Influence of Maleic Anhydride Grafted Polypropylene on the Interfacial Compatibility of Wood Flour/Poly(β-Hydroxybutyrate Valerate) Composites

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Maleic anhydride grafted polypropylene (MAPP) was utilized as a coupling agent to prepare composites of *Populus tomentosa* wood flour (WF) and poly (β-hydroxybutyrate valerate) (PHBV) through the hot-pressing process. The impacts of this coupling agent on the interfacial compatibility and physical-mechanical properties of WF/PHBV composites (WPHBVs) were analyzed and discussed by making use of scanning electron microscopy (SEM), Fourier transform infrared spectroscopy (FT-IR), and thermogravimetric analysis (TGA). The results indicated that after adding MAPP, a grafting reaction would occur. This reaction improved the interfacial compatibility between *Populus tomentosa* WF, and PHBV boosted the thermal stability of WPHBVs. When the addition amount of MAPP was 2%, the flexural strength and elastic modulus of WPHBVs reached their maximum levels, up to 27.99 MPa and 3690.47 MPa, respectively, and the strength enhancements were all above 40%. At this stage, the cross-section of the WPHBVs exhibited a smooth surface with no visible gaps. Tight interfacial bonding between phases indicated the highest level of compatibility between *Populus tomentosa* WF and PHBV.

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INTRODUCTION

With the growing severity of global environmental issues, the "white pollution" caused by traditional plastics has emerged as a pressing problem that demands immediate solutions (Dwivedi *et al*. 2021). Driven by the concepts of green and sustainable development, as well as the circular economy, biodegradable materials, owing to their characteristic of being decomposable by microorganisms in the natural environment, offer a crucial approach to addressing the issue of waste plastics in the sectors where recycling is difficult or for single-use products (Mohamad *et al*. 2023). In recent years, the biodegradable materials market has experienced rapid growth (Basalp *et al.* 2020; Borkowski and Kępka-Borkowska 2025; Makani *et al*. 2025). Based on relevant studies (Velasquez *et al.* 2025), it is expected that in the coming years, its market size will continue to expand at a considerable annual growth rate. As a result, numerous scientific research

entities and enterprises have actively engaged in the research, development, and application promotion of biodegradable materials (Dokmai and Ratanawilai 2024).

Poly (β-hydroxybutyrate valerate) (PHBV), a typical biodegradable material synthesized through microbial fermentation, demonstrates great application potential in fields such as biomedicine and packaging (Kalia *et al*. 2021; Palmeiro-Sánchez *et al*. 2022). Its excellent biocompatibility makes it suitable for biomedical applications like tissue engineering scaffolds and drug sustained-release carriers. Its biodegradability also renders it an ideal substitute for traditional plastics in the field of single-use packaging (Li *et al*. 2021; Shen *et al*. 2023; Zaafarani *et al*. 2024). However, PHBV has certain inherent performance limitations. For instance, its mechanical properties are relatively weaker compared to those of traditional general-purpose plastics, which severely restricts its widespread application in some fields with high strength requirements (Haque *et al*. 2022).

To improve the performance of PHBV and reduce costs, compounding it with wood flour (WF) has become a highly promising strategy (Bütün *et al*. 2019; Friedrich 2022). WF is widely available and low in cost, and it is rich in natural polymer components such as cellulose, hemicellulose, and lignin (Yilmaz Atay and Türkmen 2022; Khamtree *et al.* 2023). By adding WF to PHBV, on the one hand, the rigidity of WF can be utilized to enhance the mechanical properties of PHBV, and on the other hand, it can significantly reduce the material cost and improve economic benefits. Nevertheless, WF is a polar material with a large number of polar groups such as hydroxyl groups on its surface, while PHBV is a non-polar material (Khoo *et al.* 2025). There are significant differences in the chemical structures and polarities between the two materials. These differences lead to poor interfacial compatibility and weak interfacial bonding between WF and PHBV in the composite material system. When the composite material is under stress, the stress cannot be effectively transferred between the two phases, and problems such as interfacial debonding are likely to occur, which seriously affects the comprehensive performance of the composite material, including its mechanical properties, processing performance, and durability, making it difficult to meet the requirements of practical applications. Therefore, improving the interfacial compatibility between WF and PHBV has become a key issue for enhancing the performance of WF/PHBV composites (WPHBVs) and promoting their practical applications (Chan *et al*. 2018; Yin *et al*. 2019; Huo *et al*. 2022).

Many investigations (Kim *et al*. 2007; Zhu *et al*. 2014; Zhou *et al*. 2017; Sohn and Cha 2018) showed that coupling agents (MAPP, silane, titanate, aluminate, isocyanate, phthalic anhydride) boost the mechanical properties of wood/polymer composites by optimizing interfacial compatibility. MAPP is widely used as it efficiently improves interfacial adhesion (Xiao *et al.* 2020; Haque 2021; Zhang *et al.* 2023).

Although MAPP can improve the interfacial compatibility between WF and polymer matrix (Kim *et al.* 2007; Souissi *et al.* 2022; Khamtree *et al.* 2024), the MAPP-modified WPC system is complex and often contains various additives (Zhang *et al.* 2019; Subramaniam *et al.* 2022). Current research on the multiphase interactions such as synergy and competitive adsorption within the system lags behind, making it difficult to comprehensively analyze the formation mechanism of the material properties (Mengeloğlu and Çavuş 2021; Farsi *et al*. 2024).

In summary, existing studies have provided abundant fundamental data on the interfacial reinforcement mechanism of MAPP. However, for WPHBVs — a type of "natural fiber-biodegradable matrix" composite — two aspects still require further verification: first, whether the matrix biodegradability affects the interfacial chemical

reaction pathway of MAPP, and second, how to regulate the synergistic effect of physical adsorption and molecular chain entanglement on interfacial stability.

This study used MAPP as the coupling agent to prepare WPHBVs by hot-pressing. The internal mechanism by which MAPP affects the interfacial compatibility between WF and PHBV was analyzed by SEM, FT-IR and TGA. Exploring this mechanism from the molecular level and in terms of physicochemical interactions provides a solid theoretical support for optimizing the interfacial bonding and the material properties.

EXPERIMENTAL

Materials and Methods

Materials

The low-cost and widely available poplar (*Populus tomentosa* Carr.) WF was purchased from Hebei Jiashuo Building Materials Processing Co. LTD, China. It was passed through an 80-mesh sieve (177 to 180μm) and oven-dried at 105 ℃ for 24 h to a moisture content of less than 3%. Poly (β-hydroxybutyrate valerate) (PHBV), with a density of 0.9 ($g \cdot cm^{-3}$) and a melt point of 176 ℃ was purchased from Xinxing Plasticizer Co. LTD, China. Maleic anhydride grafted polypropylene (MAPP, with 1.0 MA% grafting rate and a melt flow index (MFI) of 120.0 (g/10 min) was supplied by Chuangjinxin Chemical Technology Co. LTD, China, and was used as a coupling agent.

Preparation of sample

The samples were prepared using the compression molding method with dimensions of 250 × 250 × 2 mm and a target density of 1000 $kg \cdot m^{-3}$. The mass fraction of WF was 60% and that of PHBV was 40%. WPHBVs were prepared at five levels of MAPP loading (0.5%, 1%, 1.5%, 2%, and 4% of the total mass of WF and PHBV).

First, the poplar flour was dried in an oven at 103 °C for 24 h until the moisture content was less than 3%. Dried WF, PHBV, and MAPP were weighed and then mixed in a high-speed mixer with 1800 r/min for 3 min. The mixture was oven-dried, distributed into the mold, and then hot-pressed. The hot press (Carver 3895, manufactured by Mecono Technologies Co., Ltd.) was used to press the panels at 180 °C and 7tons for 10 min. After hot pressing, the formed panel was further pressed at 3 MPa for another 3 min at room temperature using a cold press (XLB, manufactured by China Yadong Machinery Group Co., Ltd.).

In this study, the experimental design and specimen dimensions were determined in accordance with ASTM D7031-11 (2019). To guarantee the reliability of experimental data and minimize random errors, three parallel replicate specimens were prepared for each test group. Specimens with dimensions of 80 mm × 10 mm × 2 mm were used for mechanical property testing, while specimens of 5 mm × 5 mm × 2 mm were adopted for thermal stability testing, with the dimensional deviation controlled within ±0.1 mm. All specimens were cut using a hand saw, trimmed to eliminate edge defects, screened, and uniformly numbered prior to testing. The preparation is shown in Fig. 1.

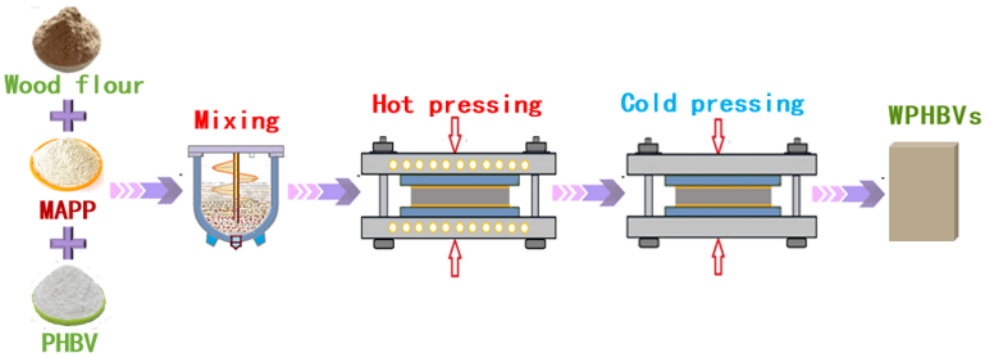


Fig. 1. Process diagram of the preparation

Characterization of Composite Specimens

Mechanical property test

The cut WPHBV specimens were mounted on a universal testing machine (Instron 3369, manufactured by Instron Corporation, USA) for three-point bending tests, following ASTM D7031-11 (2019).

The load was applied evenly during the test, and the loading speed was set at 1 mm/min. The maximum load value was recorded. Finally, the modulus of rupture (MOR) and bending modulus of elasticity (MOE) were calculated according to the formula, and the results were taken as the average of the five samples.

Scanning electron microscopy (SEM)

The morphology of the dispersed phase was observed *via* scanning electron microscopy (SEM, Model JSM-IT300LV, JEOL Ltd., Tokyo, Japan) at a magnification of 1000×. Prior to testing, the samples were sputter-coated with gold using an ion sputter coater (Model JFC-1200, JEOL Ltd., Tokyo, Japan) to eliminate charge accumulation, and the SEM scanning voltage was set to 10 kV.

Fourier transforms infrared (FT-IR) microscopy

The samples (5 to 10 mg) were ground to a particle size ⩽2μm in an agate mortar, then mixed with dried spectroscopic-grade KBr. The mixture was pressed into a thin sheet using a tablet press, followed by sample loading for testing. FT-IR spectra analysis was carried out taken on a ThermoFisher Scientific iS5 instrument (Thermo, USA). The resolution of each spectrum was 4 cm^{-1}, obtained with 16 scans. The changes in the chemical structure of the WPHBVs were analysed.

Thermogravimetric analysis (TGA)

The thermal stability of the samples was measured under 20 mL/min N_2 flow atmosphere with a TGA-Q50 thermogravimetric analyzer (TA, USA). The heating process was conducted from room temperature to 600 ℃ at a scanning rate of 10 ℃/ min. The thermogravimetric curves (TGA) and differential thermogravimetric curves (DTG) were obtained.

RESULTS AND DISCUSSION

Mechanical Property Analysis

Figure 2 shows that with the gradual increase of the MAPP addition amount, MOR and MOE of WPHBVs exhibit a typical trend of first increasing and then decreasing. When the MAPP addition amount is precisely regulated to 2%, all mechanical property indexes of WPHBVs reach their peak values. Among them, MOR increased to 27.99 MPa, and MOE increased to 3690.47 MPa. Compared with the original WPHBVs without the introduction of MAPP, their mechanical properties were significantly enhanced.

PHBV is a brittle polymer material, showing characteristics of high tensile strength and modulus (Jiang *et al*. 2018). However, its high crystallinity leads to poor ductility, that is, the elongation at break shows poor performance. After introducing MAPP into the system, the acid anhydride functional groups in the molecular structure of MAPP undergo an esterification condensation reaction with the hydroxyl groups on the surface of the *Populus tomentosa* wood fibers. This reaction mechanism effectively enhances the interfacial compatibility among various components of the composite system. This enables the formation of a stable bond between PHBV and wood fibers in the interfacial region. When the material is subjected to external loads, the stress can be efficiently transferred from the PHBV phase to the wood fiber phase through this interface. This interphase stress transfer mechanism acts on the entire WPCs, significantly alleviating the phenomenon of stress concentration, and thus essentially improving the mechanical strength of WPHBVs. In conclusion, precisely adding an appropriate amount of MAPP to the WPHBVs system can induce a more excellent "coupling synergy effect" (Ayana *et al*. 2025). However, when the addition amount of the MAPP exceeds the appropriate range, an excessive reaction will occur, and the excess MAPP will remain in a free state between the wood fibers and the PHBV matrix (Jiang *et al*. 2018; Zhu and Ma 2019; Gunjal *et al*. 2020;). This will increase the probability of slippage between molecular chains, ultimately leading to the deterioration of the mechanical properties of WPHBVs.

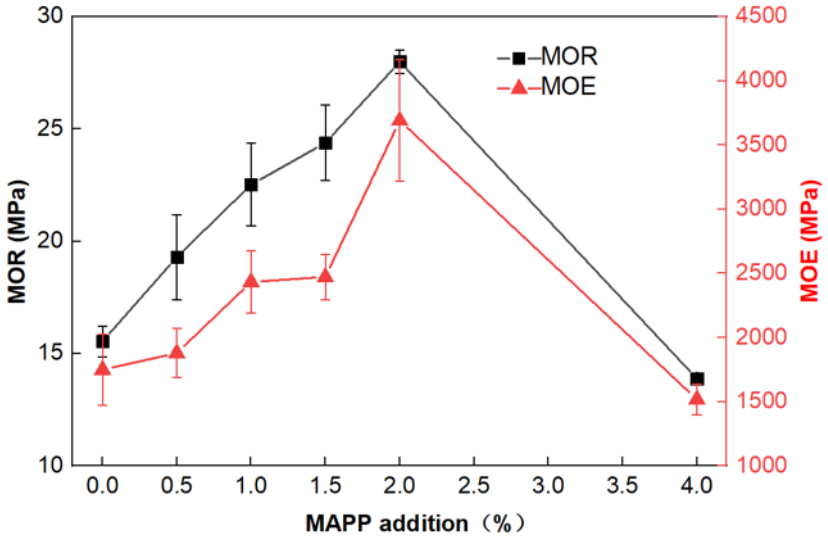


Fig. 2. The MOR and bending MOE of WPHBVs with different amount of MAPP

SEM Analysis

Figure 3 shows that fiber pull-out and fiber fracture are the main forms of structural damage in WPHBVs. Figure 3(a) shows the microscopic structure of the cross-section of WPHBVs without the addition of MAPP. The rod-shaped wood fibers in the cross-section of WPHBVs are fractured, and the fiber surface is rough, with many grooves and cracks (Chandrasekar *et al*. 2021). The bonding between PHBV and wood fibers is not tight. Therefore, when subjected to external forces, the wood fibers are directly pulled out from the matrix structure, and stress transfer cannot be achieved in a timely manner between the wood fibers (Lazrak and Hammi 2024), which is likely to cause stress concentration, making WPHBVs prone to fracture. This indicates that WPHBVs have poor mechanical properties when no coupling agent is added (Singh *et al*. 2008).

In Fig. 3(b), due to the addition of MAPP, PHBV begins to coat the wood fibers. The grooves and cracks on the wood fibers in the cross-section are significantly reduced, and the surface roughness is remarkably decreased. This shows that the addition of MAPP effectively improves the interfacial bonding force, enhances the compatibility between PHBV and wood fibers, and thus increases the mechanical strength of WPHBVs to some extent. When WPHBVs are subjected to external forces, there is a phenomenon of "being pulled out", and at this time, both ductile fracture and brittle fracture coexist.

In Fig. 3(c), with the increase of the MAPP addition amount, more and more PHBV fills between the wood fibers and gradually covers and coats the wood fibers. The roughness of the cross-section of the wood fibers continuously decreases. Moreover, when subjected to external forces, a "necking" phenomenon can occur, and at this time, the main form of structural damage basically changes from brittle fracture to ductile fracture. This indicates that the increase in the MAPP addition amount makes the connection between PHBV and wood fibers tighter. When the MAPP addition amount is 2%, the surface of the wood fibers is almost completely coated by PHBV, and the surface roughness reaches the lowest value. At this time, under the action of the coupling agent, the interfacial bonding between PHBV and wood fibers is the best, and the mechanical properties of WPHBVs reach the optimal state.

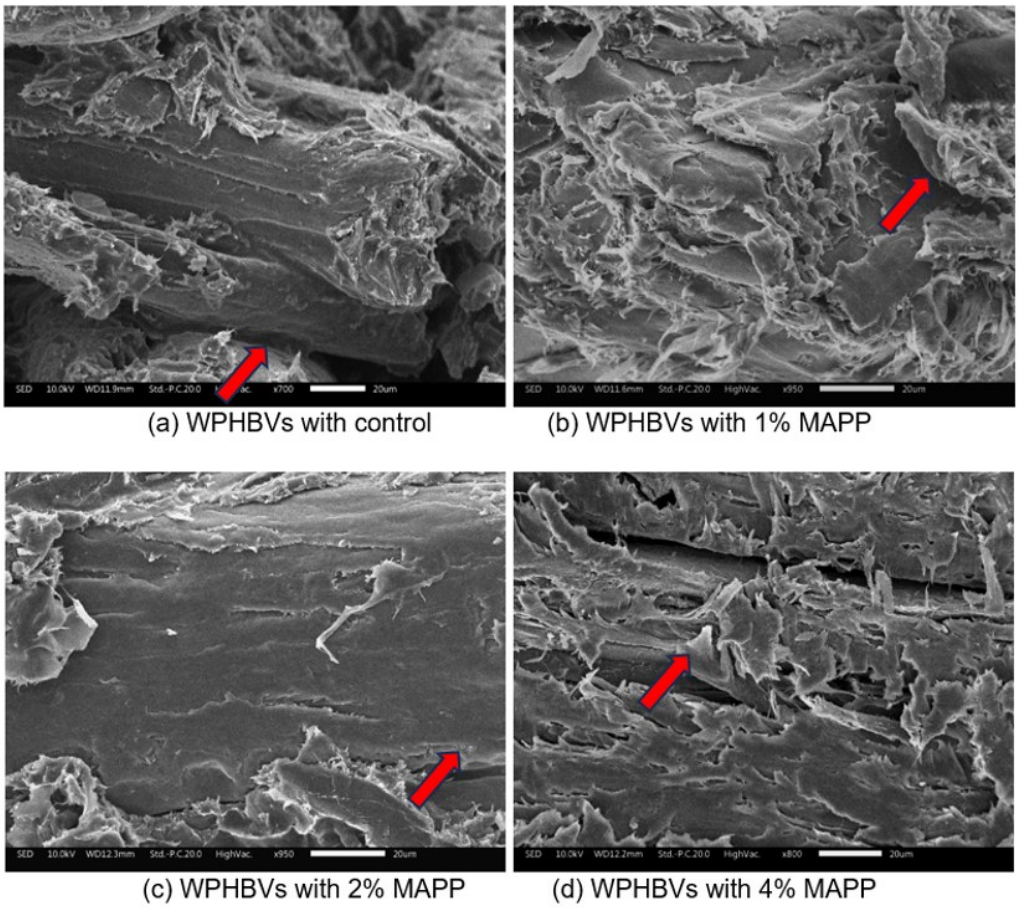


Fig. 3. Cross-sectional SEM of WPHBVs with different amounts of MAPP (The red arrows in the figure indicate grooves and cracks)

As shown in Fig. 3(d), when the MAPP addition amount is 4%, excess unreacted PHBV begins to appear on the surface of the wood fibers in the cross-section, as well as crystals formed by the precipitation of unreacted MAPP. The roughness of the cross-section increases again, and the number of grooves and cracks on the surface of the wood fibers increases significantly. At this time, the bonding between PHBV and wood fibers is no longer tight, and the mechanical properties decline greatly. This is because the excessive addition of the coupling agent will accumulate between the wood fibers and PHBV, forming a weak MAPP matrix layer. When WPHBVs are impacted, they will fracture at the weak MAPP matrix layer (Zhu *et al*. 2014). Therefore, the excessive addition of the coupling agent will lead to a significant decrease in the mechanical strength of WPHBVs.

FT-IR Analysis

Figure 4 shows the characteristic peaks in WPHBVs, with peak values at 1427 cm^{-1}, 1720 cm^{-1}, 2877 cm^{-1}, 2930 cm^{-1}, and 3427 cm^{-1}. Among them, the characteristic peak at 1720 cm^{-1} originates from the stretching vibration of C=O in wood fibers and PHBV. The characteristic peak at 2877 cm^{-1} corresponds to the symmetric and asymmetric structures of -CH_2- in PHBV. The characteristic peaks at 2930 cm^{-1} and 1427 cm^{-1} are respectively the characteristic peak of saturated alkane C-H in wood fibers and the stretching vibration peak of saturated C-H. The characteristic peak at 3427 cm^{-1} is the stretching vibration peak of O-H in wood fibers.

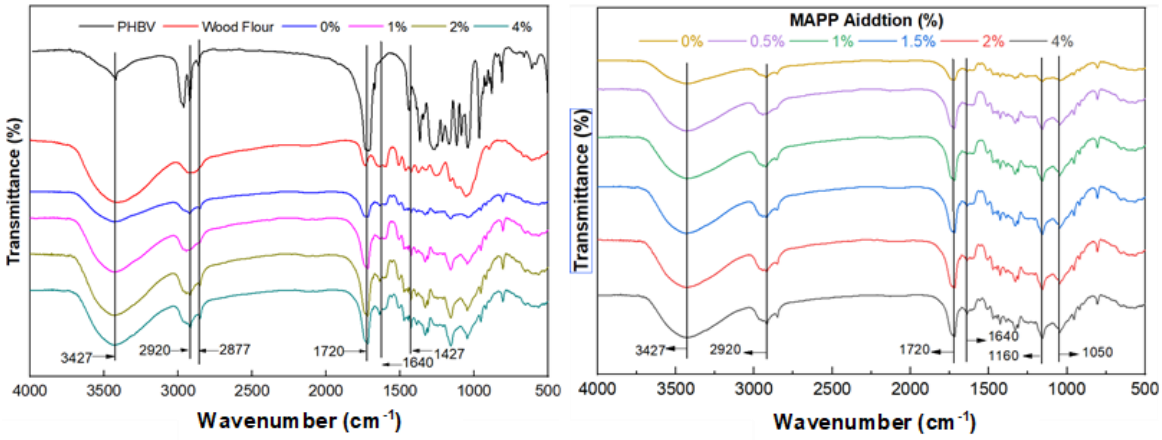


Fig. 4. Infrared spectra of WPHBVs with different amount of MAPP

The peak at 3427 cm^{-1} is the vibration peak of intermolecular hydrogen bond O-H. After adding MAPP, the peak value of this O-H vibration peak significantly increases. In addition to the large number of hydroxyl groups contained in wood fibers, it may also be due to the decomposition of the ester bonds in PHBV during the hot pressing process, which generates hydroxyl groups and carboxyl groups. The peak at 1720 cm^{-1} is the stretching peak of the ester group C=O in the molecular chain of the highly ordered crystalline structure, and the peak at 1640 cm^{-1} represents the stretching vibration peak of C=O in the glassy region of PHBV in the semi-crystalline state (Padermshoke *et al*. 2004) After adding MAPP, the peak values at these two positions increase, indicating that new ester groups are formed in WPHBVs. It also means that the number of crystalline regions and amorphous regions in WPHBVs is increasing, which is because wood fibers enhance the crystallization performance of PHBV. Moreover, the peak value at 2920 cm^{-1} increases, indicating that after adding MAPP, a reaction occurs during the preparation of WPHBVs, generating substances with saturated alkane structures.

Thermogravimetric Analysis

Figure 5(a) shows the decomposition curves of WPHBVs without MAPP addition and those modified with different MAPP addition amounts generally exhibit similar trends. Both display two thermal degradation states, and overall, they can be divided into three stages. The first stage occurs in the temperature range of 240 to 270 °C. This stage represents the main decomposition period of PHBV and also the initial stage of the pyrolysis of *Populus tomentosa* WF. The second stage lies in the range of 270 to 390 °C, which is the main decomposition stage of *P. tomentosa* WF. The third stage commences after 390 °C, during which the fiber residues further undergo pyrolysis until the reaction concludes.

PHBV shows thermal instability above 240 °C, and its thermal degradation process involves chain scission and hydrolysis. As the MAPP addition amount increases, the decomposition curves in the first stage all shift towards the high-temperature direction. This indicates that the addition of MAPP can enhance the thermal stability of PHBV. The reason for this phenomenon is that in the WPHBVs system, the treatment with MAPP strengthens the interfacial adhesion between Populus tomentosa WF and PHBV, and increases the crystallinity of the PHBV matrix, thereby raising the thermal degradation temperature of PHBV. In the second stage, the changes in the decomposition curves show

no obvious pattern. The curves with MAPP addition amounts of 4% and 2% are located at the leftmost and rightmost positions respectively. This shows that the addition of MAPP can alter the molecular structure of *P. tomentosa* WF to some extent, but it does not have a regular impact on the overall thermal stability of WPHBVs.

Figure 5(b) shows the DTG curves of WPHBVs with different MAPP addition amounts, with a heating rate set at 10 °C/min. There are mainly two weight-loss peaks in the figure, namely the weight-loss peak of PHBV around 270 to 300 °C and the weight-loss peak of WF around 380 °C. When the MAPP addition amount increases, the weight-loss peak around 270 to 300 °C shows a right-shifting trend, indicating that the addition of MAPP can improve the thermal stability of WPHBVs. After the addition of MAPP, the weight-loss peak around 380 °C also shifts to the right, showing a lag phenomenon. This means that the decomposition rate of WPHBVs slows down. The reason is that MAPP improves the thermal stability of WPHBVs: the adhesion of the generated crystals to the WF fibers increases the thermal decomposition temperature of *P. tomentosa* WF and slows down the carbonization rate.

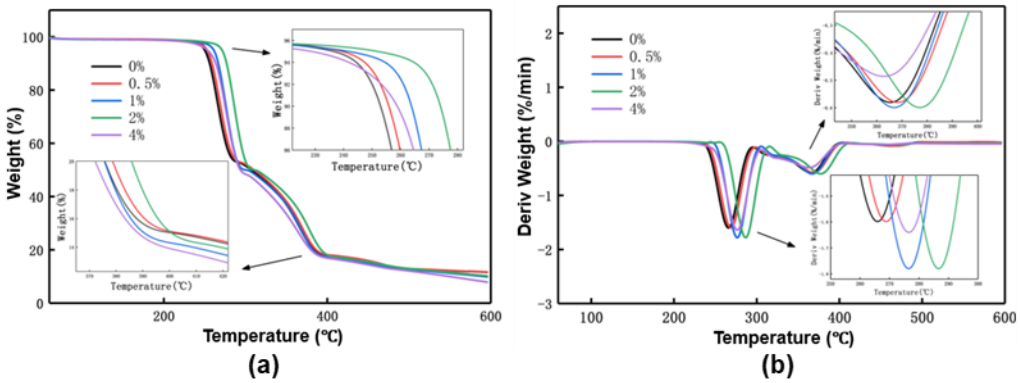


Fig. 5. (a) TGA curve and (b) DTG curve of WPHBVs with different amount of MAPP

CONCLUSIONS

1. As the addition amount of maleic anhydride grafted polypropylene (MAPP) increased, the bending strength and elastic modulus of WF/PHBV composites (WPHBVs) first increased and then decreased. Compared with the WPHBVs without the addition of the coupling agent, the appropriate addition of MAPP improved their mechanical properties. When the addition amount of MAPP was 2%, the WPHBVs exhibited the best comprehensive mechanical properties. The flexural strength and elastic modulus of WPHBVs reached their maximum levels, which could be as high as 27.99 MPa and 3690.47 MPa respectively, and the strength enhancements were all above 40%.
2. When 2% MAPP was added, the fracture surface was the flattest and the roughness was the lowest, indicating that the *Populus tomentosa* wood flour and PHBV were the most closely bonded. However, an excessive amount of MAPP accumulated between the wood fibers and PHBV, resulting in a looser bond between the two and an increase in the roughness of the fracture surface.
3. The FT-IR analysis showed that after the addition of MAPP, the numbers of ester bonds and hydrogen bonds increased, indicating that grafting and esterification reactions occurred. These bonds enhance the interfacial bonding force and compatibility between the *Populus tomentosa* wood flour and PHBV.

4. WPHBVs without MAPP and those modified with different MAPP amounts show generally similar decomposition curves. Both have two thermal degradation states and can be divided into three stages overall, indicating MAPP does not alter WPHBVs' basic thermal degradation mechanism. But as the addition amount of MAPP increased, the thermal decomposition temperature of WPHBVs rose, the decomposition rate decreased, and the thermal stability improved. The WPHBVs exhibited the best thermal properties with 2% MAPP.

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Disclosure statement

No potential conflict of interest was reported by the author(s).

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