

Research Article

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Effect of talc and diatomite on compatible, morphological, and mechanical behavior of PLA/PBAT blends

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Abstract: Biodegradable nanocomposites were prepared by melt blending biodegradable poly(lactic acid) (PLA) and poly(butylene adipate-co-butylene terephthalate) (PBAT) (70/30, w/w) with diatomite or talc (1–7%). From the SEM test, the particles were transported to the interface of two phases, which acted as an interface modifier to strengthen the interfacial adhesion between PLA and PBAT. Talc and diatomite acted as nucleating agents to improve the crystallization of PBAT in the blends by DSC analysis. Moreover, adding the particles improved the tensile and impact toughness of the blends. The elongation at break with 5% talc was 78% (vs ~21%) and the impact strength was 15 kJ/m² (vs ~6.5 kJ/m²). The rheological measurement revealed that the talc and diatomite reduced the viscosity of the blends. The results showed a good possibility of using talc- and diatomite-filled PLA/PBAT blends with high toughness for green-packaging and bio-membranes application.

Keywords: PLA/PBAT blend, talc, diatomite, compatibilization, properties

1 Introduction

So far, factories around the world have produced more than 9 billion tons of plastic, but only 9% of them are recycled, and the rest of them are thrown into landfills or the natural environment. This not only causes the waste of resources but also pollutes the environment (1–3). Biodegradable materials effectively alleviate the problem of plastic pollution and reduce the consumption of fossil energy. Poly(lactide acid) (PLA) is one of the promising bio-based and biodegradable polyesters because it owns excellent mechanical strength, sustainability, and good processing temperature (4,5). Lactic acid is the monomer for the synthesis of PLA, which was fermented from renewable plants such as corn (6–8). The tensile strength of PLA is over 70 MPa, and the melting temperature can reach 175°C. However, poor crystallizability, apparent brittleness, and low heat distortion temperature have limited the further application of PLA. The elongation at break of PLA is about ~4%, and the impact strength is less than 2.5 kJ/m². To improve the application of PLA, many methods are used to improve the properties. Polymer blending and chemical copolymerization were the most commonly used methods. Polymer blending is the most effective and economic way to improve the toughness of PLA (9). Poly(butylene adipate-co-terephthalate) (PBAT) is a copolymer based on terephthalic acid, adipic acid, and butanediol. It has good flexibility, such as high elongation at break, high impact strength, and it is fully biodegradable. PBAT is a promising blending material for PLA to improve the mechanical properties (10–12).

However, its compatibility is poor because of the weak interactions and interfacial adhesion between PLA and PBAT. The blending materials show phase separation structure on the microstructure, and neither present high strength nor toughness (13,14). To obtain composite materials with excellent properties, a good compatibilizer

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should be added to the blends. Premade copolymers (15), reactive polymers (16–18), and reactive low molecular weight chemicals (19) are considered as the most common compatibilizer in industrial production and scientific research. Adding nanoparticles to an immiscible blend could effectively improve the compatibility of PLA and PBAT (20). Particles are usually transported to the interface during melt processing, which acts as an interface modifier to enhance the interfacial adhesion between the two immiscible blends. Particles effectively prevent the coalescence of the dispersed phase. Also, the blends combine the advantages of polyester and particles (21). Moreover, particles are cheaper than polyesters and have more application value in industrial production. Therefore, it is a general method to prepare nanocomposites with good physical properties by adding particles to immiscible blends (22).

Numerous works had shown that the dispersion of particles can improve the properties of the immiscible blend (23–25). Moustafa et al. (24) showed that organo-clay modification by toxicity-free expanded rosin (ROC) or stearic acid (SOC) enhanced the biocompatibility of PLA/PBAT blends. ROC obviously enhanced the viscoelastic and tensile properties of the blends, and the nanocomposites showed strong antimicrobial activities. Urquijo et al. (26) found that carbon nanotubes (CNT) improved the impact strength of the PLA/PBAT (60/40) blend by 155% and increased the conductivity. Guo et al. (27) studied that montmorillonite clay (MMT) improved the compatibility of PLA and PBAT. MMT improved the impact strength of the blends. But more tubes weakened the interface adhesion because MMT agglomerated into bundles positioning parallel to the interface and blocked the chain entanglements. Thus, the dispersion of particles can effectively improve the compatibility of PLA and PBAT (28,29). However, more details should be researched on the crystallization, morphology, and properties of the blends.

In this research, we focus on the biodegradable PLA/PBAT blend with talc and diatomite particles. Talc is magnesium silicate. Talc crystals are in the form of pseudo-hexagons or rhombuses. The main component of diatomite is silicon dioxide, which is a kind of siliceous sedimentary rock with a porous structure. The composition and structure of the particle have a significant impact on the performance of the blends. Studies have shown that talc and diatomite are common and economical methods to promote crystallization and improve mechanical properties (30). Moreover, the addition of particles does not change the degradation performance

of biodegradable materials (31). Therefore, we mainly studied the influence of different particles on the compatibility, morphology, and performance of PLA and PBAT blends. The morphology as well as thermal and rheological properties of the blends was tested by DSC, SEM, and rheometer. Mechanical properties were obtained by tensile and impact tests.

2 Experimental methods

2.1 Materials

PLA (Grade 4032D) was obtained from Nature Works; PBAT (EcoWord FS0330) was obtained from JinHui ZhaoLong Co. Ltd; talc (~20 μm), and diatomite (~20 μm) was purchased from Liaoning Haiyan Biological Technology Co. Ltd.

2.2 Blends preparation

Before melt-blending, PLA, PBAT, talc, and diatomite were dried in a vacuum oven at 60°C for 12 h. Thermo-Scientific Process 11 extruder was used for prepared PLA/PBAT/particle blends (70/30/*x*, w/w/w) by melt-blending. The blend material is named particle-*y*% (1–7%) according to the different content of the particle. The screw diameter is 11 mm and the L/D is 40. The melt temperature was 185°C, 185°C, 190°C, 190°C, 190°C, 190°C, and 185°C, and the screw speed was 200 rpm.

2.3 Characterization

The thermal properties of blends were tested using a differential scanning calorimetry (DSC) instrument (Mettler-Toledo DSC1). A heating/cooling/heating procedure under nitrogen flow was used to study the thermal behavior, and the temperature varied from –70°C to 200°C. The first heating procedure removed the heating history, and the cooling and second heating procedures were investigated at a rate of 10°C/min. The degree of crystallinity (X_c) of PLA or PBAT in the blends was calculated using the following equation (Eq. 1):

$$X_c(\%) = 100 \times \frac{\Delta H_{c,m}}{f \times \Delta H_{c,m}^0}, \quad (1)$$

where $\Delta H_{c,m}$ is the crystallization/melting enthalpy of polyester and $\Delta H_{c,m}^0$ is the crystallization/melting enthalpy for a perfect crystalline polyester, which was 93 J/g for PLA and 114 J/g for PBAT, and f is the weight fraction of the polyester in the blends.

Thermogravimetric analysis was performed by TA Q50 instrument under flowing nitrogen. Approximately 5 mg of samples was heated from 50°C to 700°C, and the heating rate was 20°C/min.

The morphologies of extruded samples were measured using a Hitachi S-4800 SEM. The extruded samples were fractured in liquid nitrogen before using and sputter-coated with gold.

The tensile tests were measured on an Instron-5699 instrument according to ASTM D638 at room temperature, and the stretching rate was 10 mm/min. Notched impact strength was tested on a Wuzhong Material Tester XJ-300A according to ISO 179-1:2000. Five parallel samples were tested to diminish the test error. All samples were injected and molded using Thermo-Scientific Pro at 190°C under 700 bar.

Melt-rheological behaviors were measured using a HAKKE rotational rheometer RS6000, whose plate diameter is 25 mm and the gap is less than 1 mm. The frequency-dependency was monitored at 190°C, 5% strain under nitrogen flow to avoid thermal degradation.

3 Results and discussion

3.1 Morphological properties of the blends with particles

Figure 1 shows the morphological properties of PLA/PBAT blends with and without particles. The solubility parameter of PLA was $10.1 \text{ (cal/cm}^3)^{1/2}$, and PBAT was $10.7 \text{ (cal/cm}^3)^{1/2}$ according to the Hildebrand equation. Moreover, due to different structures of PLA and PBAT, the microstructure of the blend showed the obvious phase separation phenomenon. The PLA/PBAT blends without particle showed typical “sea-island” structures, where PBAT dispersed in the PLA resin with a wider size distribution, indicating low interfacial adhesion between PLA and PBAT. With the addition of the talc and diatomite, the particle size of PBAT is smaller and the interface phase between the two phases is not obvious. With the increase in the particle content from 1% to 5%, the size of the dispersed phase becomes smaller and smaller because more particles improved the compatibility of PLA and PBAT (32,33). However, with more talc or diatomite, excessive particles cannot improve the compatibility of PLA and PBAT, because in the process of blending, too much powder is easy to aggregate and

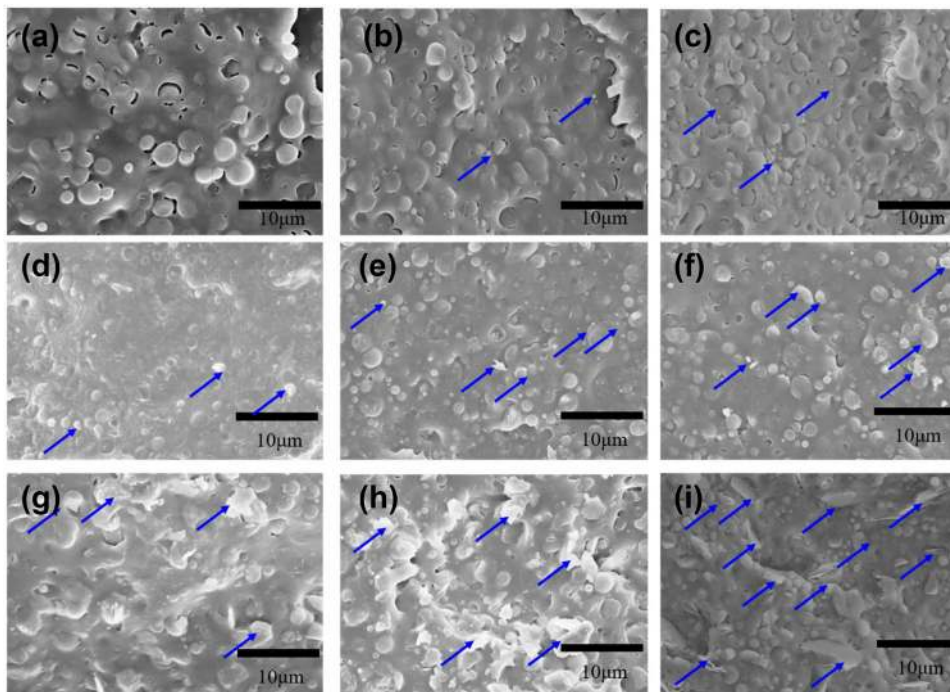


Figure 1: SEM micrographs of blends containing different amounts of particles: (a) PLA/PBAT 70/30, (b) diatomite-1%, (c) talc-1%, (d) diatomite-3%, (e) diatomite-5%, (f) diatomite-7%, (g) talc-3%, (h) talc-5%, (i) talc-7%.

cannot be effectively dispersed into the resin. Thus, as shown in Figure 1e and h, the diatomite and talc were aggregated to form nanosheets in the blend, which cannot play the role of compatibilization. Compared with the talc in the blends, diatomite is more effective in preventing PBAT from coalescence, and the size of the dispersed phase is smaller than the blends with talc. As shown in Figure 1d, the morphology of the blend with 3% diatomite was smooth and the size of the dispersed phase was small. Talc was easier to clump together than diatomite. The morphology of the blend with talc was rough and a lot of talcs gathered on the surface.

3.2 DSC analysis of the blends

DSC was used to research the thermal properties of PLA/PBAT blend with or without particles. The DSC curves are shown in Figure 2, the data calculated from these curves are presented in Table 1. From the second heating and cooling ramp, the T_g of PLA was around 61°C, and PBAT was around -30°C. No significant shift of T_g is observed in PLA/PBAT blend. However, with the addition of talc or diatomite from 1% to 5%, the T_g of PLA decreased because the talc or diatomite improved the compatibility of PLA and PBAT. The glass transition temperature of PLA was closer to PBAT. On the contrary, 7% particle could not continue to effectively improve the compatibility of PLA and PBAT. Instead, the glass transition temperature of PLA tends to increase. Also, as shown in Figure 1, the size of PBAT phase in the

blend with 7% talc and diatomite did not continue to decrease (34).

The crystallization temperature often reflected the crystalline phase properties. The lowering of T_c often reflected nucleation or the ease of chain packing. Following the heating ramp, the T_c of pristine PLA was about 169°C and the crystallinity was 3.6%. The crystallinity of the blend with 1% diatomite was 3.9%, and the cold crystallization temperature of the blend was 117.31°C. When the diatomite content was 5%, the crystallinity of the blend was 5.0% and the cold crystallization temperature was 105.47°C. Diatomite improved the crystallinity of PLA and acted as a nucleating agent. However, cold crystallization vanished in the blends with talc and double peaks of the melting endotherm in the blend indicated partial melting, recrystallization, and remelting of crystal in the blend. According to previous reports, talc acted as a nucleating agent while PBAT dispersed phase retarded for PLA to crystallize (30).

Following the cooling ramp, the crystallization temperature of PBAT was about 74°C, and the crystallinity of PBAT was 11.6%. In accordance with the heating process, diatomite has less effect on the crystallization of PLA in the blends. The crystallization temperature of PLA had no obvious change. However, talc affected the crystallinity and crystallization temperature. Talc hindered the crystallization of PLA but increased the crystallinity of PBAT. When 1% talc was added to the blends, the crystallization temperature of PBAT increased to 108°C and the crystallinity was 45.6%. The phenomenon showed that talc was more likely to move to the PBAT phase and played the role of nucleating agent to accelerate the crystallization of PBAT. The content of talc

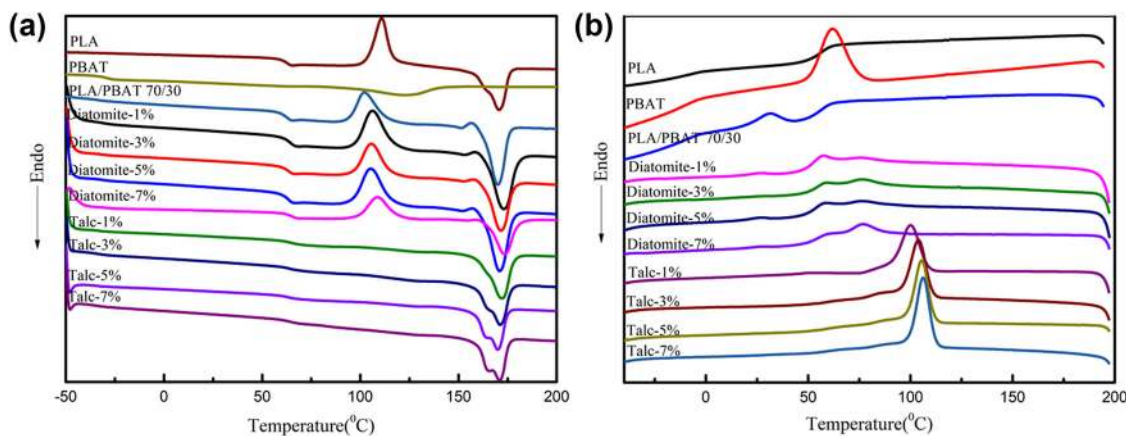


Figure 2: DSC curves of the PLA/PBAT blends: (a) blends second heating ramp, (b) blends cooling ramp.

Table 1: Thermal properties of the blends tested from DSC experiments

Samples	Heating ramp						Cooling ramp		
	T_g (PLA) (°C)	T_{cc} (°C)	T_m (°C)	ΔH_{cc} (J/g)	ΔH_m (J/g)	X_c (PLA) (%)	T_c (°C)	ΔH_c (J/g)	X_c (PBAT) (%)
PLA/PBAT	61	103	170	17.4	23.3	9.0	—	—	—
Diatomite-1%	64	117	173	16.2	18.8	3.9	—	—	—
Diatomite-3%	62	115	172	14.9	18.8	6.0	—	—	—
Diatomite-5%	62	105	171	16.8	20.1	5.0	—	—	—
Diatomite-7%	64	109	173	10.4	13.0	4.0	—	—	—
Talc-1%	67	—	172	—	22.1	—	108	15.6	45.6
Talc-3%	66	—	171	—	21.3	—	104	17.9	52.3
Talc-5%	63	—	170	—	22.3	—	105	20.8	60.8
Talc-7%	64	—	171	—	23.3	—	106	22.0	64.3
PLA	61	108	170	24.3	27.7	3.6	—	—	—
PBAT	-29	—	123	—	9.0	—	75	13.2	11.6

—: not detected.

increased leading to the continuous increase in the crystallinity of PBAT.

3.3 TGA analysis of the blends

The influence of talc and diatomite on the thermal properties of PLA/PBAT blends was evaluated using TGA. Figure 3a showed that the TGA thermograms of the blends with talc and diatomite incorporation ranging from 1% to 7%. Also, DTG curves were illustrated as a function of temperature in Figure 3b, all the blends showed two thermal degradation steps because of the two different thermal degradation temperatures of the PLA and PBAT. The initial decomposition temperature of the PLA/PBAT blends with 30% PBAT was 350.8°C, but the addition of talc and diatomite reduced the initial decomposition temperature. Çoban *et al.* (35) also studied

that volcanic particles lead to the decreased thermal stability of PLA/PBAT. The nucleation of the particles and the crystallinity were the main factors affecting the decomposition temperature of the blend. The initial decomposition temperature of the blend with 1% diatomite was 306°C, and the initial decomposition temperature of the blend with 5% diatomite was 320°C. The thermal stability of the blend had a significant increase. The initial decomposition temperature of the blend with 1% talc was 336°C and that with 5% talc was 339°C. The blends with particles had a higher thermal decomposition temperature.

3.4 Rheological analysis

Dynamic rheological tests were widely used to learn about the viscosity, phase morphology, and interfacial actions in the blends. To research the microstructure

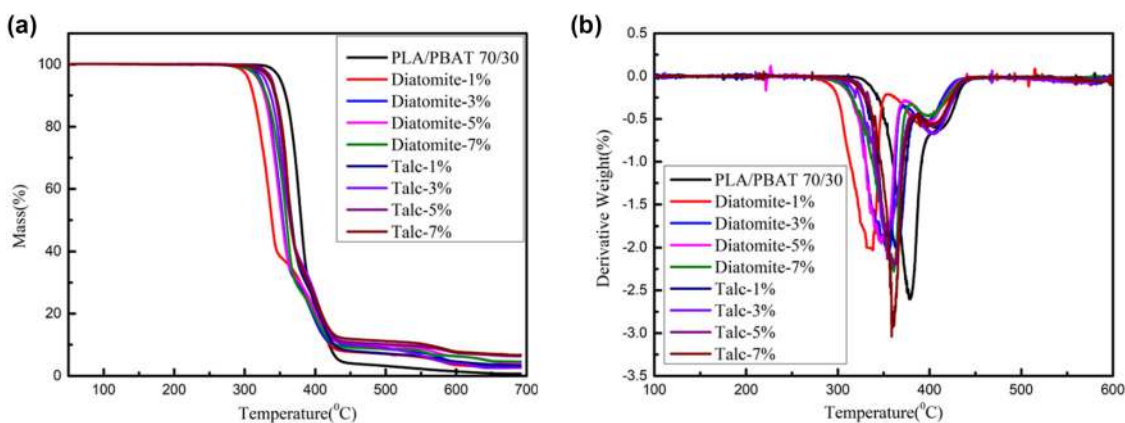


Figure 3: (a) TGA curves and (b) DTG curves of the samples under nitrogen flow.

and process properties of the blends, the elastic modulus G' , loss modulus G'' , and complex viscosity η^* versus frequency were evaluated. The effect of particles on G' , G'' , and η^* is shown in Figure 4, in the previous study, the G' , G'' , and η^* of PLA was higher than PBAT because of the different structure of PLA and PBAT. The blends without particles showed the rheological properties of the two components. The compatibility of PLA and PBAT was improved by a small number of particles. Thus, the G' , G'' , and η^* decreased with the addition of talc and diatomite. With the addition of more talc and diatomite, the chains were tightly packed together by talc and diatomite, which may cause higher modulus and viscosity. The lower viscosity was beneficial to the processing of the blends (36,37). Compared to the blend with talc, diatomite more effectively reduced the viscosity and modulus of the blends because diatomite was more effective in improving the compatibility of PLA and PBAT. The G' , G'' , and η^* of the blend with diatomite was close to that of PBAT. It is well known that the cole–cole plot is an effective way to characterize the different structures of the blends. As shown in Figure 4d, there are two different

relaxation mechanisms for immiscibility blends. Thus, the cole–cole curves usually exhibited two arcs for the immiscibility blends, the left one was attributed to the relaxation of polyester chains, and the right one showed the droplet relaxation. For the PLA/PBAT blends, the arc on the right side reflected the “sea–island” morphological structure. The addition of talc and diatomite improved the compatibility of PBAT and PLA and decreased the viscosity of the blends (38). The cole–cole plot reflected the complex rheological behavior, indicating that the droplet relaxation was incomplete, and the chains relaxation was influenced by the talc and diatomite.

3.5 Mechanical properties of the blends

The typical stress–strain curves of PLA/PBAT blends are shown in Figure 5a. The tensile strength of PLA/PBAT blend with 30% of PBAT was ~ 58.5 MPa, and the elongation at break was $\sim 25\%$, and the tensile modulus was 1.6 GPa without any particle. Because of high interfacial

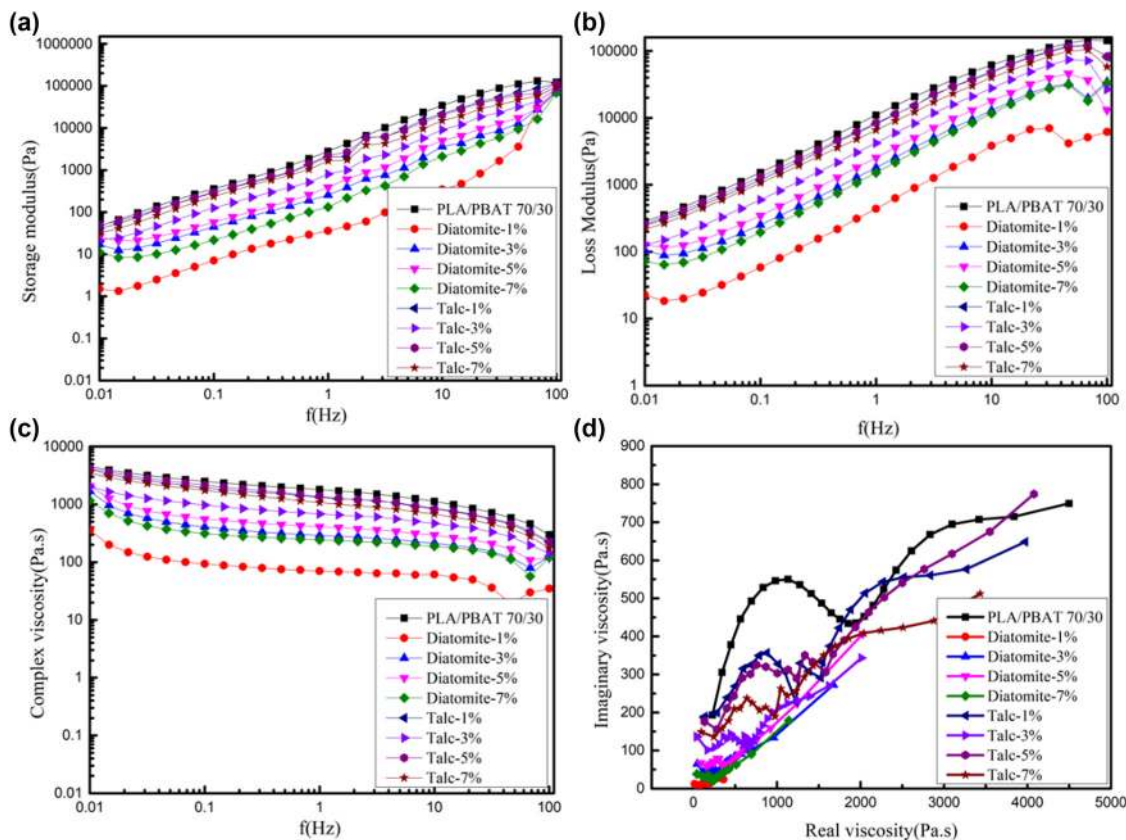


Figure 4: Effect of diatomite and talc loadings on the rheological behaviors of the blends: (a) storage modulus, (b) loss modulus, (c) complex viscosity, and (d) cole–cole plot.

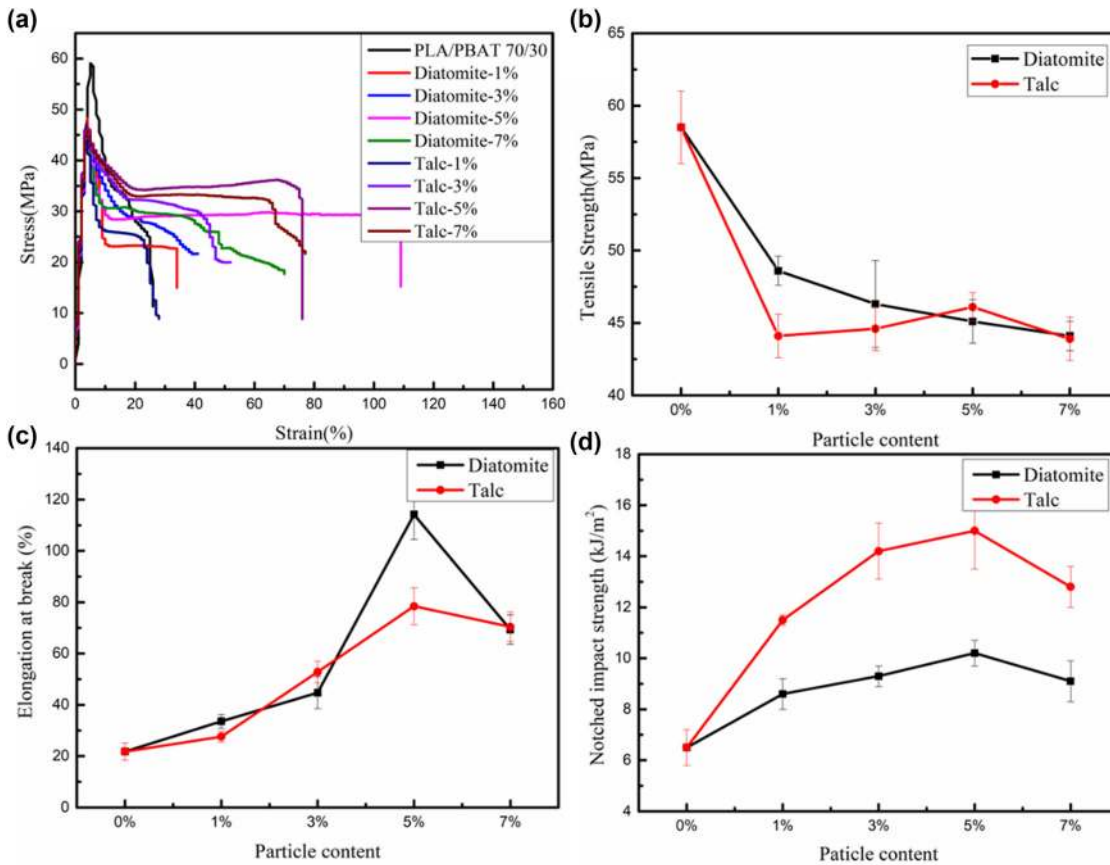


Figure 5: (a) Stress–strain curves, (b) tensile strength, (c) elongation at break, and (d) notched impact strength of the blends with talc and diatomite.

tension and poor compatibility between PLA and PBAT, brittle fracture, skin–core structures, and wire drawing phenomenon occurred during the drawing process (33). Therefore, we specify to stop the test when the sample breaks or the tensile strength is less than 30% of the max tensile strength.

The tensile strength of the PLA/PBAT blends decreased significantly in the diatomite and talc with the unfilled blends. The tensile strength of the blend with 5% diatomite decreased to 44 MPa, and 5% talc decreased to 43 MPa. Also, as given in Table 2, Young’s modulus of blends with 5% diatomite and talc decreased to 1.5 and 1.55 GPa, respectively. It is sure that a lower degree of crystallinity and the poor dispersion of the talc and diatomite in the blends should reduce the tensile strength. Moreover, the morphological change caused by the diatomite and talc was also considered as another important reason. PLA has high tensile strength and modulus, and the poor interfacial adhesion between PLA and PBAT resulted in the PLA characteristic when the blends break. The presence of the talc and diatomite

decreased the interfacial tension of PLA and PBAT. The SEM micrograph showed that talc and diatomite being a good compatibilizer changed the structure of the dispersed phase. Talc and diatomite decreased the size of the dispersed phase and decreased the interfacial tension of PLA and PBAT. Thus, compared with pure blends, the mechanical properties of the blends with particles were significantly enhanced, especially the elongation at break. Diatomite more effectively improved the mechanical properties of the blends. Young’s modulus of PLA was higher than PBAT. The Young’s modulus of the blends was close to that of PBAT because the particle improved the compatibility of PLA and PBAT (39).

Regarding the ductility of the PLA/PBAT blends, the high elongation ability of the PLA/PBAT blends is shown in the diatomite and talc components. In our previous work, incompatible polymers exhibited very low elongation at break, because of their high interfacial tension and were prone to interfacial detachment during stretching. A small amount of diatomite or talc acted as a good compatibilizer to increase the interfacial adhesion. The elongation

Table 2: The mechanical properties and thermal stabilities of the blends

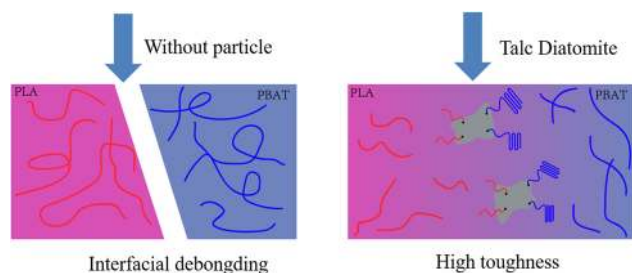
Samples	Tensile			Impact	TGA	
	Yield strength (MPa)	Elongation at break (%)	Young's modulus (MPa)		Notched impact strength (kJ m^{-2})	T_d 5% ($^{\circ}\text{C}$)
PLA/PBAT	58.5 ± 2.5	21.8 ± 3.4	1667 ± 10.5	6.5 ± 0.7	350.8	379.2
Diatomite-1%	48.6 ± 1.0	33.6 ± 2.7	1532.6 ± 50.5	8.6 ± 0.6	306.1	334.9
Diatomite-3%	46.3 ± 3.0	44.7 ± 6.2	1482.2 ± 40.4	9.3 ± 0.4	322.5	353.2
Diatomite-5%	46.0 ± 1.5	114.2 ± 9.8	1505.1 ± 70.0	10.2 ± 0.5	319.7	347.4
Diatomite-7%	45.1 ± 1.0	69.3 ± 5.7	1514.0 ± 80.0	9.1 ± 0.8	320.9	363.6
Talc-1%	44.1 ± 1.5	27.6 ± 2.1	1641.5 ± 60.0	11.5 ± 0.2	336.1	363.6
Talc-3%	44.6 ± 1.5	52.8 ± 4.2	1554.4 ± 40.5	14.2 ± 1.1	329.1	351.0
Talc-5%	46.1 ± 1.0	78.4 ± 7.2	1551.5 ± 120.0	15.0 ± 1.5	338.5	361.6
Talc-7%	43.9 ± 1.5	70.4 ± 5.8	1606.0 ± 75.5	12.8 ± 0.8	335.5	361.7

at break of the blends with 3% diatomite was more than 110%, and the blend with 3% talc was more than 80%. However, the blends with more talc or diatomite did not show the ideal elongation at break. This is because more talc or diatomite tends to agglomerate and affect the mechanical properties (40).

Like the properties of the elongation at break, the impact strength increased with the addition of the talc or diatomite. As shown in Figure 5d, the impact strength of the blend with 5% talc was 15 kJ/m^2 (vs 6.5 kJ/m^2). The affinity between the particle and the polyester improved the toughness of the blends. The particle improved the interfacial adhesion and increased the molecular chain entanglement of PLA and PBAT, which consumes more energy when damaged.

3.6 Toughening mechanism of the blends

From the above experimental results, we researched the toughening mechanism of the blends with talc or diatomite. Due to the poor compatibility of PLA and PBAT, the PBAT segments formed particles and had interfacial phase with PLA. As shown in Figure 6, under high

**Figure 6:** Scheme of toughening mechanism of the blends.

tensile strength, the PBAT led to the poor toughening effect of PLA, which is mainly manifested by poor interfacial interaction between two immiscible blends and high energy consumption. The interfacial phase exfoliation occurred in the notched impact test. After the addition of the particles, the talc and diatomite increased the interfacial adhesion and improved the compatibility of PLA and PBAT. As shown in Figure 1, the gap between PLA and PBAT disappeared and the size of the dispersed phase decreased. Therefore, in the process of stretching or impact, the blend was not easy to break from the interface. Thus, talc or diatomite effectively improved the toughness of the blend (41).

4 Conclusion

In this study, talc and diatomite acted as a compatibilizer in the blend to improve the compatibility, structure, and mechanical properties of the PLA/PBAT blends. Talc and diatomite selectively located at the interface phase induced coalescence of the PBAT spherical particles and improved the interfacial adhesion of PLA and PBAT. Nanocomposite increased the crystallization of PBAT. In the heating process of DSC, talc hindered the crystallization of PLA, and in the cooling process, talc improved the crystallization of PBAT. When 5% talc was added to the blends, the crystallization temperature of PBAT increased to 105°C and the crystallinity was 60.8%, because more talc located at the PBAT interface act as nucleating agents to accelerate the crystallization. The viscosity and modulus decreased with the addition of the nanocomposites. Thus, the tensile and impact toughness of the blends were improved for all ratios with talc or diatomite. The elongation at break and impact strength of

the PLA/PBAT blends with 5% diatomite were 115% and 10.2 kJ/m², respectively, and the blends with 5% talc were 79% and 15 kJ/m², respectively. The results showed an excellent possibility of using talc and diatomite modified PLA/PBAT blends with high toughness and excellent processing performance for green-packaging and biomembranes application.

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