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Research Article

Toughness Enhancement of Poly(methyl methacrylate) (PMMA)/Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) Nanocomposites by Multiwall Carbon Nanotubes (MWNT)

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The nanocomposites of PMMA/PHBV filled by MWNT were fabricated by melt blending. The effects of the weight fraction of MWNT on the flow and mechanical properties of the PMMA/PHBV nanocomposites were investigated. The morphologies of fracture surfaces were characterized through scanning electron microscopy (SEM). The impact test results show that the incorporation of 1.5 weight fraction of MWNT increased the impact strength by 42.4%. The flow property and bending strength increased 40.7% and 11%, respectively, by compounding 1.5 weight fraction of MWNT. The morphological behavior indicated transition of fracture surface from brittle nature to ductile. These improved mechanical properties of PMMA/PHBV nanocomposites using MWNT will lead to wider and enhanced applicability of PMMA/PHBV.

1. Introduction

PMMA is a typical transparent amorphous polymer. PMMA has advantages of good flexibility, high strength, and excellent processing property and has been widely used in construction, automobile industry, electronics, and decoration products [1, 2]. However, PMMA has poor wear resistance and solvent resistance and is not easy to degrade and environmental protection, which limits its application [3, 4].

Polyhydroxyalkanoates (PHA) are a biodegradable thermoplastic synthesized by bacterial fermentation of sugar and lipid [5]. PHBV is a kind of PHA, which is synthesized with starch as raw material and fermentation technology. PHBV has good biocompatibility and complete biodegradability. It has been used in many fields such as packaging, biomedical, and agricultural applications [6]. However, PHBV has high crystallinity and brittleness, and its degradation temperature is close to the melting temperature, which makes its processing more difficult. An alternative to overcome the limitations of using PHBV as a potential thermoplastic is to introduce other materials to improve its properties [7].

Carbon nanotubes (CNT) display remarkable mechanical, chemical, and electrical properties. As one-dimensional nanomaterials, CNT has high aspect ratios, which can greatly enhance stress transfer. CNT has been used extensively in polymer composites because of its unique properties [8, 9]. Mazaheri et al. found that CNT can significantly improve the mechanical properties of PA6/ABS composites [10]. Li found that the tensile strength of HIPS can be increased from 6 kJ/m^2 to 12 kJ/m^2 by adding 2.5 wt.% CNT [11]. Choi et al. found that the PMMA-grafted MWNT could improve the mechanical strength and electrical conductivity of polycarbonate (PC) [12]. In this work, PMMA/PHBV/MWNT composites were prepared by melt blending. The PHBV can improve the degradation performance of PMMA and PMMA can improve the processability of PHBV. At the same time, MWNT is used to enhance the strength of composites. The effects of the content of MWNT on the flow properties, mechanical properties, and micromorphology of the composites were analyzed, and the optimum amount of MWNT was determined.

2. Experimental

2.1. Materials. PMMA (IF850) was provided by LG Chemical Company of Korea. The relative molecular mass is 2.2

$\times 10^4$. PHBV (Y100P) was supplied by Ningbo Tianan Biomaterials Co., Ltd. The relative molecular mass is 2.6×10^5. MWNT (XFQ037) was offered by Nanjing Xianfeng Nanometer Material Co., Ltd. The purity of the MWNT is greater than 95%. The diameter of the MWNT is 5-15 nm, and the length is 10-30 μm.

2.2. Preparation of Composites. The preparation process of PMMA/PHBV/MWNT nanocomposites is illustrated in Figure 1.These raw materials were first placed in an oven (DGG-9003 Shanghai Sen Xin Experimental Instrument Co., Ltd.) and dried for 6 hours at 80°C to remove excess moisture. Then, the dried mixtures were premixed by high-speed mixer (SHR-10 Zhangjiagang Second Light Machinery Co., Ltd.) according to Table 1. The speed of the high-speed mixer was 1410 rpm, and the time was 10 minutes. Then mixed materials were blended by twin-screw extruder (CTE35 Coperion (Nanjing) Machinery Co., Ltd.) for melt extrusion. The temperature range was 160°C-250°C, and the rotational speed of the main engine was about 45 rpm. Finally, the extruded pellets were injected into a standard spline by injection molding machine (FT-110 Zhejiang Sound Plastic Machinery Co., Ltd.).

2.3. Characterization of Composites. The melt flow performance was tested by a melt flow tester (XRL-400-A Chengde Precision Testing Machine Co., Ltd.) according to the China standard GB/T 3682-1983 with the temperature of 230°C and the load of 3.8 kg. The tensile property was measured by a universal tensile machine (WDT-W Chengde Precision Testing Machine Co., Ltd.) according to the China standard GB/T 1040-1992. The speed of universal tensile machine was 5 mm/min, and the size of the dumbbell standard spline at the narrowest part was 10 mm × 4 mm. The bending strength was tested according to the China standard GB/T9341-2008. The speed of the universal tensile machine is 6 mm/min, and the thickness of the dumbbell standard spline is 2 mm. The impact performance was tested by the cantilever impact tester (XB-5 Chengde Precision Testing Machine Co., Ltd.) according to the test method specified in the China standard GB/T 1843-1996. The size of the standard notched spline was 10 mm × 4 mm, and 1 J was selected for pendulum. After vacuum gold spraying, the impact fracture spline was observed by SEM (Evo18 Carl Zeiss AG).

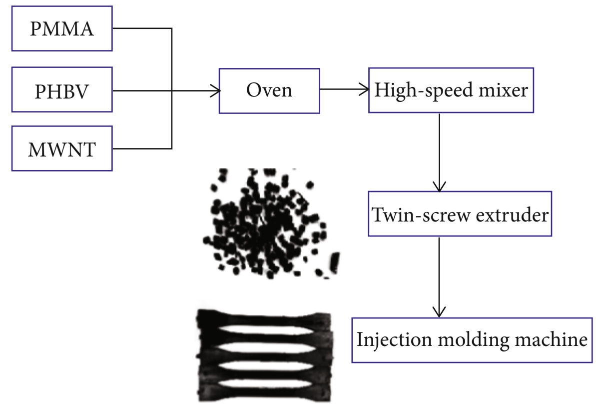


Figure 1: Schematic illustration of the preparation process of PMMA/PHBV/MWNT nanocomposites.

Table 1: Formula of the PMMA/PHBV/MWNT.

	PMMA (wt.%)	PHBV (wt.%)	MWNT (wt.%)
1	80	20	0
2	79.6	19.9	0.5
3	79.2	19.8	1
4	78.8	19.7	1.5
5	78.4	19.6	2

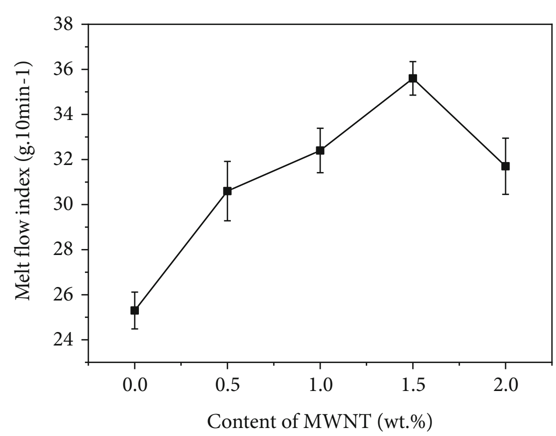


Figure 2: Effect of the content of MWNT on the flow performance of PMMA/PHBV nanocomposites.

3. Results and Discussions

3.1. Melt Flow Performance. Polymer materials need to flow before they can be processed into final products. The fluidity of materials directly affects the performance of final products. The effect of MWNT weight fraction on the fluidity of PMMA/PHBV nanocomposites is shown in Figure 2. The values of the flow properties increase with increasing the MWNT weight fraction, and the value of the flow properties is up to the maximum at the MWNT weight fraction of 1.5 wt.%. MWNT is a kind of nanomaterial with larger aspect ratio. A small amount of addition of MWNT will help improve the flow performance of PMMA/PHBV. When the amount of MWNT reaches a certain value, too much MWNT will hinder the transition of polymer chain flow, resulting in poor melt flow performance.

3.2. Mechanical Performance. The effect of MWNT weight fraction on the tensile strength of PMMA/PHBV nanocomposites is shown in Figure 3. With the increase of MWNT weight fraction, the tensile strength of PMMA/PHBV nanocomposites decreases first and then increases. The addition of a small amount of MWNT is not easy to form uniform dispersion in the composite, so it will reduce the tensile strength. With the increase of MWNT content, the dispersion of MWNT in the composites begins to become uniform, and

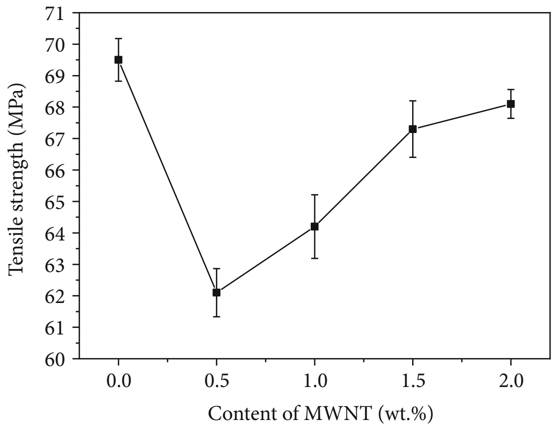


FIGURE 3: Effect of the content of MWNT on the tensile strength of PMMA/PHBV nanocomposites.

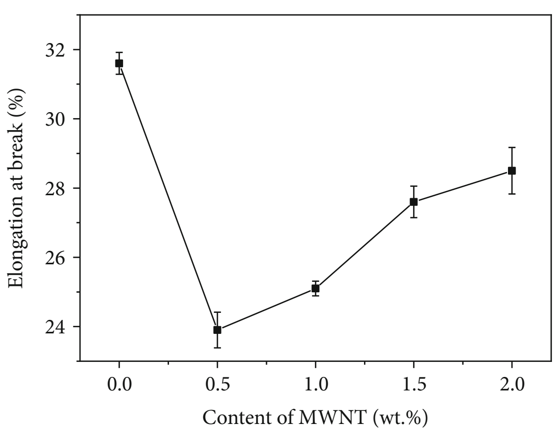


FIGURE 4: Effect of the content of MWNT on the elongation at break of PMMA/PHBV nanocomposites.

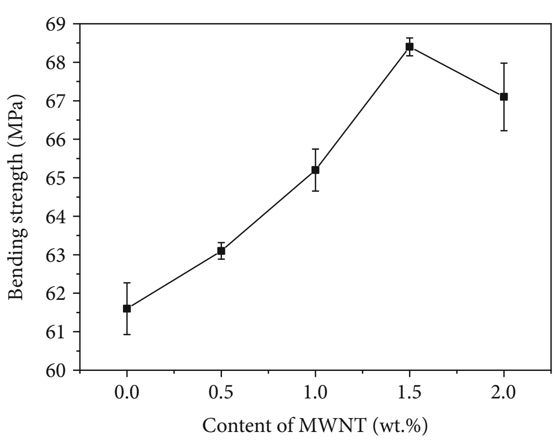


FIGURE 5: Effect of the content of MWNT on the bending strength of PMMA/PHBV nanocomposites.

the tensile strength increases. Li found the similar phenomena in the research of PP composites reinforced with graphene, and it is difficult to disperse a small amount of graphene uniformly in PP [9].

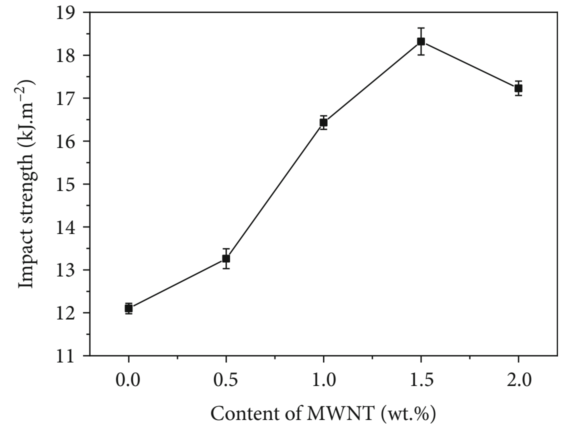
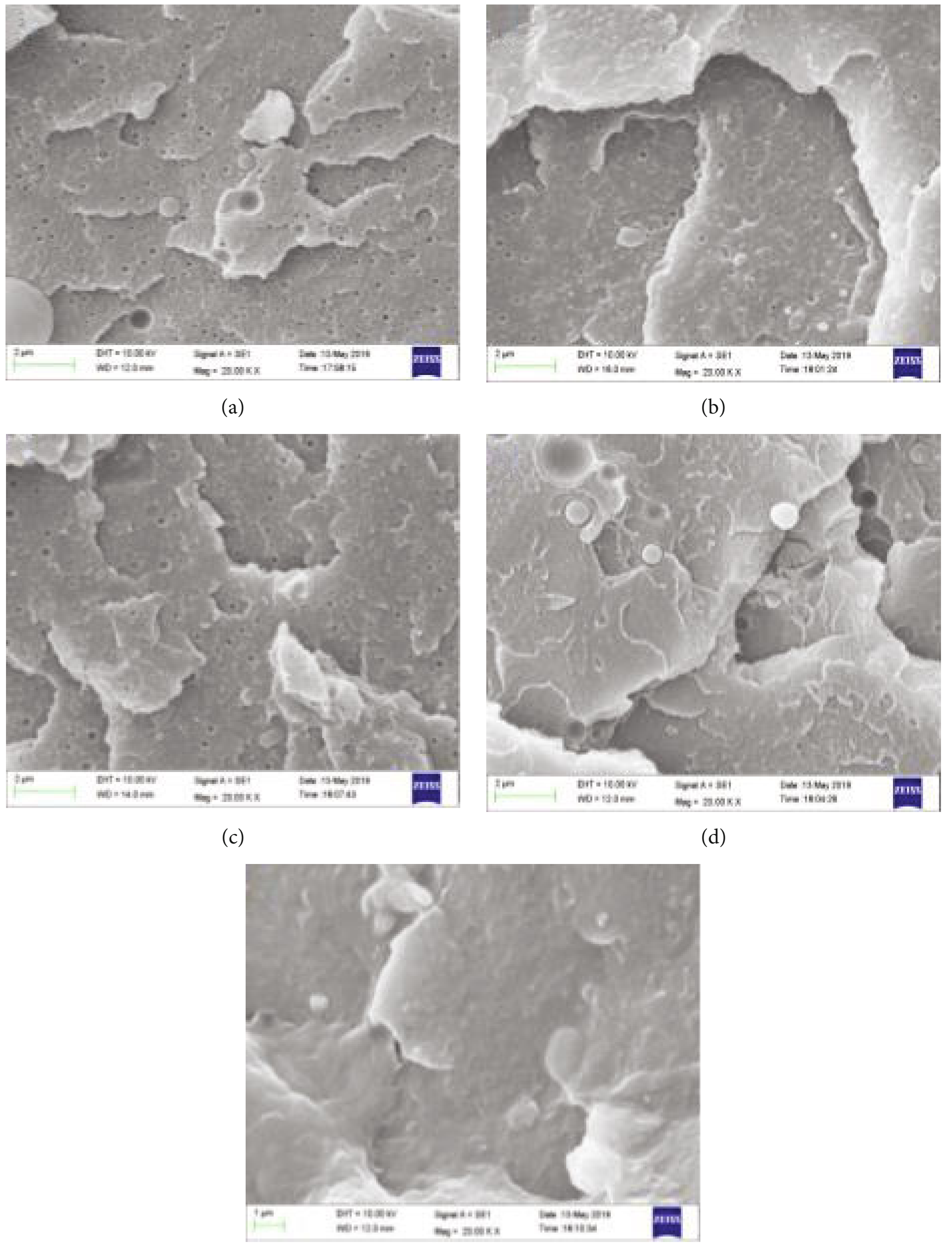


FIGURE 6: Effect of the content of MWNT on the impact strength of PMMA/PHBV nanocomposites.

The effect of MWNT weight fraction on the elongation at break of PMMA/PHBV nanocomposites is shown in Figure 4. With the increase of MWNT weight fraction, the elongation at break of PMMA/PHBV nanocomposites decreases first and then increases. As the loading of the MWNT is 0.5 weight fraction, the elongation at break of the composites decreases by 24.2%. When the addition amount of MWNT is less than 0.5 wt.%, MWNT is difficult to be evenly dispersed in the matrix, which easily forms stress concentration and reduces the elongation at break. With the increase of MWNT content, the dispersion of MWNT in the matrix begins to become uniform, and the elongation at break increases.

The effect of MWNT weight fraction on the bending strength of PMMA/PHBV nanocomposites is shown in Figure 5. With the increase of MWNT weight fraction, the bending strength of the nanocomposites increases greatly. As the loading of MWNT is 1.5 weight fraction, the bending strength of the nanocomposites increases about 11%. Then, the bending strength of the nanocomposites begins to decrease with the increase of the amount of MWNT. When fillers are loaded into polymer materials, they will play a role of skeleton in the matrix, and the movement of molecular chains will be limited, leading to improve the stiffness of the composite systems [13]. For PHBV, the MWNT will play a role of heterogeneous nucleation, which leads to increase the crystalline degree and improve correspondingly the stiffness of the composite systems.

Impact strength is an important index to measure the mechanical properties of engineering plastics. Figure 6 presents the dependence of the impact strength on the MWNT weight fraction of the PMMA/PHBV nanocomposites. It is shown that the values of the impact strength of the PMMA/PHBV nanocomposite systems sharply increase with an addition of the MWNT weight fraction and the value of the impact strength is up to the maximum at the MWNT weight fraction of 1.5. The impact strength of composites is related to toughness. MWNT has high mechanical strength. In addition, MWNT has high aspect ratio, which can effectively disperse the impact force when the material is impacted by external



(a) (b) (c) (d) (e)

Figure 7: SEM of composite fracture surface with different MWNT weight fractions: (a) 0 wt.%; (b) 0.5 wt.%; (c) 1 wt.%; (d) 1.5 wt.%; (e) 2 wt.%.

force. Therefore, the addition of MWNT can improve the impact strength of the composites.

3.3. Morphology of Fractured Surfaces. The SEM test results of the composites are shown in Figure 7. The morphology of the fractured surface of the PMMA/PHBV is shown in Figure 7(a). The fractured surface is smooth, and it means that the microcrack in the specimen of PMMA/PHBV expands directly under impact load which leads to low-impact fracture strength. With the increase of MWNT weight fraction, the section morphology begins to become rough and pits appeared. Figure 7(d) shows the SEM micrograph of the fractured surface of the PMMA/PHBV containing 1.5 weight fraction of MWNT. The roughness of the microsection of the composite is the largest, which is consistent with the change of impact strength properties. Liang et al. found the similar phenomena in the research of PP composites reinforced with MWNT. In addition to the fact that MWNT could induce the PP matrix to generate deformation and it could absorb the impact energy, he thought that the nanomaterials would block the development of the microcracks [14].

4. Conclusions

In this paper, PMMA/PHBV/MWNT nanocomposites were prepared by melt blending, and the effects of content of

MWNT on the properties of the composites were studied. The experimental results show that MWNT can improve the flow performance and the impact strength and the bending strength of the nanocomposites. Adding 1.5 wt.% MWNT, the flow properties of the composites have improved by 40.7%. And the bending strength and impact strength increase by 11% and 42.4%, respectively. The SEM micrographs of fractured surfaces of the PMMA/PHBV with and without MWNT show that MWNT could increase the toughness of the composites.

Data Availability

All data are included in this paper.

Conflicts of Interest

The author declares that there are no conflicts of interest.

Acknowledgments

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