Chip Morphology's Effect on Properties of PLA-based Bamboo–Plastic Composites Produced Using Hot-pressing

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This study explored the effect of raw material morphology on the properties of bamboo–plastic composites produced using hot-pressing. To provide a reference for reducing the production cost and improve the product properties of the composites, polylactic acid (PLA)-based bamboo–plastic composites were prepared using bamboo chips with a shaved morphology (BS) and fiber morphology (BF) and PLA as the matrix material, *via* hot-pressing. The properties of the bamboo–plastic composites formed with BS and BF chips were studied and compared with those of composites with conventional granular morphology (BM) and powder morphology (BP). The results showed that when the content of the bamboo chips was at 50% (the same below), the mechanical properties of the BF/PLA composites were remarkably better than those of the other PLA-based composites. However, the BF/PLA composites showed a high degree of hydrophilicity, with a water contact angle of 70.0° and a water absorption of 10.8% at 288 h. More holes could be seen in the BF/PLA composites using a scanning electron microscope. Among the four types of PLA-based composites, better melt fluidity was found only in the BF/PLA composites, and the melting index was 65.3 g/min.

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INTRODUCTION

With the increasing awareness of the urgency of environmental and resource sustainability, the use of green and sustainable plant fibers to prepare composite materials has attracted much attention (Khan *et al*. 2023). Plant fibers, including wood fiber, bamboo fiber, and straw fiber, can be used as reinforcement or filler materials in thermoplastic resins to replace non-renewable synthetic fibers derived from petroleum. They possess various advantages such as light weight, good degradability, and sustainability (Thamae *et al*. 2008; Zhang and Ma 2018). In recent years, the application of plant fiber-based thermoplastic composites has been expanding in fields such as construction, transportation, and daily use products.

Generally, there are three types of molding method for plant fiber thermoplastic composites: extrusion molding, injection molding, and hot-press molding. Of these, extrusion molding and injection molding are the most widely used, but there are problems associated with their use. These include low production efficiency, low plant fiber addition ratio (generally 30% to 40%), limited fiber shape size (usually 40 to 100 mesh), and high energy consumption (Wang and Sheng 2019). Hot-pressing technology has various advantages, such as the material processing is not affected by the shape of the plant fiber, the product shape can be varied according to the mold, it is associated with a high production efficiency, simple operation, and low power energy consumption; thus, the technology has become one of the most popular research directions in recent years (Friedrich 2021; Li 2021). Liu (2014) compared and analyzed the effects of different forms of wood fibers on the properties of wood fiber (WF) / polylactic acid (PLA) composites prepared using hot-pressing, and found that the bending strength and elastic modulus of WF/PLA composites prepared from medium forms of wood fibers with a diameter of 0.012 to 0.05 mm were higher than those of the other two kinds of wood-plastic composites. Qi *et al*. (2016) prepared oriented wood–plastic composite (WPC) particleboard using hot-pressing and blending poplar wood shavings with HDPE film. When the HDPE mass fraction reached 12%, the overall performance of the particleboard was the best. Using wood flour and PE resin as raw materials, Fan (2019) studied the effects of the hot-pressing time, hot-pressing temperature, and material density on physical and mechanical properties, such as the static bending strength, elastic modulus, and screw holding force. The results showed that the tensile and bending strengths and elongation at the point of breakage of the composite materials increased first and then decreased with the increase in temperature. Cao *et al*. (2020) made WPCs using hot-pressing and blending *Eucalyptus* wood silk and filamentary HDPE as raw materials. They considered the properties and possible applications of the WPCs prepared using this method in packaging. Li *et al*. (2019) studied the influence of PVC film perforation pretreatment on the physical and mechanical properties of, and the lamination effect exhibited by, a new WPC to explore and solve the heat transfer problem in the hot-pressing process.

Bamboo resources are abundant in China, and they are known as the "second forest". However, due to its thin wall, hollow center, sharp cutting degree, and other characteristics, bamboo produces a lot of leftovers during its processing, and the utilization efficiency of raw materials is generally low. With the increase in labor and transportation costs, how to make high-quality and efficient use of bamboo processing leftovers has become a key issue in improving the comprehensive utilization rate of bamboo (Zheng *et al*. 2021). In recent years, with the development of bamboo pulp and paper making, the production of bamboo fiber has gradually increased. The application of bamboo shavings and bamboo fibers in boards is mainly in the production of bamboo particle board or bamboo fiber board bonded using urea-formaldehyde resin and other adhesives. Formaldehyde is released during this process (Gao *et al*. 2022), which will cause harm to the environment and personnel. Bamboo–plastic composite materials are mostly made of bamboo powder after it is screened, while polylactic acid (PLA) is a biodegradable thermoplastic resin made from plant corn (Fan *et al*. 2020; Xue *et al*. 2020). PLA has been widely studied and applied (Shojaeiarani *et al*. 2022; Wang *et al*. 2022). In this paper, bamboo–plastic composites were prepared using the hot-pressing process, and the properties of the PLA-based bamboo–plastic composites prepared with wood shavings and fibrous bamboo fragments were studied. The effects of the morphology of the fragments on the properties of the composites were compared with those of bamboo–plastic composites made from commonly used particle and powder forms of bamboo. This study aims to provide a reference for reducing the production cost of bamboo plastic composite materials, improving product performance, and expanding the applications of bamboo processing residues in molding materials.

EXPERIMENTAL

Materials

Bamboo shavings (BS), leftovers from bamboo strip processing, were provided by Jiangxi Shuangshuang Machinery Manufacturing Co., Ltd., China. They were dried in the oven at 103 ℃ until the moisture content was less than 3% before use.

Bamboo granules (BM), the remaining material from bamboo strip roughing, were obtained from Jiangxi Shuangshuang Machinery Manufacturing Co., Ltd., China. They were screened, passing through 60 mesh and being retained on 90 mesh sieves, and dried in the oven at 103 ℃ until the moisture content was 3%.

Bamboo powder (BP), which is the leftovers from bamboo strip roughing process, was obtained from Jiangxi Shuangshuang Machinery Manufacturing Co., Ltd., China. It was sieved with 120 mesh to 150 mesh sieves, dried in the oven at 103 ℃ until the moisture content was less than 3% before use.

Bamboo fiber (BF), the leftovers of bamboo, was processed with biological enzymes and synthesized through mechanical grinding. They were provided by Jiangxi Heide Bamboo Industry Development Co., Ltd., China, and dried in the oven at 103 ℃ until the moisture content was less than 3% before use.

Polylactic acid resin (PLA), grade 4032D, density 1.25 g/cm^3, melting temperature 160 ℃, glass transition temperature 58 ℃, was purchased from Nature Works (Blair, NE, USA) and dried at 60 ℃ oven for 24 h for reserve use.

Test Standards

Shape test of different bamboo scraps

The shape characteristics of different bamboo fragments were observed under a stereoscopic microscope (SZX10, OLYNPUS), and the diameter and length–diameter ratio of the bamboo fragments were measured under an electron microscope (BK6000, Chongqing Optec Instrument Co., Ltd.).

Mechanical property tests of PLA-based bamboo–plastic composites

The tensile and bending properties of the composites were tested using a universal mechanical testing machine (CM755, MTS). The tensile properties were tested according to GB/T 1447 (2005), using the specimen size of 80 mm × 10 mm × 4 mm, and the tensile speed was 5 mm/min. The bending performance was tested according to GB/T 9341 (2008). The size of the specimen was 80 mm × 10 mm × 4 mm, the distance between the supports was 30 mm, and the drop speed of the indenter was 5 mm/min. The impact properties of the composites were tested using a pendulum impact testing machine (PIM550B-2, Shenzhen Tianyi Technology Co., Ltd), and the impact properties of the non-notched composites were measured according to GB/T 1043.1 (2008).

Water absorption test of PLA-based bamboo–plastic composites

According to GB/T 17657 (2022), a water absorption test was performed to determine the water absorption of the composite materials. A specimen with the dimensions of 50 mm × 50 mm × 4 mm was immersed in a glass container filled with distilled water at 23 ℃ and sealed with metal mesh to ensure the specimen was fully immersed in the water.

Surface wettability test of PLA-based bamboo–plastic composites

The composite material was processed into a specimen with the dimensions of 200 mm × 20 mm × 4 mm. The surface of the specimen was rinsed with deionized water and dried until it was completely dry. Deionized water was used as the liquid phase to test the surface wettability of the composite using an optical contact angle measuring instrument (HARKE-SPCAX3, Beijing Hake Test Instrument Factory). The contact angle of the blend was determined by drawing the contact angle change curve and observing the number of contact angles displayed after it becomes stable. Each sample was tested 5 times, and the average value was used as the final result.

Profile analysis of PLA-based bamboo–plastic composites

The surface morphology of the tensile section of the composites was observed using cold field emission scanning electron microscopy (JSM-7500F, Hitachi). Test conditions used were voltage of kV and current of 10 mA.

Melting Index

Referring to GB/T 3682.1 (2018), the melting index of the composites were tested at 190 ℃ and a load of 2.16 Kg as the melt index of the blend by the melt index meter (GOTTFERT MI-2.2, GOTTFERT).

Methods for Composite Preparation

To prepare low-cost and high-performance bamboo-plastic composite materials, the proportion of fixed bamboo scraps used was 50%. The dried PLA particles were put into the open mill machine (SY-6215-A, Shiyan Precision Instrument Co., Ltd) for mixing and melting at 180 ℃. After melting completely, the fully dried bamboo scraps were added step by step and blended until they were fully incorporated and mixing continued for 15 min. Immediately after the mixing was completed, the mixture was quickly poured into a mold with dimensions of 200 mm × 200 mm × 4 mm and molded in the hot-pressing machine (HBSCR100T/600X, Qingdao Xincheng Huabo Machnology Co., Ltd). The hot-pressing was performed at 170 ℃, time of 10 min, and pressure of 10 MPa. After the hot-pressing was completed, the material was quickly sent to the cold-pressing machine (XLB500×500×2, Qingdao Xincheng Yiming Rubber Machinery Co., Ltd). The cold-pressing was performed at pressure of 10 MPa and pressing time of 6 min. The molded material was sealed and stored for 24 h before being made into standard splines for testing.

RESULTS AND DISCUSSION

Microscopic Morphologies of Four Kinds of Bamboo Fragments

Figure 1 shows the morphologies of the four kinds of bamboo fragments observed under a microscope at 6-fold magnification. As shown in Fig. 1 and Table 1, the BS was flaked and curly, its length and width were large, and it had a certain thickness; the length range was 1500 to 8000 μm, the width range was 300 to 5000 μm, and the length-to-diameter ratio was 1.0 to 2. The length of BM was 300 to 1000 μm, the width was 100 to 300 μm, and the length-to-diameter ratio was the highest, in the range of 2 to 4 (54%). The BP length–diameter ratio was small, ranging from 40 to 250 μm in length and 15 to 250 μm in width. The length–diameter ratio was the greatest in the 1 to 2 range, accounting for 59% of the BP. The BF was needle-like, its diameter was small, and it was easy to interweave together because the mechanical grinding process was not performed; due to the inclusion of the thick stalk, its width was up to 300 μm, and compared with the first three, its length–diameter ratio range was wider, in which 22% of the BF had a length–diameter ratio in the range of 1.0 to 2, and 57% of the BF had a length–diameter ratio in the range of 4 to 10 or more than 10.

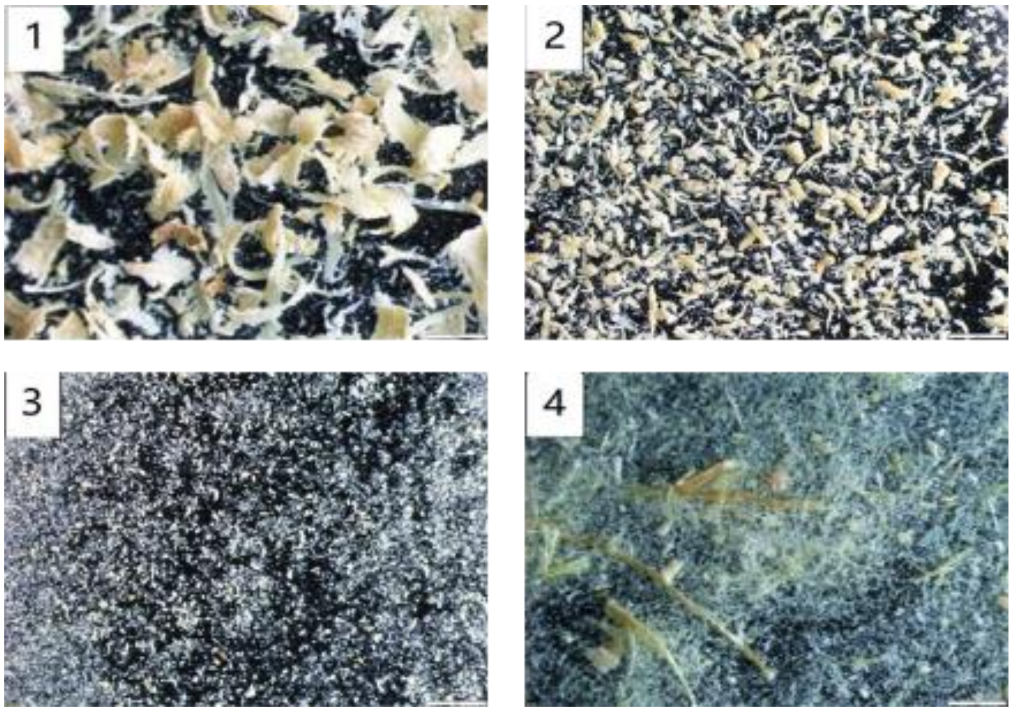

Fig. 1. Morphology of bamboo chips with different shapes under 6.2X body microscope: 1: BS; 2: BM; 3: BP; and 4: BF

Table 1. Analysis of the Aspect of Bamboo Chips

Chips Morphology	Chips Length (μm)	Chips Width (μm)	Length–Diameter Ratio			
			1 to 2	2 to 4	4 to 10	Over 10
BS	1500 to 8000	300 to 5000	57%	25%	18%	/
BM	300 to 1000	100 to 300	20%	54%	26%	/
BP	40 to 250	15 to 80	59%	23%	18%	/
BF	200 to 3000	8 to 300	22%	21%	39%	18%

Mechanical Properties Analysis

Figure 2 shows the mechanical properties of the PLA-based bamboo–plastic composites prepared in four forms. The bending and tensile strengths of the planed bamboo–plastic composites (BS/PLA) were 62.7 and 50 MPa, respectively, which was only slightly lower than those of the granular bamboo–plastic (BM/PLA) and powdered bamboo–plastic (BP/PLA) composites. The bending and tensile strengths of the BM/PLA composite were 3.0% and 8.7% lower than those of the BM/PLA composite, and the bending modulus and tensile modulus were slightly higher than those of the BM/PLA and BP/PLA composites. The bending performance of the BS/PLA composite material was much higher than the rigidity requirements for a bending strength ≥ 20 MPa and bending modulus ≥1800 MPa according to the national standard GB/T 24137 (2009). The bending strength, bending modulus, and tensile strength of the BF/PLA composites were considerably higher than those of other bamboo–plastic composites: 87.9, 5830, and 68.2 MPa, respectively, which were 35.9%, 19.8%, and 24.5% higher than those of the common BM/PLA composites, respectively. The elongation at breakage and impact toughness of the composites were affected by the shape of the bamboo fragments. The larger the shape, the smaller the elongation at breakage and impact toughness. The elongation at breakage and impact toughness of the BS/PLA composites were relatively low, at 2.54% and 3.88 kJ/m^2, respectively, which were 8.6% and 25.1% lower than the common BM/PLA composites. The fracture elongation and impact toughness of the BF/PLA composites were 3.50% and 8.67 kJ /m^2, respectively, which were 25.8% and 67.4% higher than those of the common BM/PLA composites, respectively. The mechanical properties of the BF/PLA composite were better, which may be due to the smaller BF monomer and the full contact made between the PLA and BF in the melting process, thus improving the structural compactness of the bamboo–plastic composite (Ayrilmis *et al*. 2012). At the same time, the overall length and diameter of the BF were larger, and an interwoven network structure was easily formed in the composite's structure (Landes and Letcher 2020). This may improve the strengthening effect on the matrix to a certain extent, which is prominent in the impact performance. In addition to the impact toughness, the other mechanical properties of the BS/PLA composites studied in this paper were not noticeably different from those of the common BM/PLA composites, which indicates that the wood particle form has the potential for applications in the preparation of bamboo–plastic composites through the use of the hot-pressing method. If successfully applied, it has significance for reducing the production energy consumption and thus the production cost of the creation of bamboo–plastic composites.

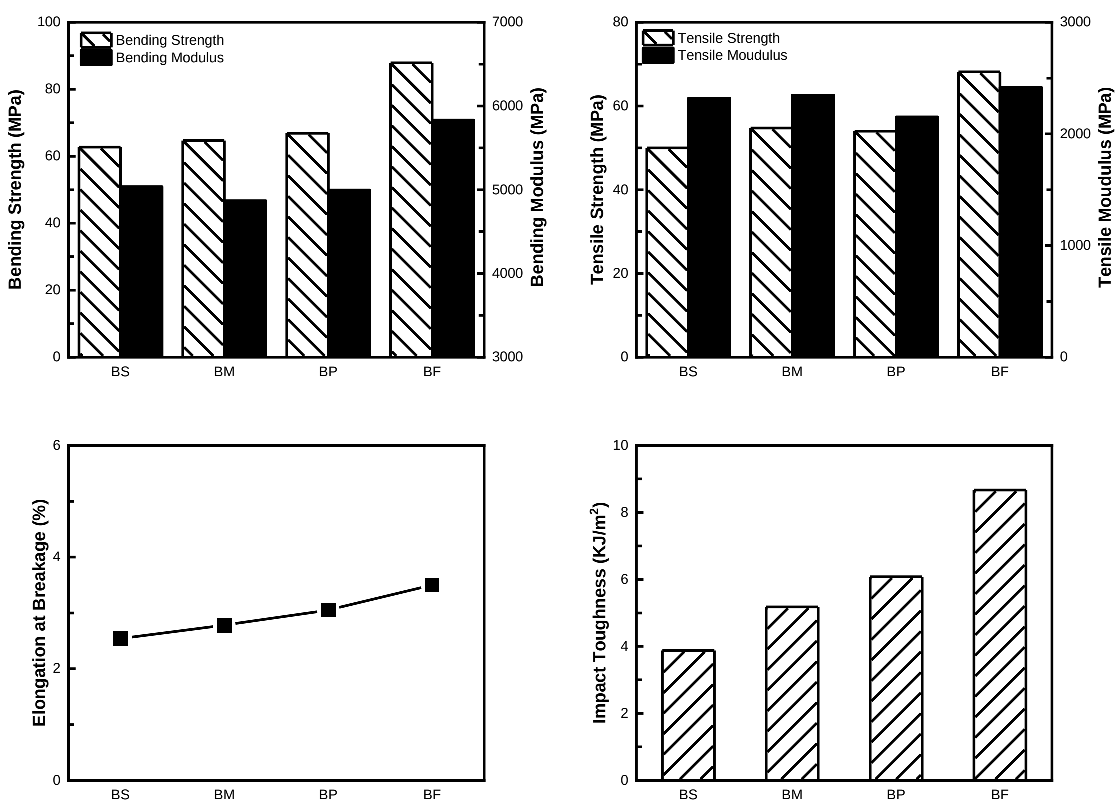

Fig. 2. Mechanical properties of PLA-based bamboo-plastic composites with four morphologies of bamboo chips

Water Absorption Analysis

The water absorption of the PLA-based bamboo–plastic composite with four kinds of bamboo fragments is shown in Fig. 3. The trend in the water absorption of the BS/PLA and BF/PLA composites was similar to that of the BM/PLA and BP/PLA composites. With the extension of time, the water absorption of the four kinds of composite materials increased rapidly at first and then slowly, and the increase in the water absorption slowed down after 72 h. The water absorption of the BF/PLA composite was higher than that of the other three forms, reaching 3.5% at 24 h and 10.83% at 288 h, which was 83.2% and 45.8% higher than that of the BM/PLA composite with the lowest water absorption, respectively. The high water absorption of the BF/PLA composite may be due to its small BF monomer and larger specific surface area (Zhao *et al*. 2023), which exposes more hydroxyl groups on the composite surface, which easily combine with hydroxyl groups in water to undergo wet expansion (Xu *et al*. 2021), resulting in greater water absorption. The water absorption of the BS/PLA composite was slightly higher than that of the BP/PLA composite, and its water absorption at 24 h and 288 h was 29% and 12.5% higher, respectively. Among the four forms of bamboo fragments, the BS fragment was on average the largest and had less hydroxyl groups exposed, but its composite material has the second highest water absorption rate, which may be due to the large size and partial curling of the BS fragment, which cannot make full contact with PLA, forming more pores and defects in the composite material and increasing the opportunity for water molecules to enter (He *et al*. 2014).

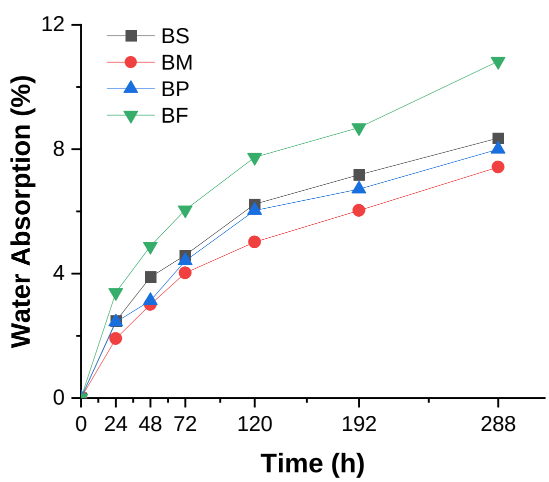

Fig. 3. Water absorption of PLA-based bamboo-plastic composites with four morphologies of bamboo chips

Contact Angle Analysis

The contact angle is a characteristic that reflects the wettability of the surface of the composite material, in addition it results from the attraction and surface tension between the surface of the composite material and the medium molecules. The contact angles of the bamboo–plastic composites prepared from four kinds of bamboo scraps are shown in Fig. 4 and Table 2. The contact angles of the four bamboo composites were all less than 90°, and they were hydrophilic, which was attributed mainly to the strong water absorption of bamboo. In addition, the amount of bamboo fragments added in this study was as high as 50%. The contact angle of the BF/PLA composite was the lowest (69.95°); the contact angle of the BS/PLA composite was close to that of the BP/PLA composite (75.90°); and the BM/PLA composite's contact angle was the largest. The variation in the surface contact angle of the bamboo–plastic composites with different morphologies was consistent with the water absorption of the different composites. Due to the large surface area of the BF, there were more hydroxyl groups, more wettability, lower contact angle, and more water absorption capabilities (Ye 2023). Because of the large size of the shavings, there may be insufficient contact between the shavings and PLA, so that water molecules could easily enter, and the contact angle was relatively small.

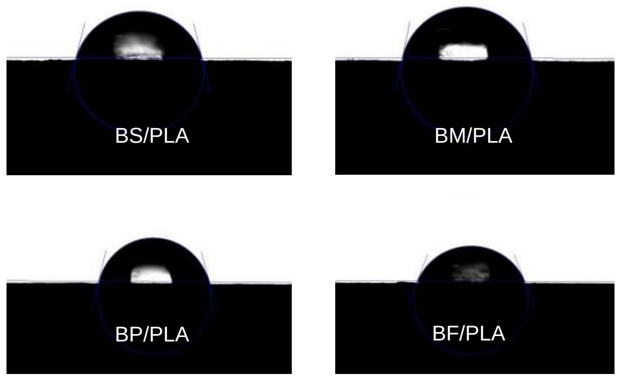

Fig. 4. Water wettability of PLA-based bamboo-plastic composites with four morphologies of bamboo chips

Table 2. Surface Contact Angle of PLA-based Bamboo-Plastic Composites with Four Morphologies of Bamboo Chips

Composite Material	BS/PLA	BM/PLA	BP/PLA	BF/PLA
Contact Angle (°)	75.9	77.9	76.0	70.0

Analysis of Section Morphology

The SEM morphologies of the four kinds of bamboo–plastic composites are shown in Fig. 5. Due to the large individual size of the BS, they appeared in a small number in the composite structure. From the view of the cross-section, the shavings were thick and long strips, and the combination with the PLA matrix had an appearance similar to that of reinforced concrete. This may also be the reason why the mechanical properties of the BS/PLA composite were not sharply reduced compared with the BM/PLA and BP/PLA composites to a certain extent. Compared with the BS/PLA composite, the remaining three forms of bamboo fragments were relatively dispersed and more evenly distributed in the composite, but there was also a partial agglomeration phenomenon, which was caused by the difference in the polarity between the two phases of the bamboo fragments and PLA matrix (Xie *et al*. 2020). In this experiment, no interface modification was made in the preparation of the bamboo–plastic composite, and the two-phase interface was obvious at the joint. There were more fine fibers in the BF/PLA composite material. Most of the bamboo fibers were wrapped in the PLA matrix, with a small amount exposed outside the matrix, and some traces were left because the fibers were not tightly combined with the matrix. At the same time, compared with the other bamboo–plastic composites, more pores can be seen in the BF/PLA composite, which is one of the reasons for its higher water absorption.

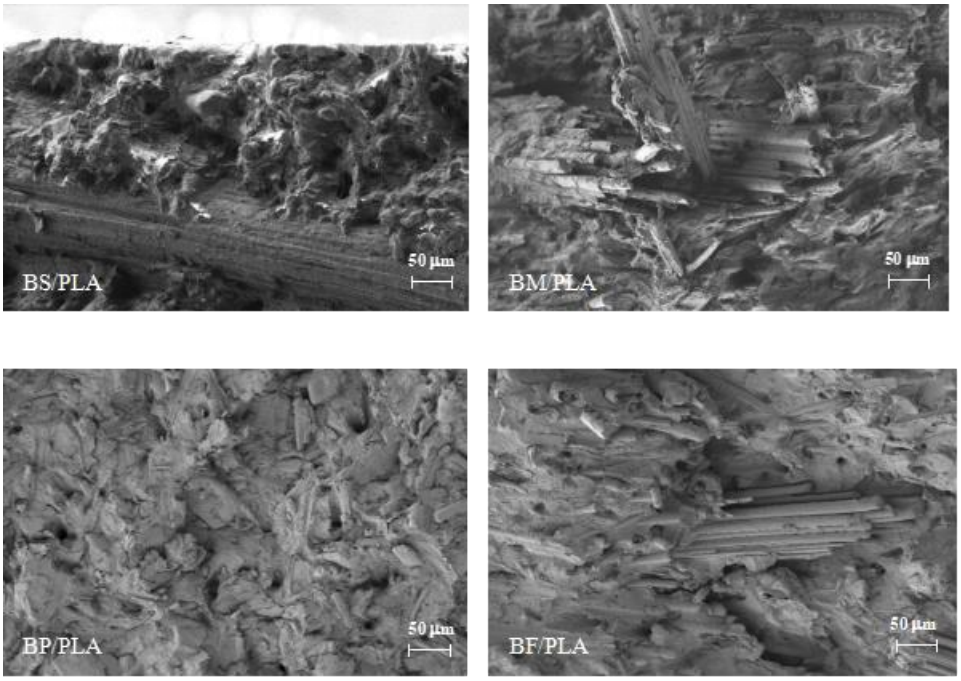

Fig. 5. SEM images of PLA-based bamboo-plastic composites with four morphologies of bamboo chips

Melting Index

In the processing of composite materials, the fluidity of resin has an important effect on the performance, yield, and energy consumption of the product. The melt flow index of the four kinds of bamboo–plastic composites is shown in Table 3.

Table 3. Melt Flow Indices of Composite Materials

Composite Material	Melting Mass (g)	Flow Time (s)	MFR $(g\cdot 10min^{-1})$
BS/PLA	*	*	*
BM/PLA	*	*	*
BP/PLA	*	*	*
BF/PLA	1.32	12.1	65.3

Note: * means "did not flow," such that no data were obtained.

It can be seen from Table 3 that among the four types of bamboo–plastic composite materials, only the BF/PLA composite material had relevant data for its melting fluidity. The BF/PLA composite had a flow time of 12.1 s, a melting mass of 1.32 g, and a melting index of 65.3 g/min. The other three forms of bamboo plastic composites all did not flow at 190 ℃, and so a melting index could not be calculated. This may be due to the high proportion of bamboo scraps in this experiment, which obstructs the flow of the PLA, and is consistent with the results of the pre-experiment of this study: when the mixture was mixed using a twin-screw extruder, the extrusion material could not be generated by flow. The BF/PLA composites mixed at a high temperature showed fluidity because the BF fragment was small, light in weight, and slender, and the BF contributed little resistance to flow in its mixture with the PLA (Wang 2019). Therefore, the BF/PLA composites exhibited good fluidity. At the same time, the flow effect of PLA promoted the uniform distribution of BF, so the BF dispersion was good according to the electron microscopy. However, the bamboo fragments of the other forms of bamboo plastic composites had a large individual area and overall quantity, which contributed a large resistance to PLA flow, so the composites did not exhibit fluidity.

CONCLUSIONS

1. The mechanical properties of the bamboo fiber/poly(lactic acid) (BF/PLA) composites were significantly better than those of the other bamboo–plastic composites, and the bending strength, tensile strength, elongation at break, and impact toughness were 35.9% and 24.5% higher than those of the conventional granular morphology (BM/PLA) composites, respectively. The bending and tensile strengths of the shaved morphology (BS/PLA) composites were only slightly lower than those of the conventional BM/PLA and bamboo powder (BP/PLA) bamboo–plastic composites. The bending modulus and tensile modulus of the BS/PLA composites were slightly higher than those of the BM/PLA and BP/PLA bamboo–plastic composites, but their impact toughness was relatively low.
2. The water absorption and wettability of the BS/PLA composite were similar to that of the BP/PLA composite; the BF/PLA composite exhibited strong hydrophilicity, with a water absorption of 3.5% and 10.83% at 24 h and 288 h, respectively, which was 83.2% and 45.8% higher than the BM/PLA composite, which had the lowest water absorption.
3. The BF/PLA composite was observed under SEM to have more fine fibers and more pores, which may be the reason for its higher water absorption. Only the BF/PLA composite had a melt flow index, which indicates that the melt matrix of the BF/PLA composite was less affected by BF fragment resistance; its melt index was 65.3 g/10 min.

FUTURE WORK

High proportion bamboo fiber/PLA composites were prepared by high-temperature mixing and hot-pressing. The addition of BF can improve the mechanical properties of PLA, and the composite has a certain fluidity. However, there are still some problems, such as lower water resistance and poorer impact performance. Future research will focus on the following aspects:

a) Adding compatibilizers to improve the interface bonding between two aspects, and to prepare the composite materials by different bamboo chips with a high-performance and low-cost.

b) At the same time, functional composite materials will be prepared by introducing elastic substances, surface modification, and other modification treatments.

c) In order to enrich the application range, the surface properties of composites will be characterized, such as roughness and wear resistance.

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