

Effect of Mesh Number of Wood Powder and Ratio of Raw Materials on Properties of Composite Material of Starch/Wood Powder

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Utilizing wood powder and corn starch as the main materials with polyurethane adhesive as a cross-linking agent, the starch/wood powder composite material was prepared *via* molding forming techniques. The effects of wood powder mesh and addition of wood powder on the properties of composite material were investigated. The compatibility of starch and wood powder and the thermal stability of composite were evaluated by scanning electron microscopy (SEM), thermogravimetric analysis (TGA), and dynamic mechanical thermal analysis (DMA), respectively. The mechanical properties and water absorption of composite material were also tested. The results showed that the mechanical properties and water resistance of composite material first increased and then fell with increasing mesh, and the best performance was obtained when the mesh ranged from 80 to 100. The best compatibility, mechanical properties, water resistance, and thermal stability of composite material was obtained with 10% polyurethane crosslinking agent addition.

Keywords: Wood; Corn starch; Flour; Polyurethane polyurethane crosslinking; Molding

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INTRODUCTION

The study and development of bioplastics is a very attractive area because of the increasing costs of petroleum alternatives and environmental pollution caused by synthetic plastics (Gironès *et al.* 2012; Kaewtatip and Thongmee 2012; Jiang *et al.* 2015). Wood plastic composite is a new material that has generated increasing interest due to its excellent combination of properties (Wechsler and Hiziroglu 2007; Kirchoff *et al.* 2012). The plastics used in traditional wood plastic composites are generally polyethylene (PE), polypropylene (PP), and polyvinyl chloride (PVC), along with other petrochemical products. But this kind of wood plastic composite cannot degrade, even after several times repeated use, and can induce environmental pollution. (Castano *et al.* 2012). With the increasingly serious deterioration of the environment, as well as on-going energy shortage problems, the development of new composites using renewable natural resource materials has become important. To this trend, many studies have focused on the preparation and study of biodegradable polymers-based composites and nanocomposites (Yang *et al.* 2005).

Starch is the second most abundant renewable polymer found in nature after the lignocellulose group and it is relatively inexpensive (Gironès *et al.* 2013). Because of its

low cost, renewability, biodegradability, and lack of toxicity or smell, it has been considered for many years to have high potential for use in packing and disposable items instead of petroleum-based polymers (Kaewtatip and Thongmee 2013; Zuo *et al.* 2014). Unfortunately, the hydrophilicity and poor mechanical properties are often limiting factors for the use of thermoplastic starch in many applications. Improved water resistance has been achieved by rendering starch more hydrophobic through chemical modifications (Kosan *et al.* 2006; Simi and Abraham 2007; Duanmu *et al.* 2010). The further development and utilization of starch resources has become a topic of common concern in the materials research field (Ma *et al.* 2005). Different approaches can be used to design thermoplastic starch materials with enhanced structural and functional stability during use, which could broaden their application range (Moriani *et al.* 2011). When starch is employed as the matrix and wood fibers serve as the reinforcement, the resulting material is considered a bio-composite or “green” composite because it is completely biodegradable (Guadalupe *et al.* 2014)

Polyurethane (PU) is a commonly used component in manufactured materials (Murayama and McMillin 1983; Decay and Charles 2012). Although the dosage of a polyurethane cross-linker accounts for only approximately 5% of wood adhesives, its application amount has been quite considerable. This trend has greatly promoted the fast growth of the polyurethane industry in recent years (Dakai and Charles 2012; Phetphaisit *et al.* 2013). Multi-isocyanate monomers can be directly used as adhesives and can also pre-polymerize with polyester polyols into polyurethane, of which the polyurethane cross-linker is the most extensively applied (McDonnell *et al.* 1951). At present, there are many methods of preparation for wood plastic composites. The main molding methods of wood plastic composites in China include the impregnation method, the mold pressing method, the extrusion molding method, and the injection method.

In the present investigation, a mold pressing method was employed to produce a starch-wood powder composite. In this study, the effects of the different mesh numbers of the wood powder, as well as the ratio of the wood powder-starch composites, on the properties of the composites were explored. The research results can provide a theoretical basis for the preparation of natural pollution-free, biodegradable composites.

EXPERIMENTAL

Materials

Industrial grade corn starch was obtained from Dacheng Corn Development Co., Ltd. (Changchun, Jilin, China); the size of the starch flour is 80 to 150 mesh. It was loaded in a vacuum drying oven at 50 °C for 24 h to eliminate moisture before use. It was prepared with use of different crushing and screening mesh periods of cypress wood powder fiber, according to the need for screening of 20 to 40 mesh, 40 to 60 mesh, 60 to 80 mesh, 80 to 100 mesh, and 100 to 120 mesh. The source material was purchased from Baiquan Wood plastic composite material base (Heilongjiang, China). Polyethylene glycol (M_w 400 Daltons) and acetone, analytical grade, was bought from Tianjin Guangfu Chemical Reagent Co., Ltd. (Tianjin, China). Polymethylene polyphenyl isocyanate (PAPI) pre-polymer was obtained according to the methods of Zhang *et al.* (2012).

Methods

Preparation of starch/wood powder composite materials with different meshes

Starch (40 g) was added to 60 g of 20 to 40 mesh, 40 to 60 mesh, 60 to 80 mesh, 80 to 100 mesh, and 100 to 120 mesh wood powder fibers, respectively. Polyurethane was then added to the mixture. The proportion of the polyurethane cross-linker in the composites was 10% of the total content. After mixing, the mixture was sealed and incubated for 10 min to obtain a uniform composite. The size of the composite manufactured is 200×200 mm square. The parameters of the composite material used in the molding method were as follows: pressure, 7 MPa; temperature, 110 °C, and time, 1.0 min/mm.

Preparation of starch/wood powder composite materials with different ratio of starch and wood powder

The ratio of the wood powder to starch was 10:0, 9:1, 7:3, 5:5, 3:7, and 1:9. Starch (40 g) was added to different wood powder ratio with mixing. The proportion of the polyurethane cross-linker in the composites was 10% of the total content. After mixing, the mixture was sealed and incubated for 10 min to obtain a uniform composite. The size of the composite manufactured was 200×200 mm square. The parameters of the composite material used in the molding method were as follows: pressure, 7 MPa; temperature, 110 °C, and time, 1.0 min/mm.

Scanning electron microscope (SEM)

The starch/wood composites were characterized with a scanning electron microscope (SEM) (QUANTA 200; Phillips, Netherlands). The SEM was operated at an acceleration voltage of 20 kV. The samples were cooled in liquid nitrogen and then broken into pieces. The fractured faces were sputter-coated with gold for SEM observation.

Mechanical properties

The tensile and flexural properties of composite materials were tested according the GB/T 13022-1991 (General Administration of Quality Supervision 1991) and GB/T17657-1999 (General Administration of Quality Supervision 1999), respectively. The kind of bending method was three point's bending, and the distance between supports in case of the bending specimen was 80 mm. The cross-section dimensions of the bending specimens were 30 mm×30 mm, respectively. The experiments were determined in a CMT-5504 Materials Testing Machine (Sans, Shenzhen, China) at a crosshead speed of 2 mm/min. The data were averages of five specimens.

Water absorption

According to the National Standard of China GB/T17657-1999 (General Administration of Quality Supervision 1999), the bars of samples were cut into small pieces of 10 by 10 mm and the pieces were vacuum-dried at 50 °C for 48 h. The dried pieces were weighed immediately, and then they were soaked in distilled water at room temperature. After 2 h and 24 h, the specimens were removed from distilled water, blotted dry with filter paper, and then weighed again. Data were recorded as averages of the three specimens. Water absorption of the starch plastic was calculated as follows (Eq. 1),

$$\text{Water absorption} = \frac{W_2 - W_1}{W_1} \times 100\% \quad (1)$$

where W_2 is the mass of the sample after water absorption and W_1 is the mass of the dry sample.

Thermogravimetric analysis (TGA)

Thermogravimetric analysis was carried out to determine the thermal stability of the composites. The pan was heated from room temperature to 800 °C at a rate of 10 °C/min under an argon atmosphere (flow 40 mL/min). The sample weight was plotted as a function of temperature for all samples, and its first derivative was used for the analysis.

Dynamic mechanical analysis (DMA)

Dynamic mechanical analysis (DMA) was performed with a D204 DMA by NETZSCH instruments (Germany). Tests were run at 5 Hz with a strain of 0.1% and the temperature was ramped from 25 °C to 300 °C at a rate of 5 °C/min.

RESULTS AND DISCUSSION

Compatibility Analysis

The composites were prepared by the crosslinking of polyurethane with the wood powder and starch. As a consequence of such crosslinking, the compatibility of the wood powder and starch was affected to a certain degree. The analysis results obtained by SEM are as shown in Fig. 1 and 2.

As illustrated in Fig. 1, if the mesh number of the wood powder fiber was smaller, then wood powder fiber was longer; and if the mesh number was larger, then the size of the wood powder fiber was smaller. When the mesh number of the wood powder was 20 to 40 mesh, the starch in the composites exhibited a granular distribution. They also played enhancing and filling roles in the skeletal structure of the wood powder (wood fiber). However, because fibers of the wood powder of 20 to 40 mesh were long, their compatibility with the starch was poor. Cracks and the enrichment of starch particles were evident on the surface of wood fiber. It could not be distributed into the wood fiber very well or evenly, thereby causing enhanced local brittleness, poor reinforcement, and poor filling properties. Therefore, the mechanical strength of the wood powder composites prepared by the 20 to 40 mesh wood powder was relatively lower than that of the composites prepared by the other mesh numbers. With the increase of the mesh number of the wood powder, the particle size of the wood powder fibers decreased, and also the gap between the wood powder and starch interface gradually decreased. Therefore, its compatibility with wood powder mesh was increased, from which the interface of the two phases of the fiber and starch can be clearly seen. The particles were closely linked to the fiber, and the composite between the starch particles and the wood powder was easily conducted, and therefore its mechanical properties were greatly improved. Also, there was a high compatibility of the composites prepared by selecting the 80 to 100 mesh, as well as the 100 to 120 mesh of the wood powder. Interfacial cracks and holes almost disappeared, enrichment of starch granules was only partial, and the granules were closely integrated with the wood powder fibers.

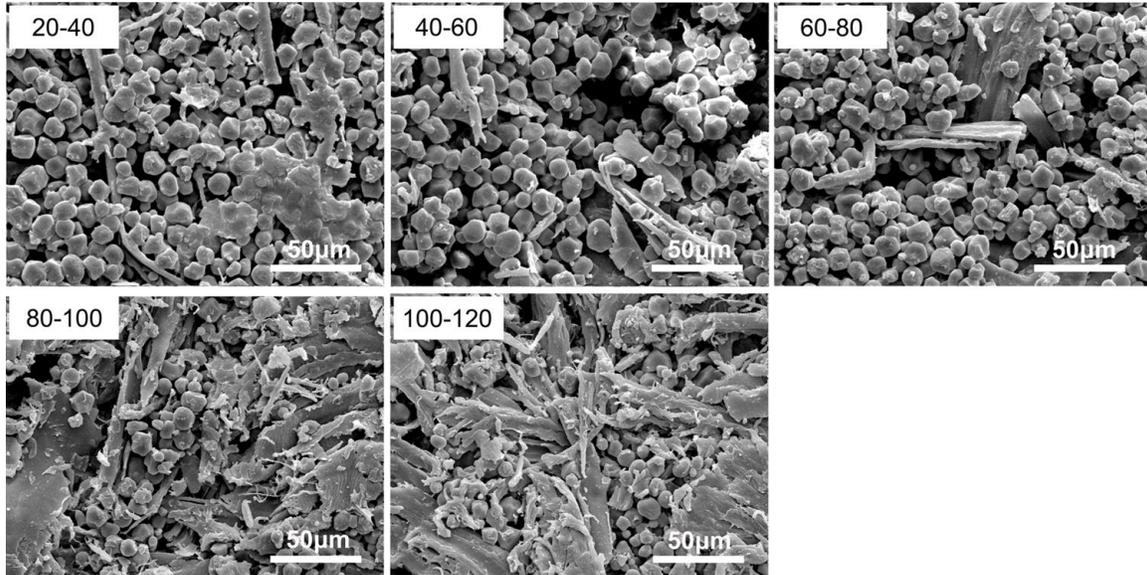


Fig. 1. SEM images of the composites prepared by different wood powder mesh

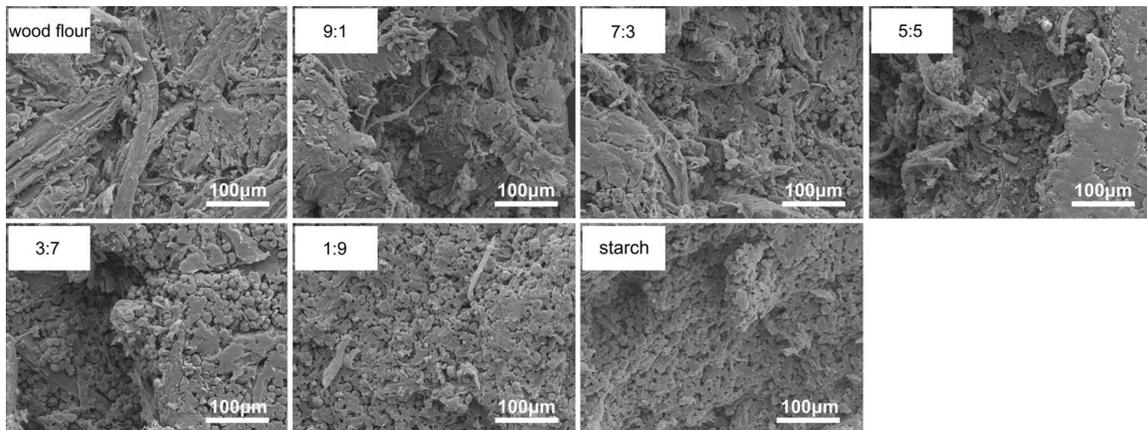


Fig. 2. SEM images of the composites prepared by different addition of the wood powder

Figure 2 illustrates the effects of the ratio of the wood powder and starch on the interfacial compatibility. As shown in Fig. 2, with the increase in content of starch, an increasing number of starch particles were distributed on the surface of the wood fibers. However, the starch particles, which truly played enhancement roles, were minimal. On the contrary, with the increase of the starch particles, the plate strength was reduced and the brittleness increased. Therefore, using a ratio of the wood powder to starch of 9 to 1 was found to be an effective compatibility ratio.

Mechanical Property Analysis

The mechanical properties of the composites were closely related to the morphological structure and the combination mode of starch in wood powder, while the dispersion state was also related to the compatibility (Liu *et al.* 2005). Figures 3 and 4 illustrate the effects of the different mesh numbers of the wood powder, as well as the different wood powder/starch composite proportions, on the mechanical properties of the prepared composites.

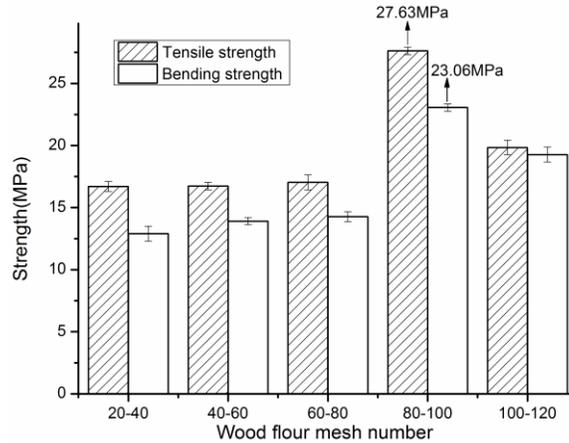


Fig. 3. Mechanical properties curves of the composites prepared by different wood flour mesh

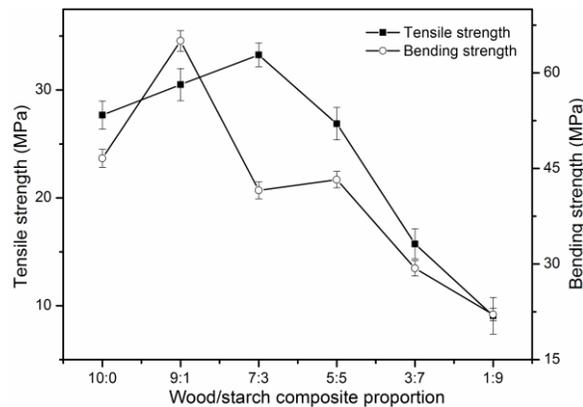


Fig. 4. Mechanical properties of the composites prepared by different addition of the wood powder

As shown in Fig. 3, with the increase of the mesh number of the wood powder, the tensile strength first increased from 16 MPa to 27 MPa, and then it decreased. When the mesh number of the wood powder was 80 to 100 mesh, the mechanical properties were greatly enhanced, and the tensile and bending strengths reached the maximum values of 27.63 MPa and 23.06 MPa, respectively. The properties of the wood powder with 100 to 120 mesh were slightly lower than those of 80 to 100 mesh wood powder, and the tensile and bending strengths were 18.09 MPa and 19.27 MPa, respectively. The mechanical properties of the composites prepared with the wood powder mesh numbers of 20 to 40 mesh, 40 to 60 mesh, and 60 to 80 mesh were poor. This was mainly due to the large size of the wood fiber, and the starch not reaching gelatinization, causing the homogeneity of the starch and wood powder mixture to have poor results. In addition, the starch could not completely wrap the wood powder; therefore the pores were large, and the mechanical properties were poor. The mixing of the wood powder with the mesh numbers of 80 to 120 mesh with the starch was even, and the addition of the wood powder reduced the brittleness of the original starch, giving it a certain toughness and strength. As shown in Fig. 4, it can be concluded from the comprehensive combination of the tensile and bending strengths that when the wood powder/starch composite ratio was 9 to 1, the bending strength increased to 65.01 MPa, and the tensile strength increased to 30.85 MPa. In combination with the SEM analysis results, it can be seen that when the

mesh size of the wood powder was large and the particle size was small, the compatibility of the wood powder and starch was acceptable. When the ratio of the wood powder/starch composite was 9 to 1, the dispersion compatibility effect of the starch in the wood powder was the most effective, and the filling and reinforcing effects were improved.

Water Resistance Analysis

Both starch and wood powder contain hydrophilic groups. Therefore, the water absorption performances of the composites prepared by the different mesh numbers of the wood powder, and the composites prepared using different ratios of wood powder/starch composites were tested. The results of this test are shown in Figs. 5 and 6.

As displayed in Fig. 5, with the increase of the mesh number of the wood powder, the water absorption rate of the composite exhibited first an increase, and then a decreasing trend in a 24 h test period. However, it showed no effect on the water absorption rate in a 2 h test period. These results can be mainly attributed to the large particle size of the wood powder with a size of 20 to 40 mesh and because the water absorption capacity of the wood fiber was low. For the wood powder with 60 to 80 mesh, the surface roughness was high; its structure was loose; the blending effect of the starch with the wood powder was poor; the polyurethane crosslinking agent did not produce a good cross-linking effect; and there were many more bare particles and holes around the wood powder, causing it to easily absorb water due to its poor water resistance. For the wood powder with 80 to 120 mesh, the length diameter ratio and particle size were small, but the compatibility of the interface of the starch and wood powder was high. Therefore, the probability of reaction and cross-linking of the polyurethane cross-linking agent and the starch and wood powder increased. Also, the water resistance improved when compared to those of the other mesh sizes.

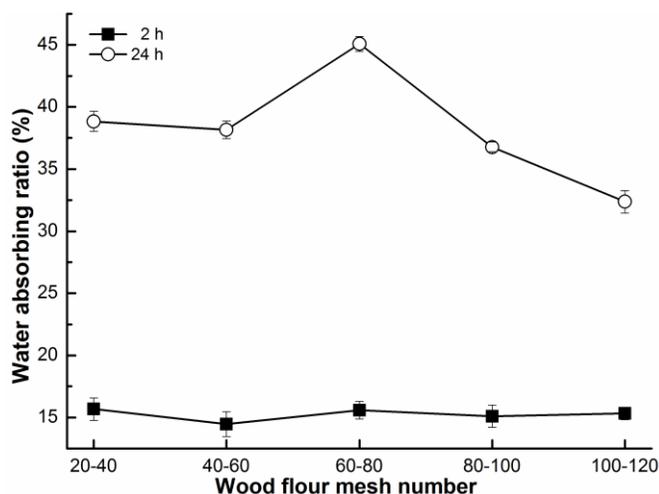


Fig. 5. Water absorbing ratio of the composites prepared by different wood powder mesh

As shown in Fig. 6, when the ratio of the wood powder/starch composite was 9 to 1, the wood powder and starch combined with each other *via* a polyurethane crosslinking agent. Thereby, the content of starch that did not participate in the reaction was small, and the water absorption of the composite was the lowest. When the ratio of the wood powder/starch composite was 10 to 0, and only the wood powder was present, the wood

powder easily absorbed water. With the decrease of the ratio in the wood powder/starch composite, the content of starch was increased, and the water absorbing performance of the starch improved. The starch in the composites did not participate in the reaction, and the starch particles attached on the surface of fiber increased in number, so that the water absorption performance was gradually enhanced.

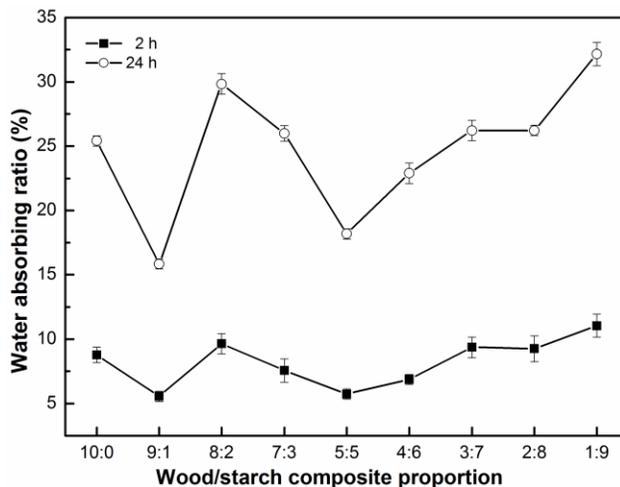


Fig. 6. Water absorbing ratio of the composites prepared by different addition of the wood powder

Thermal Stability Analysis

The thermal properties of the starch/wood powder composite were analyzed by TGA, and the results are illustrated in Figs. 7 and 8.

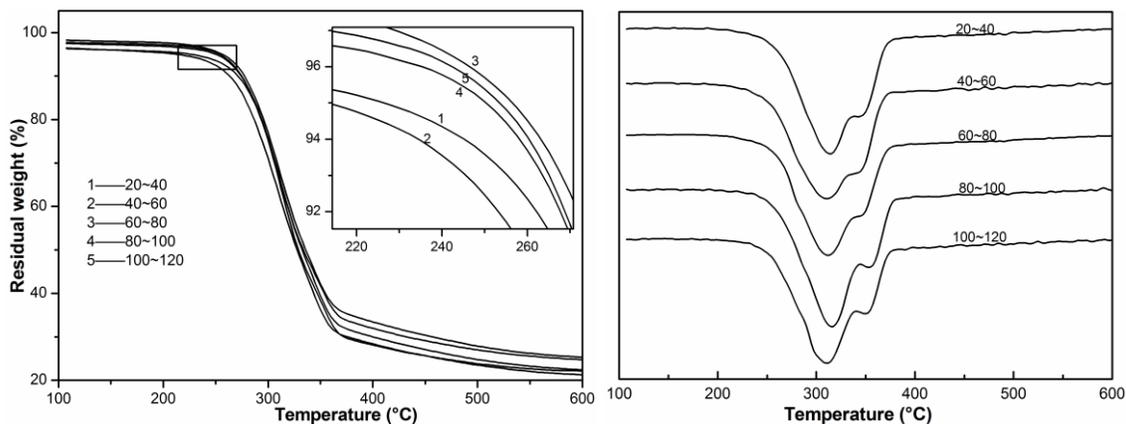


Fig. 7. TGA and DTG curves of the composites prepared by different wood flour mesh

As shown in Fig. 7, the mesh number of the wood powder had a certain effect on the thermal stability of the composite. Within the 200 to 260 °C temperature ranges, the stability of the wood powder with a mesh number of 60 to 80 was found to be the best, followed by that of the 100 to 120 mesh, and then the 80 to 100 mesh. The reason for this was that the interfacial compatibility of the wood powder with 80 to 120 mesh and the starch was high, so its thermal stability was also high. The length of the fibers of the wood powder with 60 to 80 mesh was moderate, and although its compatibility with the

starch interface was not as high as that of the 80 to 120 mesh, it also played a maximum role of the composite material, and therefore its stability was the highest.

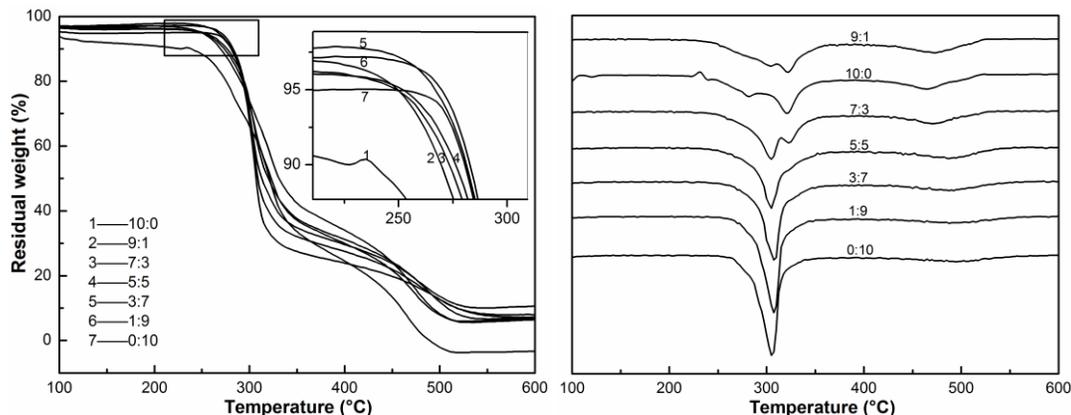


Fig. 8. TGA and DTG curves of the composites prepared by different additions of wood powder

Figure 8 shows that when the ratio of the wood powder/starch composite was 9 to 1, the starting temperature for weight loss of the composite material was the highest, and the thermal stability was the best. With the decrease of the ratio of the wood powder and starch, the starch was gradually increased, the thermal weightlessness starting temperature of composite was reduced, and also the thermal stability was decreased. Due to the fact that the heat stability of the starch was poorer than that of the wood powder, the ratio of the added starch into the wood powder/starch composite should not be too high in the preparation of the starch and wood powder composites. Therefore, in this study, the ratio of 9 to 1 was chosen, and the thermal stability was the most acceptable. In combination with the best compatibility of the wood powder and starch discussed above, the starch completely filled the inside of the wood fiber, and fulfilled an excellent enhancement and filling role.

DMA Analysis

Figures 9 and 10 illustrate the effects of the mesh number of the wood powder on the thermal mechanical properties of the composites. The curves obtained were the storage modulus and the loss angles under different temperatures. As illustrated in Fig. 9, the storage modulus of the composite exhibited a decreasing trend with the increase of temperature within the entire range of temperatures. This is attributed to the fact that when the temperature was high, the material's rigidity was reduced, and the storage modulus was weak. When the temperature was being raised from $-50\text{ }^{\circ}\text{C}$ to $100\text{ }^{\circ}\text{C}$, the storage modulus of the composites gradually decreased. When the temperature continued to rise, the rate of decrease of the storage modulus decreased and was almost zero when the temperature was $250\text{ }^{\circ}\text{C}$. Thus, these results indicated that the effect of temperature on the storage modulus of the composites was relatively large when the temperature range was lower, while at high temperature range, the effect of temperature on the storage modulus decreased. It can be seen from the storage modulus of the five types of mesh numbers of the wood powder (wood fiber morphology) that the storage modulus of the composites exhibited first a decreasing, then increasing, followed again by a decreasing trend, with the increase of the mesh number of the wood (decreases in fiber size). The

storage modulus of the composite with the mesh number of 80 to 100 mesh was the largest, followed by that of the 100 to 120 mesh. These results were consistent with the trends of the static bending and tensile strengths. Also, smaller fiber was easily agglomerated into the matrix, leading to a failure of the stress conduction. Too much fiber easily produces voids, which leads to stress concentration (Zhang *et al.* 2013).

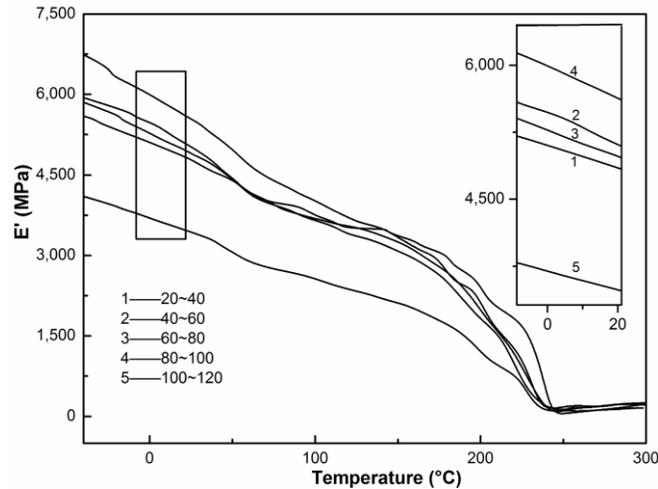


Fig. 9. DMA curves of the composites prepared by different wood powder mesh

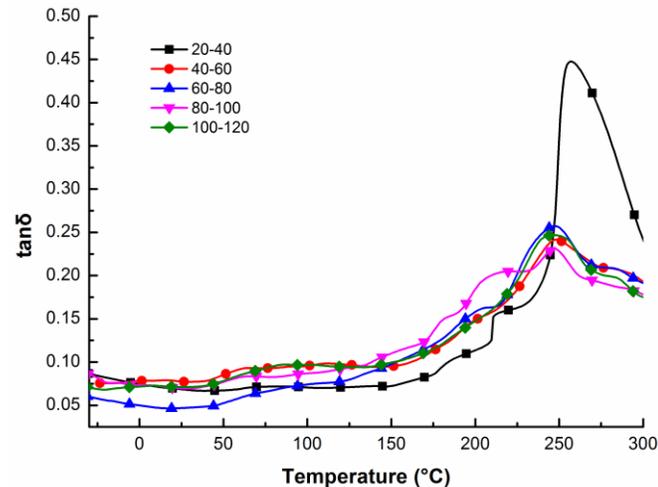


Fig. 10. DMA curves of the composites prepared by different addition of the wood powder

Figure 10 illustrates the curve of the loss angle tangent of the composite with various temperatures. As shown, there was a relaxation transition peak of the composite at 0 °C to 100 °C; it may be the glass transition of starch in the composite. With the increase of the temperature, the loss angle rate of the composite was quick. The composites prepared using the 5 types of wood powder with different mesh (wood fiber morphology) were compared, and the results indicated that the loss tangent of composite (that is the loss modulus) exhibited first an increasing, and then a decreasing trend with the increase of the mesh number of the wood powder. With the increase in the mesh number of the wood powder (wood fiber decreased), the energy required for the melting of the composites showed first an increasing, and then a decreasing trend. Also, the

molecule movement was increasingly difficult at first and then became easier, and the storage modulus was also increased at first and then decreased. Due to the loss tangent rate in temperatures lower than 100 °C being fast, and also because the temperature change was quick, the speed loss was similar, so that the maximum temperature difference of the loss tangent was minimal.

CONCLUSIONS

1. The effects of the mesh number of the wood powder and the ratio of the wood powder and starch composite on the properties of the composites were pronounced, especially in terms of the ratio of the wood powder/starch composite.
2. With the increase in mesh number of the wood powder, the particle size decreased; the compatibility increased gradually; and the water resistance was enhanced.
3. When selecting the 80 to 100 mesh wood powder, the mechanical properties were greatly enhanced, and the tensile and bending strengths reached maximum values of 27.63 MPa and 23.06 MPa, respectively.
4. The effects of the ratio of the wood powder/starch composite on the performance of composite were substantial when the ratio of the wood powder and starch was 9 to 1.
5. The SEM analysis results showed that the compatibility of the starch and wood powder was high: the ratio of the wood powder and starch was 9 to 1.
6. The water content and 24 h water absorption of the composite were found to be low, and the moisture content was close to 0. The 24 h water absorption rate was 15.84%, while the bending performance was improved with the bending strength reaching the maximum value of 65.01 MPa and the tensile strength reaching 30.85 MPa. The best thermal stability was achieved with this ratio 9 to 1.

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