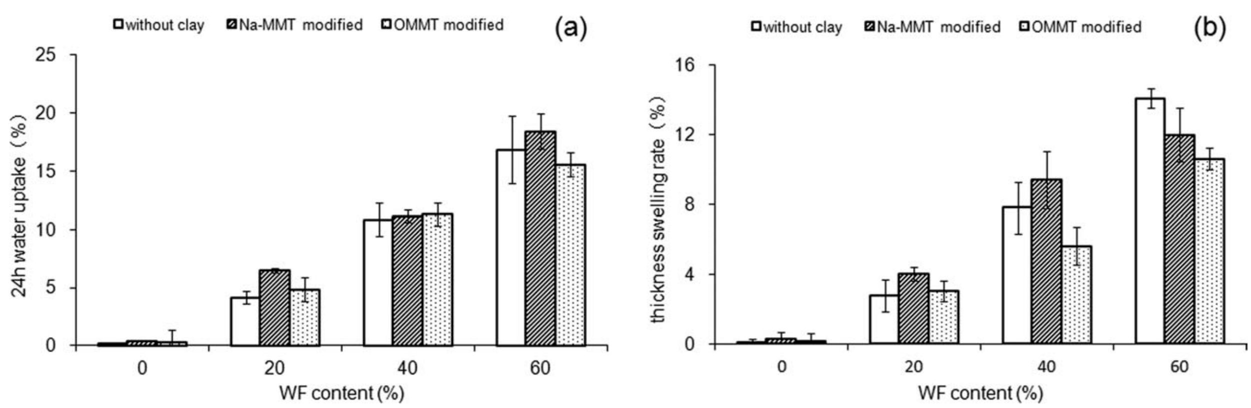

Figure 1. 24 h water uptake (a) and thickness swelling rates (b) of PLA /WF composites.

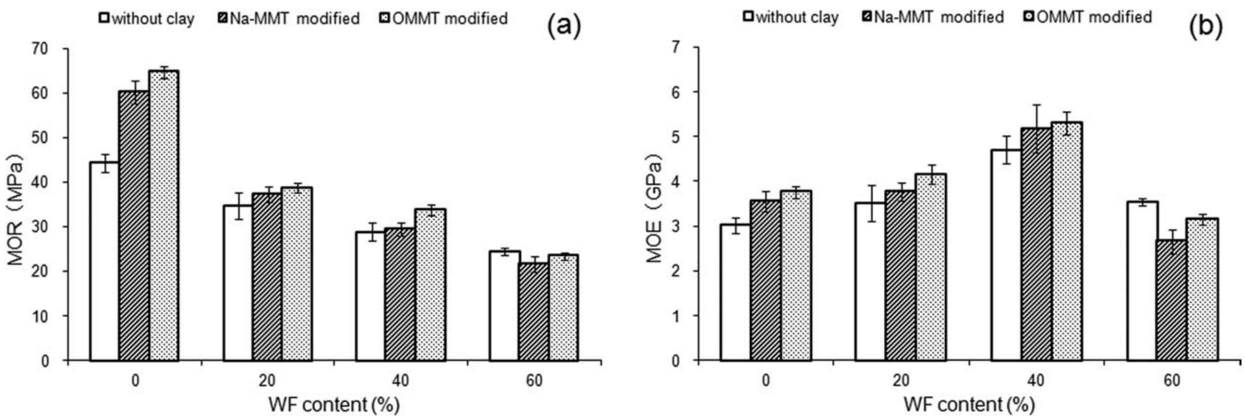

Figure 2. Flexural MOR (a) and MOE (b) of PLA /WF composites.

higher WF contents such as 40 and 60 wt%, the thickness swelling rates decreased obviously after the addition of OMMT due to its hydrophobic property. Besides, compared with Na-MMT, OMMT has a wider interlayer distance, which makes it easier to obtain exfoliated structure and finally some of OMMT layers can enter into the WF cell wall and prevent it from swelling. As a result, at 60 wt% WF content, the thickness swelling rate of OMMT modified composite was reduced to 10.61% from 14.10% for unmodified PLA/WF control, which corresponded a 24.7% reduction.

Flexural Strength Properties

The flexural modulus of rupture (MOR) and modulus of elasticity (MOE) were shown in Figure 2. From Figure 2(a), the flexural MOR decreased with the increasing WF content. The addition of 60 wt% WF decreased the flexural strength from 44.40 MPa (pure PLA) to 24.51 MPa. This was considered to be caused by the reduced interfacial adhesion in PLA/WF composites, which was in consistent with the research results obtained for other WF/polymer composite.[31–34] The addition of clay enhanced the flexural MOR at lower WF content ($\leq$40 wt%). For example, the flexural MOR of pure PLA is 44.40 MPa, while after adding OMMT it rises to 64.77 MPa, which is almost 1.5 times of the value for pure PLA. Sinha et al[35] reported some enhancements of mechanical properties for PLA/clay composites. When the clay is well dispersed in the polymer matrix, it would lead to the strong interaction between PLA matrix and clay and increase the level of intercalation.[36] The same reason can be used to explain the better performance of OMMT than Na-MMT. However, when the content of WF increased to 60 wt%, the flexural MOR dropped with the addition of clay. This may be due to the formation of weak interface caused by the aggregation between excessive WF and clay.

The flexural MOE as shown in Figure 2(b) increased with the increasing WF content at first and then dropped after reaching the maximum at 40 wt% WF content. The trends are the same for unmodified, Na-MMT modified, and OMMT modified PLA/WF composites. The increase of flexural MOE was associated with the increasing stiffness and brittleness caused by WF and clay for their high modulus. Also, the restriction of the PLA molecules by WF and clay prevented the small PLA chains from freely moving.[31] The highest MOE of 5.30 GPa was found for OMMT modified composite at 40 wt% wood content, which was nearly doubled compared with 3.02 GPa for pure PLA. However, as the WF content increased to 60 wt%, the MOE value sharply decreased due to the poor interfacial adhesion of WF and PLA matrix at high WF content. The effect of Na-MMT and OMMT on flexural modulus was almost the same as flexural strength, which had already been discussed.

XRD Analysis

The XRD scans for the two types of clay, PLA and PLA/WF composites at WF content of 60 wt% modified with clays were shown in Figure 3. For better investigation of the structure of the clays, 2θ from 2° to 10° was chosen for discussion. The interlayer distance of clays was deduced from XRD peaks according to eq. (1)

$$d = \frac{n\lambda}{2\sin\theta} \tag{1}$$

where d refers to interlayer distance; n refers to the integer wavelength number ($n = 1$); λ refers to the X-ray wavelength; θ refers to the maximum diffraction angle.

The characteristic peaks of Na-MMT and OMMT were found at $2\theta = 6.05°$ and $2\theta = 3.98°$, which corresponded to the d-spacings of 14.59Å and 22.17Å, respectively, suggesting a wider interlayer distance of OMMT. After being added into PLA or PLA/WF composites, the diffraction peak of clays moved. For PLA modified with Na-MMT (M), the peak moved to $2\theta = 5.42°$, corresponding to slightly wider interlayer distance of 16.28Å, which indicated that some PLA chains had intercalated into Na-MMT. While for PLA modified with OMMT (O), the sample exhibit a peak at $2\theta = 2.27°$ and the corresponding distance between silicate layers was 38.87Å. It is possibly due to the fact that OMMT had a better dispersion than Na-MMT and the silicate layers were further enlarged in PLA matrix. However, after introducing WF into PLA/clay system, the results varied. For PLA/WF modified with Na-MMT (6-M), the peak appeared at $2\theta = 6.05°$, which is the same as Na-MMT itself. While for PLA/WF modified with OMMT, a small broaden peak appeared around $2\theta = 2.81°$ with interlayer distance of 31.40Å. The

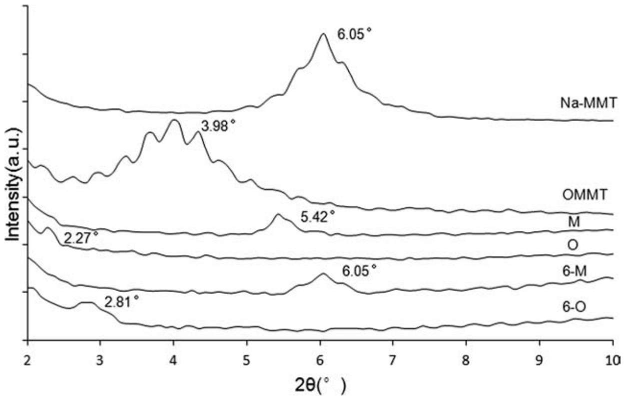

Figure 3. XRD patterns of clays, PLA modified with clays and PLA /WF composites modified with clays. Note: the labels of samples are referred to Table 1.

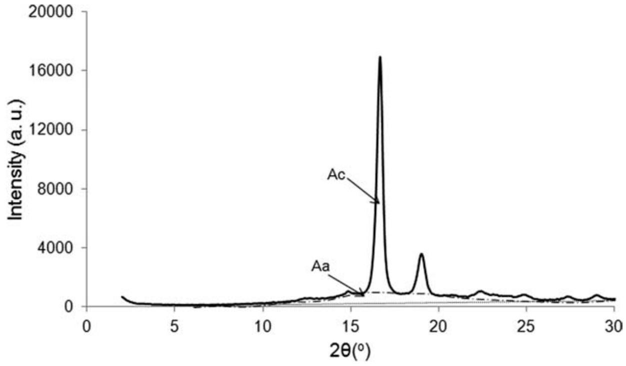

Figure 4. Calculation of the relative crystallinity of PLA or PLA /WF composites.

negative effect caused by WF was considered to be related with the inclination of clays attached on WF surface rather than dispersed into PLA matrix at a higher WF content (60 wt%). However, it was worth mentioning that OMMT performed better than Na-MMT since the interlayer distance (31.40Å) of OMMT modified PLA/WF composites at WF content of 60 wt% was greatly larger than OMMT itself, while Na-MMT was not obvious.

The relative crystallinity of samples calculated from XRD data was listed in Table II. The degree of crystallinity of samples was quantitatively determined by integration of 1-d diffraction pattern intensity that measures the volume fraction crystallinity and calculated according to eq. (2).[37]

$$X = \frac{Ac}{Ac + Aa} \times 100\% \quad (2)$$

where X refers to relative crystallinity; Ac refers to the crystallized area of the sample on the X-ray diffractogram; Aa refers to the amorphous area of the sample on the X-ray diffractogram. An example of calculating crystallinity from the X-ray diffractogram is shown in Figure 4.

It can be seen that, with the increasing WF content, the crystallinity showed a slight decrease. This could be explained by the lower crystallinity of WF (about 37%) than pure PLA (43.38%). In other words, compared with WF, the crystallinities of the composites were increasing since the crystallized structure of PLA penetrated into wood cell wall. The addition of clay slightly decreased the crystallinity of PLA/WF composite at lower WF content (≤40 wt%) due to its weak crystallinity, while increased at WF content of 60 wt%. This might because more clay attaching to WF rather than diffusing into PLA matrix. OMMT resulted in a higher crystallinity than Na-MMT, which can explain the better performance of OMMT in physical and mechanical properties of PLA/WF composites.

Table II. Crystallinity of PLA/WF Composites

	Crystallnity (%)		
		2 phc clay	
WF content (wt%)	Without clay	Na-MMT	OMMT
0	43.38	42.04	43.29
20	42.28	41.08	41.65
40	39.62	39.23	39.26
60	37.20	38.05	39.45

phc, per hundred compound.

SEM Analysis

Figure 5 showed the SEM images of the micro-structure of the two types of clay, pure PLA, PLA modified with clay and PLA/WF composites modified with clay at WF content of 40 and 60 wt%, respectively. Comparing Figure 5(a) with 5(b), we can clearly see that the Na-MMT mainly accumulated into large aggregation with irregular surface structure, while OMMT was characterized by much looser structure with some even exfoliated into separate layers. Figure 5(c–e) showed the section images of pure PLA, PLA modified with Na-MMT, and OMMT, respectively. It was obvious that Na-MMT had very poor compatibility with PLA matrix by showing tremendous microvoids. However, OMMT modified PLA showed very homogenous texture, suggesting that OMMT had very good compatibility with PLA.

Figure 5(f–k) showed the section images of PLA/WF control, PLA/WF modified with Na-MMT, and OMMT at WF contents of 40 and 60 wt%, respectively. Clay especially OMMT can improve the homogeneity of WF distribution with PLA but the difference between PLA/WF control and PLA/WF modified with clay is not very obvious at WF content of 40 wt% [Figure 5(f–h)], suggesting that PLA and WF could be well combined at lower WF contents below 40 wt%. While after introducing more WF into clay/PLA system, namely, at WF content of 60 wt% [Figure 5(i–k)], the effect of Na-MMT and OMMT became significant. The SEM image of PLA/WF without clay [Figure 5(i)] exhibited some big voids surrounding the WF, separating WF from PLA matrix. In Na-MMT modified PLA/WF composite [Figure 5(j)], the size of voids became smaller but more voids appeared than in control PLA/WF composite without clay. That explains the higher 24 h water uptake of Na-MMT modified PLA/WF than PLA/WF control in Figure 1(a).

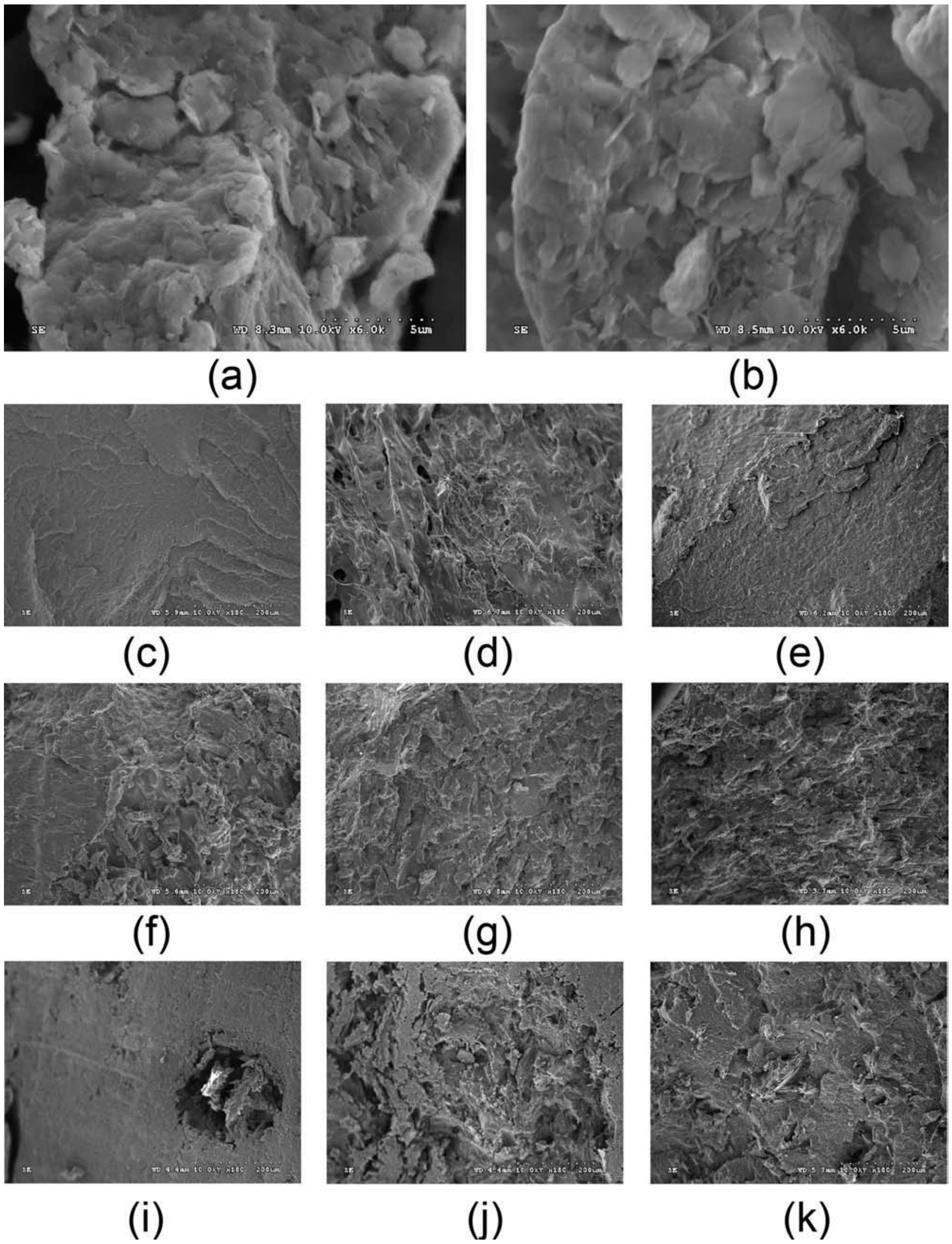

Figure 5. SEM images of Na-MMT (a), OMMT (b), pure PLA (c), Na-MMT modified PLA (d), OMMT modified PLA (e) PLA/WF control without clay at 40 wt% WF (f) and 60 wt% WF (i), Na-MMT modified PLA/WF composite at 40 wt% WF (g) and 60 wt% WF (j), and OMMT modified PLA/WF composite at 40 wt% WF (h) and 60 wt% WF (k). Note: the scale bars of SEM images were × 6000 for (a) and (b), × 180 for (c–k).

However, the voids in OMMT modified PLA/WF composite [Figure 5(k)] became even smaller and most part of WF and PLA matrix were evenly distributed, suggesting that OMMT improve the interfacial adhesion of WF and PLA matrix so that the water uptake and thickness swelling lowered.

FT-Raman Analysis

The FT-Raman results of the unmodified and clay modified PLA, as well as the PLA/WF composites with or without clay at WF contents of 60 wt% were showed in Figure 6(a,b), respectively.

In Figure 6(a), it can clearly seen that the peaks of Na-MMT modified PLA (M) was almost the same with pure PLA (C), suggesting the Na-MMT simply dispersed in PLA matrix rather than reacted with it. While PLA modified with OMMT (O) showed some differences. A strong peak appeared at 483 cm^{-1}, which should be assigned to Si–O–C bond and could be produced from reaction between PLA and OMMT. Peaks around 1450 cm^{-1} were assigned to CH_3 in-plane asymmetric bending, a characteristic peak for PLA. However, when it was modified by OMMT, the peak was broken into a shoulder and shifted to 1463 cm^{-1}. This might caused by the overlapping of Si–CH_3

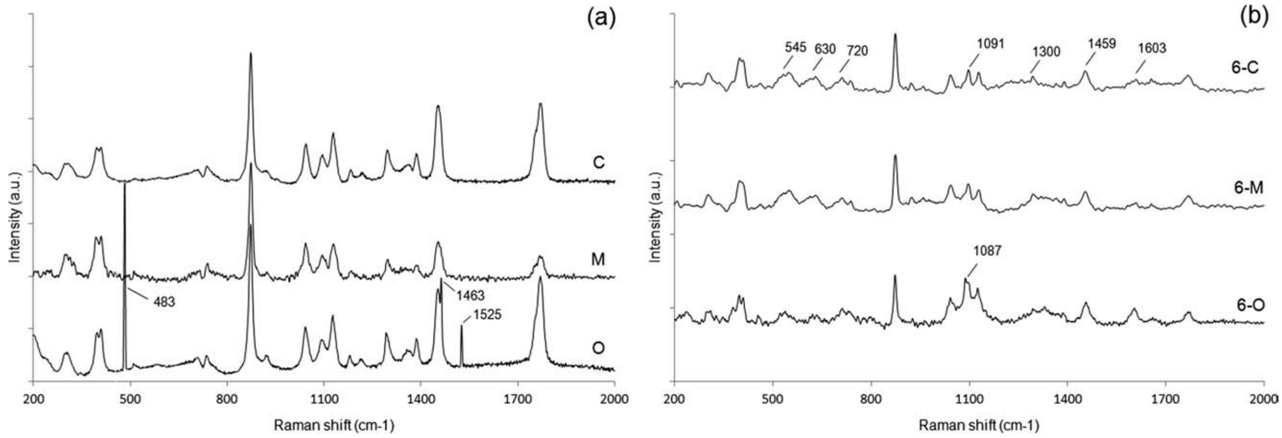

Figure 6. FT-Raman spectram of unmodified and clays modified PLA (a), and PLA /WF composites with or without clay at WF content of 60 wt % (b), respectively. Note: the labels of samples are referred to Table 1.

bond. Another peak at 1525 cm^{-1} was contributed to the modifier of the quaternary ammonium salt in OMMT. These indicated that the OMMT not only dispersed in PLA matrix but also intercalated and reacted with it.

Figure 6(b) gave the FT-Raman results of PLA/WF composites at WF content of 60 wt%. Compared with Figure 6(a), extra peaks at 1459 cm^{-1} was assigned to symmetric angle vibration of CH_2 for cellulose and hemicelluloses and 1603 cm^{-1} was assigned to symmetric aryl ring stretching of C=C of lignin. The small broad peaks around 1300 cm^{-1} were assigned to the overlapping of benzene and phenol derivatives in lignin. The peak at 1090 cm^{-1} assigned to symmetric stretching of C–O–C of PLA was stronger, which was intensified by acetyl groups from cellulose or hemicelluloses. New peaks in the low frequency Raman spectra below 800 cm^{-1}, like 720 cm^{-1}, 630 cm^{-1}, and 545 cm^{-1} were contributed to extractives in WF. All the above information proved the addition of WF into PLA matrix. However, there was no big difference between unmodified PLA/WF control and PLA/WF composites modified with clays. For OMMT modified composite, only a peak was broken and shifted to 1087 cm^{-1} from 1090 cm^{-1}. This was caused by the Si–O–Si group, suggesting the addition of clay. The peak at 483 cm^{-1} for Si–O–C bond and 1463 cm^{-1} for Si–C bond in O group both vanished after adding WF, suggesting that much OMMT might be attaching on WF surface rather than dispersing into PLA matrix at higher WF contents (60 wt%).

CONCLUSIONS

Both Na-MMT and OMMT used in this study had an influence on the physical and mechanical properties of PLA/WF composites, and the effect was highly dependent on WF content. OMMT showed better performance than Na-MMT by improving most of the tested properties including the resistance to water sorption and swelling, flexural MOR and MOE, etc. While for Na-MMT modified composites, although the flexural strength properties were mostly improved, the water absorption and thickness swelling were weakened in most cases. This might owe to the better distribution and exfoliated structure of OMMT in PLA/WF system. WF content was also an important factor to determine the properties. The properties of water absorption, thickness swelling and MOR were greatly weakened with increasing WF loading, while the change on MOE was adverse below WF content of 40 wt%. As for the effect of different types of clay on WF content, 40 wt% WF served as a critical point above which the clay would be more attached on the surface of WF rather than diffused in PLA matrix and finally resulting in poorer interfacial adhesion at high WF contents.

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