Preparation of Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV)-based Composites Using Poly(butylene adipate-co-terephthalate) (PBAT) and Bagasse

Yiqing Qin, Guichang Jiang,* Qi Xu, Peipei Li, Shihang Wei, and Jingyi Liu

Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) matrix composites were prepared by powder blending and a hot-pressing process with poly(butylene adipate-co-terephthalate) (PBAT) as a toughening agent and bagasse as a reinforcing agent. The effects of different contents of PBAT and bagasse on the mechanical properties, the thermal properties, and the section morphology of the composites were studied. By comparing the powder blending process with the conventional twin-screw melt extrusion blending process, it was found that the composites prepared by the powder blending process had better appearance and mechanical properties. With the increase of the PBAT content, the mechanical strength and modulus of the PHBV/PBAT composites decreased, but the flexibility increased. The addition of PBAT also caused the crystallinity of the composites to decrease and the thermal stability to increase. Based on the mass ratio of 60% PHBV and 40% PBAT, different contents of bagasse were added. It was found that the mechanical strengths of the composites were lower than that of the composites without bagasse, but the modulus was greatly improved. The crystallization properties of the composites improved, and the thermal stability decreased. This indicated that the addition of bagasse could reduce the material cost and improve the properties of PHBV.

INTRODUCTION

Due to the shortage of petroleum resources and the increasingly serious environmental problems, biodegradable polymers have attracted extensive attention in academia and industry. As a polyester family, polyhydroxyalkanoates (PHAs) are a kind of biomaterial that uses renewable natural resource starch as the main raw material and are fermented by a variety of microorganisms to obtain carbon sources as energy and stored in microorganisms (Bittmann et al. 2015). Polyhydroxyalkanoates can be completely biodegraded into carbon dioxide and water, and they are potential substitutes for petroleum-based polyester materials (Meereboer et al. 2020). Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) is the second-generation product of PHAs. Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) has attracted extensive attention because of its good biodegradability, biocompatibility, gas barrier properties, and high mechanical strength. At present, PHBV has been applied in the fields of biomedicine, packaging,
automotives, among others (Ibrahim et al. 2020; Zhao et al. 2021). The rigidity and gas barrier properties of PHBV are comparable to polypropylene (PP). However, PHBV material has drawbacks such as high cost, high brittleness, low impact resistance, and poor thermal stability (Javadi et al. 2010b). To improve the properties of PHBV, the most economical and effective method is to blend it with polymers with complementary properties.

Poly(butylene adipate-co-terephthalate) (PBAT) is a biodegradable material with excellent flexibility and high thermal stability, but its mechanical strength and degradation rate are low (Javadi et al. 2010a). Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) and PBAT are complementary in properties. Blending PHBV and PBAT can achieve complementary advantages and disadvantages and improve the comprehensive properties of the composites (Jin et al. 2018; Duangphet et al. 2019). Li et al. (2018) synthesized PEG-mb-PBAT multiblock copolymers by chain extension/coupling of PEG and PBAT prepolymers, which were used to plasticize and toughen poly(L-lactide) (PLLA) simultaneously. Among them, PEG segments could plasticize PLLA and improve the compatibility of PBAT microphase and PLLA matrix, while PBAT microphase could provide excellent toughness and ductility of the blends. Zhao et al. (2019) used PBAT as a good toughening agent for PLA. However, due to the poor compatibility of the two materials, they added polypropylene carbonate polyurethane (PPCU) as a compatibilizer for PBAT and PLA blends and obtained PLA/PBAT/PPCU ternary blends. The results showed that PBAT could improve the elongation at break of the composites, and PPCU could improve the tear strength of the composites. Xiao et al. (2019) used PBAT and ethylene-methyl acrylate-glycidyl methacrylate terpolymer (EGMA) as toughening and compatibilization agents of industrial hemp hurd (HH)/polylactic acid (PLA) biocomposites, respectively, to produce fused deposition model filaments. In summary, PBAT can also be used as a good toughening agent to improve the toughness of highly brittle PHBV materials.

Commonly, the strength and stiffness properties of any polymers or polymer composites can be improved with the addition of fillers with dimensions in the micron and nanometer ranges (Pal et al. 2020). Such addition, in principle, could offset the decrease of strength and modulus accompanied by the increase of toughness when PBAT is added to PHBV. Plant fibers are the most abundant natural fillers and have the advantages of low cost, low density, high modulus, good thermal stability, and renewability (Yu and Li 2014; Zhang et al. 2014). Plant fibers such as bagasse, corn stalk, and wheat straw, as agricultural wastes, have attracted increased attention in recent years due to their high yield and low utilization rate (Nagarajan et al. 2013b; Berthet et al. 2015; David et al. 2020). Bagasse is the main by-product of sucrose manufacturing. The global annual output of bagasse is as high as hundreds of millions of tons (Liu et al. 2020). At present, the utilization of bagasse is relatively low. A large amount of bagasse is incinerated for power generation or discarded, and only a small amount is used for papermaking or converted into biomass energy. This causes a great waste of resources and creates environmental pollution problems. Using bagasse as polymer reinforcement can improve the comprehensive utilization of bagasse, increase the added value of agricultural products, and reduce the burden of shortage of wood resources. The utilization of bagasse as a polymer reinforcement can also reduce the cost of composites while maintaining biodegradability and providing better mechanical properties. Therefore, the research of bagasse reinforced PHBV-based composites has high economic and social value.
In this work, a PHBV-based composites with excellent comprehensive properties were prepared by powder blending and a hot-pressing process with PBAT as a toughening material and bagasse as a reinforcing material. The mechanical properties, thermal properties, and section morphology of the PHBV/PBAT composites and PHBV/PBAT/bagasse composites were studied to explore the effects of different amounts of PBAT and bagasse on the composites. This work provides insight on the preparation of an economical and sustainable biocomposite material.

EXPERIMENTAL

Materials

The PHBV powder (ENMAT Y1010) was obtained from Ningbo Tianan Biologic Material Co. (Zhejiang, China). This commercial grade of PHBV contained some nucleating agents and antioxidants. The PBAT (Ecoflex C1200) was obtained from BASF (Ludwigshafen, Germany). This commercial grade of PBAT was granular and required liquid nitrogen freezing and grinding into powder. The bagasse (60-mesh) was obtained from fresh sugarcane. The sugarcane was peeled, juiced, and treated with hot water to remove water-soluble sugar. The obtained bagasse was dried in a drying oven at 80 °C for 24 h, pulverized by a high-speed pulverizer (Taisite Instrument Co., Ltd., Tianjin, China), and passed through a 60-mesh screen to obtain bagasse powder.

Preparation of the Composites

The bagasse was dried at 80 °C for 12 h, and the PHBV powder and PBAT powder were dried at 60 °C for 6 h. The dried raw materials were weighed according to a certain mass ratio, put into a high-speed mixer (Dongguan Benfan General Machinery Co., Guangdong, China), and blended at 1,500 revolutions per min for 5 min. Approximately 38 g of the blended raw materials were evenly distributed in a hot-pressing mold with dimensions of 200 mm × 150 mm × 1 mm. The material was pressed for 8 min at 178 °C and a pressure of 15 t. The blended materials were then taken out after the mold was cooled. The preparation process of the composite materials is shown in Fig. 1.

Fig. 1. The process flow chart for the preparation of the composite materials

This experiment was divided into two parts. Both the PHBV/PBAT composites and the PHBV/PBAT/bagasse composites with different mass ratios were studied. The bagasse was added to the composites based on the PHBV/PBAT mass ratio of 60/40, as this ratio
can provide sufficient flexibility while maintaining a certain mechanical strength of the material. The various formulation compositions of the prepared composites are shown in Table 1.

**Table 1. The Formulation Composition of the Prepared Composites**

<table>
<thead>
<tr>
<th>Sample Name</th>
<th>Abbreviation</th>
<th>Blend Matrix</th>
<th>Bagasse (wt%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PHBV</td>
<td>100/0</td>
<td>100</td>
<td>0</td>
</tr>
<tr>
<td>80%PHBV/20%PBAT</td>
<td>80/20</td>
<td>80</td>
<td>20</td>
</tr>
<tr>
<td>60%PHBV/40%PBAT</td>
<td>60/40</td>
<td>60</td>
<td>40</td>
</tr>
<tr>
<td>50%PHBV/50%PBAT</td>
<td>50/50</td>
<td>50</td>
<td>50</td>
</tr>
<tr>
<td>40%PHBV/60%PBAT</td>
<td>40/60</td>
<td>40</td>
<td>60</td>
</tr>
<tr>
<td>20%PHBV/80%PBAT</td>
<td>20/80</td>
<td>20</td>
<td>80</td>
</tr>
<tr>
<td>PBAT</td>
<td>0/100</td>
<td>0</td>
<td>100</td>
</tr>
<tr>
<td>90%[60%PHBV/40%PBAT]/10%Bagasse</td>
<td>54/36/10</td>
<td>60</td>
<td>40</td>
</tr>
<tr>
<td>80%[60%PHBV/40%PBAT]/20%Bagasse</td>
<td>48/32/20</td>
<td>60</td>
<td>40</td>
</tr>
<tr>
<td>70%[60%PHBV/40%PBAT]/30%Bagasse</td>
<td>42/28/30</td>
<td>60</td>
<td>40</td>
</tr>
<tr>
<td>60%[60%PHBV/40%PBAT]/40%Bagasse</td>
<td>36/24/40</td>
<td>60</td>
<td>40</td>
</tr>
<tr>
<td>50%[60%PHBV/40%PBAT]/50%Bagasse</td>
<td>30/20/50</td>
<td>60</td>
<td>40</td>
</tr>
</tbody>
</table>

**Mechanical Properties**

The tensile and flexural properties of the composites were tested by an Instron 3369 universal testing machine (Norwood, MA, USA) according to the GB/T 1040 (2006) and 9341 (2008) standards. At least five samples were measured for each sample and the average value was taken.

**Scanning Electron Microscopy (SEM)**

An SU1510 scanning electron microscope (Hitachi, Tokyo, Japan) was used to observe the fracture surface of the composites under the scanning voltage of 10 kV. The sample was brittle fractured by liquid nitrogen, pasted on the test platform with conductive adhesive to ensure that the fracture surface was flush with the platform, and then the surface was sprayed with gold.

**Differential Scanning Calorimetry (DSC)**

The DSC analysis of the composites was performed using a DSC 8000 (PerkinElmer, Waltham, MA, USA) under a constant nitrogen flow. The samples (approximately 5 mg to 10 mg) were subjected to two thermal cycles to observe the thermal behavior of the composites. In the first thermal cycle, the temperature was fixed from room temperature to 200 °C at 20 °C/min and then held isothermally at 200 °C for 3 min to help remove the hot processing history. Afterwards, the temperature was reduced from 200 °C to -60 °C at 10 °C/min. In a second heating, the samples were heated from -60 °C to 200 °C at 10 °C/min. The peaks of the cooling curve ($T_c$) and the second heating curve ($T_m$) were recorded. The crystallinity of the PHBV in the composites was calculated by Eq. 1,

$$X_{PHBV} = \frac{\Delta H_m}{W_{PHBV} \times \Delta H_0} \times 100\%$$

where $\Delta H_m$ is the melting enthalpy of the second heating curve (J/g), $W_{PHBV}$ is the weight fraction of PHBV in the composites, and $\Delta H_0$ is the melting enthalpy of 100% crystalline PHB (146 J/g).
Thermogravimetric Analysis (TGA)

The thermal stability and thermal decomposition temperature of the composites were measured by a TGA-Q50 (TA Instruments, New Castle, DE, USA). The samples (approximately 5 mg to 10 mg) were heated from room temperature to 600 °C at a rate of 10 °C/min. The whole process was carried out under a nitrogen atmosphere with a nitrogen flow rate of 50 mL/min.

RESULTS AND DISCUSSION

Mechanical Properties of the PHBV/PBAT Composites

The effects of different contents of PBAT on the mechanical properties of the PHBV/PBAT composites are shown in Fig. 2. The pure PHBV material had high mechanical strength and modulus values, but the elongation at break value was low, which is typical for a rigid material. The pure PBAT is a flexible material with extremely low mechanical strength and modulus properties, but the elongation at break was more than 900%. Therefore, the addition of PBAT gradually decreased the mechanical strength and modulus of the PHBV/PBAT composites, while it gradually increased the elongation at break properties. This trend can be seen in Figs. 2a-e, which show that the addition of PBAT can improve the flexibility of PHBV.

The tensile strength of the composites in Fig. 2a was an exception, which initially decreased with the addition of PBAT. The tensile strength reached its lowest when the PBAT content was 80%, after which it increased. This may be because when the PBAT content exceeded 80%, the PBAT occupied an absolute dominant position in the blend matrix rather than the PHBV. The PBAT changed from a dispersed phase to a continuous phase, and the intermolecular force of the composites increased, resulting in fracture under higher tensile force. Different from the tensile strength, the flexural strength of the material

Fig. 2. Effect of the PBAT content on the mechanical properties of the PHBV/PBAT composites. (a) Tensile properties, (b) elongation at break, (c) tensile stress-strain curves, (d) flexural properties, and (e) flexural stress-strain curves
is mainly affected by the degree of softness of the material. Therefore, the flexural strength
of the composites in Fig. 2d gradually decreased with the increase of PBAT content. The
pure PBAT material is very soft, so its flexural strength is extremely low.

As shown in Fig. 2b, powder blending did not significantly improve the elongation
at break values of the composites. When the PBAT content was 70%, the elongation at
break was still less than 10%, which was because the PBAT raw materials were too
dispersed in the mixed materials and could not connect and interact with each other. When
the PBAT content exceeded 80%, the PBAT formed a continuous phase and the elongation
at break was greatly improved, which was consistent with the changing characteristics of
the tensile strength. However, the addition of PBAT greatly improved the flexibility of the
composites, which can be intuitively observed from the flexural stress-strain curves in Fig.
2e. Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) is a brittle material that breaks before
yielding. With the increased PBAT content, the composites gradually transformed from
brittle fracture to flexible yielding, and the composites with the high PBAT content did not
break when bending. The mechanical properties analysis of the PHBV/PBAT composites
proved the complementarity of the two materials in mechanics, and composites with
different rigidity and toughness can be prepared by adjusting the blending ratio of PHBV
and PBAT.

Since the purchased PBAT raw material was granular, while PHBV and bagasse
were powdered, the PBAT particles were frozen and ground into powder, and the
composites were prepared by powder blending and hot pressing (Allaf et al. 2015; Zaidi
and Crosky 2019). At the same time, comparison materials were prepared by melt-blending
granular PBAT, powdered PHBV, and powdered bagasse with a twin-screw extruder and
then hot pressing. The mechanical tests and appearance comparison of the composites
prepared by the powder blending process in this experiment and the conventional twin-
screw melt extrusion blending process were shown in Table 2.

<table>
<thead>
<tr>
<th>Blending Process</th>
<th>Tensile Strength (MPa)</th>
<th>Tensile Modulus (MPa)</th>
<th>Flexural Strength (MPa)</th>
<th>Flexural Modulus (MPa)</th>
<th>Elongation at Break (%)</th>
<th>Appearance</th>
</tr>
</thead>
<tbody>
<tr>
<td>Melt extrusion blending 60%PHBV/40%PBAT</td>
<td>10.70</td>
<td>775.03</td>
<td>21.82</td>
<td>1007.94</td>
<td>35.09</td>
<td></td>
</tr>
<tr>
<td>Powder blending 60%PHBV/40%PBAT</td>
<td>14.95</td>
<td>986.91</td>
<td>30.84</td>
<td>1439.45</td>
<td>3.48</td>
<td></td>
</tr>
<tr>
<td>Melt extrusion blending 80%(60%PHBV/40%PBAT)/20%Bagasse</td>
<td>8.62</td>
<td>1023.04</td>
<td>18.43</td>
<td>1335.31</td>
<td>1.54</td>
<td></td>
</tr>
<tr>
<td>Powder blending 80%(60%PHBV/40%PBAT)/20%Bagasse</td>
<td>9.53</td>
<td>1141.36</td>
<td>20.85</td>
<td>1544.84</td>
<td>1.96</td>
<td></td>
</tr>
</tbody>
</table>

As shown in Table 2, the comparison of appearance showed that powder blending
made the raw materials evenly mixed and had a beautiful appearance, while the pellets and
powders in the melt extrusion blending process were difficult to be fully mixed evenly.
Besides, after adding bagasse, the bagasse undergoes gelatinization during melt extrusion
because of the prolonged high temperature environment. By comparing the mechanical parameters of the composites, it was found that powder blending was better than melt extrusion blending, except in regards to elongation at break (Zimmerling et al. 2021), because powder blending was more uniform than pellet melt blending. When bagasse fibers were added, the elongation at break of the composites prepared by melt extrusion blending process was greatly reduced due to the inherent rigidity of the fibers. At the same time, the powder blending process reduces one heat treatment compared with the melt extrusion blending process, thereby avoiding the molecular weight loss caused by multiple thermal processing of PHBV, and also avoiding the gelatinization problem caused by multiple thermal processing of fibers in the later stage. The prepared composites had better appearance and better mechanical properties. Therefore, powder blending process is a good choice for fiber and plastic blending materials.

SEM Analysis of the PHBV/PBAT Composites

Figure 3 shows the cross-sectional SEM micrographs of the PHBV/PBAT composites, where the interfacial compatibility of PHBV and PBAT materials is visible. As shown in Fig. 3a, the section of pure PHBV was very smooth, which also reflects the brittleness of the PHBV material, while the pure PBAT material shown in Fig. 3f was soft, with a rough and textured section (Quiles-Carrillo et al. 2018). There were no obvious defects in the cross section of the PHBV/PBAT composites and no obvious interface separation between the two materials, which proves that PHBV and PBAT have a certain compatibility and can partially melt with each other, which is also the reason why the mechanical properties of the composites can keep decreasing gradually with the increase of PBAT content.

![Fig. 3. SEM micrographs of the a) 100/0, b) 80/20, c) 60/40, d) 40/60, e) 20/80, and f) 0/100 PHBV/PBAT composites](image)

DSC Analysis of the PHBV/PBAT Composites

Figure 4 shows the DSC curves of the PHBV/PBAT composites at cooling and the second heating, while Table 3 shows the DSC data. The addition of PBAT reduced the crystallinity of the PHBV in the composites. With the increase of PBAT content, the crystallization temperature ($T_c$) and the melting temperature ($T_m$) of the PHBV/PBAT composites decreased initially before they increased, reaching the lowest at the ratio of
50/50. However, the $T_c$ and $T_m$ values of all the blends were lower than the pure PHBV and higher than the pure PBAT. Crystallization temperature ($T_c$) can be considered as an indirect measure of crystallization rate. Usually, a lower $T_c$ indicates slower crystallization. As shown in Fig. 4a, the increase of the PBAT content gradually reduced the peak intensity and broadened the crystallization peak of the composites, which indicated that the addition of PBAT would hinder the crystallization of PHBV, delay the growth rate of PHBV crystals, and result in slower crystallization and lower crystallinity.

The DSC heating curves of semi-crystalline polyesters often show a bimodal melting phenomenon, as shown in Fig. 4b, which is due to the melting recrystallization behavior of polyesters (Larsson et al. 2016). During the heating process, the imperfect crystallized part of PHBV melts first and can be partially transformed into a more perfect crystal structure, resulting in the first melting peak. The more perfect crystallized part is then melted at a higher temperature to obtain the second melting peak. After the first heating to eliminate the thermal history, the PHBV material in the second heating curve has only one melting peak, while the composites with high PBAT content still show bimodal melting, which may be caused by a large amount of PBAT hindering the crystallization of PHBV. Therefore, PBAT should be added at an appropriate content.

![Fig. 4. DSC curves of the PHBV/PBAT composites. (a) Cooling curves and (b) second heating curves](image)

**Table 3. DSC Data of Cooling Cycle and Second Heating Cycle of PHBV/PBAT Composites**

<table>
<thead>
<tr>
<th>PHBV/PBAT Content (%)</th>
<th>$T_c$ (°C)</th>
<th>$T_m$ (°C)</th>
<th>$\Delta H_m$ (J/g)</th>
<th>$X_{PHBV}$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>100/0</td>
<td>119.07</td>
<td>171.82</td>
<td>92.45</td>
<td>63.32</td>
</tr>
<tr>
<td>80/20</td>
<td>98.44</td>
<td>169.66</td>
<td>62.21</td>
<td>53.26</td>
</tr>
<tr>
<td>60/40</td>
<td>93.73</td>
<td>168.61</td>
<td>44.91</td>
<td>51.27</td>
</tr>
<tr>
<td>50/50</td>
<td>86.84</td>
<td>167.29</td>
<td>37.09</td>
<td>50.81</td>
</tr>
<tr>
<td>40/60</td>
<td>90.24</td>
<td>168.09</td>
<td>27.77</td>
<td>47.55</td>
</tr>
<tr>
<td>20/80</td>
<td>100.82</td>
<td>168.89</td>
<td>12.71</td>
<td>43.53</td>
</tr>
<tr>
<td>0/100</td>
<td>79.66</td>
<td>128.15</td>
<td>10.21</td>
<td>-</td>
</tr>
</tbody>
</table>
TGA Analysis of the PHBV/PBAT Composites

The effect of different contents of PBAT on the thermal stability of PHBV/PBAT composites was further explored by TGA. As shown by the TG curves in Fig. 5a, the thermal decomposition temperature range of PHBV was very narrow. It began to decompose at approximately 260 °C and was completely decomposed at 300 °C. The thermal decomposition rate was extremely fast, which led to a narrow processing range of PHBV. However, the PBAT started to decompose slowly from 350 °C and was basically completed at approximately 440 °C with some carbon residues retained. The thermal decomposition of PHBV/PBAT composites consisted of two separate decomposition steps accompanied by two separate decomposition peaks (Bittmann et al. 2015). As shown in Fig. 5b, with the increase of the PBAT content, the mass loss of the PHBV/PBAT composites gradually shifted to higher temperature and the initial decomposition temperature increased. This shows that the addition of PBAT can improve the thermal stability of the composites, which is beneficial to reduce the thermal decomposition of the materials and expand the processing temperature range.

![TG and DTG curves of PHBV/PBAT composites](image)

**Fig. 5.** (a) TG and (b) DTG curves of PHBV/PBAT composites

Mechanical Properties of the PHBV/PBAT/Bagasse Composites

Figure 6 shows the effect of the bagasse content on the mechanical properties of the PHBV/PBAT/bagasse composites. When the bagasse content was 10%, the tensile and flexural properties of the composites decreased compared to those without the bagasse. This was ascribed to the fact that the addition of the bagasse destroyed the intermolecular forces of the PHBV/PBAT blend matrix. In addition, the interfacial adhesion between the hydrophilic bagasse fibers and the hydrophobic polymer matrix is poor, and effective interfacial stress transfer cannot be carried out when resisting external forces (Zhang et al. 2014). As the bagasse content increased, the strength and modulus of the composites increased gradually, which is attributed to the force between the fibers and the interaction between the fibers and the matrix. The higher the content of bagasse, the greater the strength and modulus of the composites. In general, compared with the composites without the bagasse, the mechanical strength of the composites that contained bagasse decreased, but the modulus increased. When the bagasse content was 50%, the tensile modulus and flexural modulus of the composites increased by 110.21% and 106.75%, respectively. The modulus is the ratio of stress to strain. The greater the modulus, the higher the stiffness and deformation resistance of the material (Wu et al. 2019). The improved modulus of
composites may be attributed to the addition of fibers to the polymer matrix, which hinders the free movement of polymer molecules and restricts the migration of molecular chains, thereby increasing the stiffness of the composites (Nagarajan et al. 2013a). Therefore, bagasse can be used as a good reinforcing material for the PHBV/PBAT blend matrix.

**SEM Analysis of the PHBV/PBAT/Bagasse Composites**

The SEM micrographs of the PHBV/PBAT/bagasse composites are shown in Fig. 7. As shown in Fig. 7a, the composite material without bagasse had a smooth cross-section and no obvious defects, and the PHBV and PBAT materials were closely combined. The section of the composite added with bagasse became rough, the interfacial compatibility between bagasse fiber and plastic matrix was poor, the interface bonding force was weak, and the fiber could not be well coated by the matrix. Therefore, fiber pullout, voids, and gaps between the fiber and the matrix were observed. This can explain the phenomenon of the mechanical strength reduction. In the later stage, bagasse can be
modified to improve the interfacial compatibility between bagasse and plastic matrix and improve the comprehensive properties of the composites.

**DSC Analysis of the PHBV/PBAT/Bagasse Composites**

Figure 8 shows the DSC curves of the PHBV/PBAT/bagasse composites, and Table 4 shows the DSC data of the composites. The addition of the bagasse was able to shift the \( T_c \) of the composites to higher temperatures, which indicates that bagasse can improve the crystallization properties of the composites. Bagasse fibers act as crystallites to form nucleation sites, allowing the composites to crystallize at higher \( T_c \) values (Zaidi and Crosky 2019). As the bagasse content increased, the \( T_m \) of the composites first increased and then decreased. The decrease in the \( T_m \) value was likely because the hydroxyl group on the fibers caused the hydrolysis of the PHBV at high temperature (Srithep et al. 2013). The crystallinity of the PHBV in the composites showed an increasing trend, but suddenly decreased when the bagasse content was 50%. This indicates that an appropriate amount of bagasse acts as a nucleating agent in the composites, but excessive bagasse restricts the movement of molecular chains in the polymer matrix, which hinders the crystallization of the PHBV.

![DSC curves](image)

**Fig. 8.** DSC curves of the (a) cooling curves and (b) second heating curves of PHBV/PBAT/bagasse composites

**Table 4.** DSC Data of the Cooling Cycle and Second Heating Cycle of PHBV/PBAT/Bagasse Composites

<table>
<thead>
<tr>
<th>Bagasse Content (%)</th>
<th>( T_c ) (°C)</th>
<th>( T_m ) (°C)</th>
<th>( \Delta H_m ) (J/g)</th>
<th>( X_{PHBV} ) (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>93.73</td>
<td>168.61</td>
<td>44.91</td>
<td>51.27</td>
</tr>
<tr>
<td>10</td>
<td>95.47</td>
<td>170.30</td>
<td>40.79</td>
<td>51.74</td>
</tr>
<tr>
<td>20</td>
<td>97.92</td>
<td>170.36</td>
<td>36.58</td>
<td>52.20</td>
</tr>
<tr>
<td>30</td>
<td>96.25</td>
<td>170.56</td>
<td>32.35</td>
<td>52.76</td>
</tr>
<tr>
<td>40</td>
<td>97.09</td>
<td>169.08</td>
<td>28.85</td>
<td>54.89</td>
</tr>
<tr>
<td>50</td>
<td>97.54</td>
<td>168.12</td>
<td>21.83</td>
<td>49.84</td>
</tr>
</tbody>
</table>
TGA Analysis of the PHBV/PBAT/Bagasse Composites

The TGA curves of the PHBV/PBAT/bagasse composites are shown in Fig. 9. After the bagasse was added, the composite material became a three-step thermal decomposition process, corresponding to the three thermal decomposition peaks of the PHBV, bagasse, and PBAT, respectively. The weight of the composites at 200 °C decreased slightly with the increase of the bagasse content, which was due to the evaporation of absorbed water in the bagasse. After adding bagasse, the weight residual of the composites increased, which was mainly the charcoal residue after the thermal decomposition of the cellulose, hemicellulose, and lignin in the bagasse. As shown in Fig. 9b, as the bagasse content increased, the thermal decomposition peak of the composites shifted to the left and the initial thermal decomposition temperature gradually decreased. This indicated that the addition of bagasse reduced the thermal stability of the composites. This may be because the thermal decomposition temperature of hemicellulose in bagasse is lower than that of PHBV. The exposed fibers decompose first and cause mutual thermal decomposition between the fibers and the plastic matrix (Guo et al. 2019). This result is consistent with those of other related studies (Yu and Li 2014; Li et al. 2020).

![Fig. 9. (a) TG and (b) DTG curves of the PHBV/PBAT/bagasse composites](https://example.com/fig9.png)

CONCLUSIONS

1. In this study, poly(3-hydroxybutyrate-co-3-hydroxyvalerate)/poly(butylene adipate-co-terephthalate) (PHBV/PBAT) composites and PHBV/PBAT/bagasse composites were prepared by powder blending and hot pressing process, in which PBAT was used as a toughening agent for PHBV, and bagasse was used as a reinforcing agent for PHBV. The powder blending process was compared with the conventional twin-screw melt extrusion blending process, and it was found that the composites prepared by the powder blending process had better appearance and mechanical properties.

2. As the PBAT content increased, the mechanical strength and modulus properties of the PHBV/PBAT composites decreased, but the elongation at break properties improved, and the material gradually changed from a rigid material to a flexible material. There were no obvious defects in the cross section of the composites. The addition of PBAT reduced the crystallinity and improved the thermal stability of the composites. These results showed that PBAT is a good toughening material.
3. As the bagasse content increased, the tensile and flexural properties of the PHBV/PBAT/bagasse composites gradually increased, and the modulus and stiffness increased greatly, but the mechanical strength was lower than that of the composites without bagasse. The cross section of the composites became rough, the crystalline properties improved, and the thermal stability decreased. These results indicated that although bagasse is an agricultural waste with low mechanical properties, it can still be used as a natural reinforcing material for polymers, which plays a positive role in improving the application of bagasse.

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