Article

Effect of Recycled High-Density Polyethylene on the Impact Strength of Polybutylene Terephthalate/Polyamide 6

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ABSTRACT: Recycling high-density polyethylene (HDPE) is crucial to addressing plastic waste challenges. This study investigates the mechanical properties of blends composed of HDPE, polybutylene terephthalate (PBT), and polyamide 6 (PA6). Blends with varying HDPE content (0, 70, 80, 90, and 100%) were analyzed using injection molding to determine their impact toughness and structural characteristics. PBT and PA6 (blended in a 50:50 ratio) were combined with HDPE to create composites with enhanced properties. Testing included unnotched impact strength analysis and scanning electron microscopy (SEM). HDPE, a flexible thermoplastic, was paired with PBT and PA6, known for their strength and heat resistance, to produce a blend with superior mechanical performance. Results reveal that incorporating HDPE enhances the impact toughness of the composites compared to the pure PBT/PA6 blend, offering promising potential for many diverse applications in materials engineering in the automotive industry, household products, and protective casings of electronic products.

Keywords: HDPE; PBT/PA6 blends; Impact strength; Microstructure

1. Introduction

The rapid growth in global plastic production stems from its cost-effectiveness, versatility, and high physical and chemical stability. Common thermoplastics such as polyethylene terephthalate (PET), high-density polyethylene (HDPE), polyvinyl chloride (PVC), and others dominate consumer products. However, their durability creates a significant environmental challenge, as plastics take decades to decompose [1,2]. In Vietnam, plastic waste generation has escalated dramatically, with current estimates reaching 3.27 million tons annually, including 0.28–0.73 million tons discharged into the ocean [3]. Recycling remains underutilized, with only 27% of plastic waste being recycled, while the remainder is primarily treated through landfilling and incineration [4,5]. This problem highlights the urgent need for innovative solutions to enhance recycling and reduce the environmental impact of plastic waste.

This study focuses on HDPE's unnotched impact toughness and microstructure combined with a polybutylene terephthalate (PBT)/polyamide 6 (PA6) blend. HDPE, known for its flexibility, impact resistance, and chemical stability, is a promising material for blending with PBT/PA6, which offers high strength, heat resistance, and excellent chemical stability. However, the compatibility between PBT and PA6 remains challenging, with limited studies exploring their blends. By combining HDPE with a 50:50 PBT/PA6 blend, this research aims to overcome these limitations, enhancing impact resistance and achieving a homogeneous material with superior mechanical properties.

The study investigates the optimal HDPE ratio in the blend, employing mixing, granulation, injection molding, and subsequent impact strength and microstructure analysis. This research addresses material compatibility challenges and

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contributes to reducing plastic waste, saving resources, and creating high-value materials for diverse applications [6,7]. Through the above research, we will combine the results of the application topic to manufacture battery cases and holders using a mixture of HDPE/PBT/PA6 plastic. The combination of superior properties from recycled HDPE, PBT, and PA6 will help increase the mechanical properties of the HDPE/PBT/PA6 plastic mixture compared to conventional plastic materials. The cost of manufacturing this new plastic mixture will help save more than virgin plastic and will also reduce the amount of waste from production.

2. Materials and Methods

This study used three types of plastic materials: PBT with a density of 1.3 g/cm^3, PA6 with 1.13 g/cm^3, and HDPE with a density of 0.923 g/cm^3 derived from recycled materials of Dong Nhan Phat Co., Ltd. (Di An City, Binh Duong Province, Vietnam). PBT/PA6 and HDPE were mixed according to the ratio in Table 1. The mixture was dried at 110 °C for about 2 h, achieving a moisture content of less than 0.03%, using the injection molding method using a 100-ton Toshiba plastic injection molding machine (Japan). The mixture is mixed in proportions according to the table below:

Table 1. Ratio of HDPE/PBT/PA6 blends.

Sample	Ratio (%)		
	HDPE	PBT	PA6
100HDPE	100	0	0
90HDPE	90	5	5
80HDPE	80	10	10
70HDPE	70	15	15
0HDPE	0	50	50

The injection molding process was conducted with recycled HDPE/PBT/PA6 blends at weight ratios of 100:0:0, 90:5:5, 80:10:10, 70:15:15, and 0:50:50. To remove moisture and improve processing, all material ratios were pre-dried in an oven at 100 °C for 1 h and 30 min before injection molding.

The HDPE/PBT/PA6 blends were prepared by mixing the dried pellets in the specified ratios using a laboratory mixer. The mixed materials were then cooled to room temperature before being processed further. The injection molding of the test specimens was carried out using a Toshiba 100T injection molding machine. Key parameters for the injection process included:

- Melt temperature: 180 °C;
- Holding pressure: 50 MPa;
- Mold temperature: 50 °C.

The impact properties of the molded specimens were evaluated using the Izod Impact Strength parameter, following the ASTM D256 standard test method. The tests were performed at room temperature to assess the blends' unnotched impact strength.

Theory/Calculation

The formula for calculating impact strength without a V-notch groove, commonly referred to as unnotched impact strength, is:

$$Impact\ strength = W/A \quad (1)$$

where, W: Energy absorbed by the specimen during fracture (kJ); A: Cross-sectional area of the specimen at the point of impact (m^2).

$$A = b \times h \quad (2)$$

where, b: the depth of the specimen (m); h: the width of the specimen (m).

3. Results

3.1. The Unnotched Impact Strength

The V-notch impact strength was measured according to ASTM D256 using the Izod Tinius Olsen IT504 impact toughness tester (Tinius Olsen Company, Horsham, PA, USA).

Table 2 shows that the impact strength value decreases gradually when we add the PBT/PA6 mixture to HDPE. For the 90HDPE sample, it gives us a value of 109.08 kJ/m^2 and gradually decreases to 28.14 kJ/m^2 (80HDPE sample) and 21.99 kJ/m^2 (70HDPE sample) and finally 16.17 kJ/m^2 (0HDPE sample). It can be seen that compared to the ratio of 100HDPE, the impact toughness value decreases slightly by 1.023 times and decreases sharply by 6.9 times (0HDPE sample). Increasing the PBT/PA6 content generally increases the impact toughness of the HDPE/PBT/PA6 mixture but will decrease slightly compared to the original 100HDPE. At a ratio of 90/5/5, we will get the best results to meet the requirements for combining PBT/PA6 and recycling HDPE blends.

Table 2. Impact strength measurement results of samples.

Times	100HDPE	90HDPE	80HDPE	70HDPE	0HDPE
1	109.37	113.69	29.53	21.51	16.24
2	110.05	108.34	27.96	22.92	16.23
3	110.88	105.75	29.22	21.26	15
4	113.97	114.04	25.52	21.31	16.38
5	113.76	103.57	28.48	22.97	17
Average	111.6	109.08	28.14	21.99	16.17
Standard deviation	2.13	4.68	1.59	0.87	0.73

Figure 1 briefly compares the unnotched impact test specimens before and after the test. Compared to before the test, the shape of the specimen has minor deformation, and the material is not broken, indicating that the material has good ductility and absorbs impact energy quite well.

(**a**) (**b**)

Figure 1. Unnotched impact test specimen. (**a**) Sample before testing; (**b**) Sample after testing.

Figure 2 shows the impact strength of HDPE/PBT/PA6 blends. Looking at the bar chart, the impact strength of the sample containing 100% HDPE has the highest average impact strength with a value of 111.6 kJ/m^2 compared to other samples. The sample containing 0% HDPE has the lowest, with a value of 16.17 kJ/m^2. Compared to pure HDPE, the impact toughness value is reduced by 6.9 times. It gradually decreases in the samples of 80% HDPE (28.14 kJ/m^2) and 70% HDPE (21.99 kJ/m^2) with high PBT/PA6 content. These results were calculated using Formulas (1) and (2).

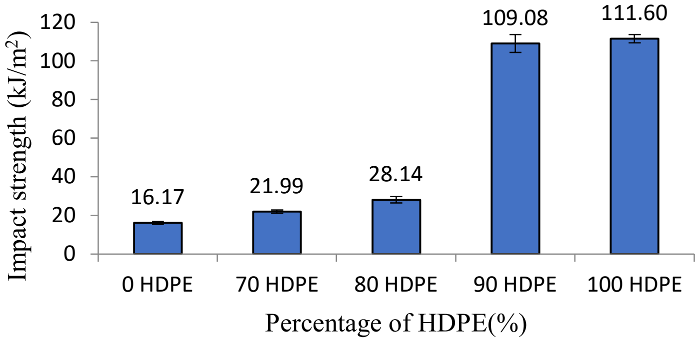
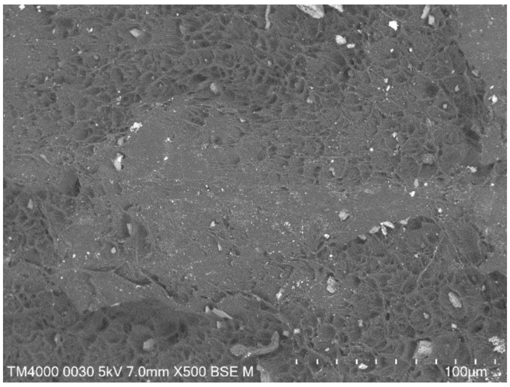


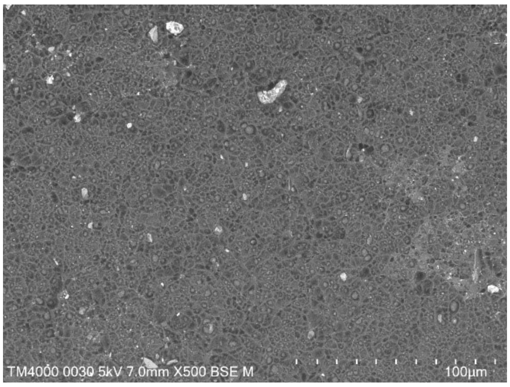
Figure 2. Average impact strength chart of HDPE/PBT/PA6 blend samples.

3.2. Microstructure

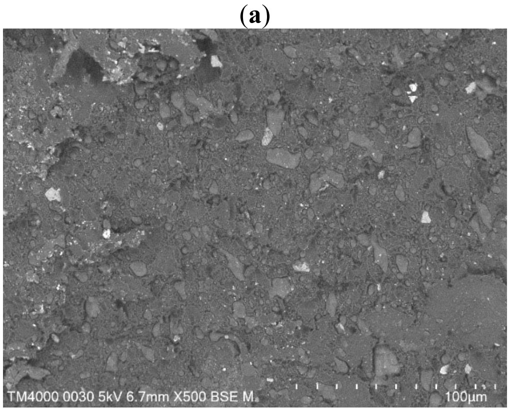
Based on the micrograph in Figures 3 and 4a, we can see that the surface structure of the 100HDPE sample is relatively smooth, characterized by a mesh-like crystal lattice structure. This result is why HDPE can more effectively induce the cracking and shearing of the matrix to absorb the breaking energy, leading to increased impact strength. The HDPE matrix is relatively homogeneous in structure, without voids. From Figures 3 and 4b, when increasing the PBT/PA6 content, we gradually see the appearance of small spherical particles of uniform size and distribution on the substrate, leading to a significant increase in the bonding between PBT and PA6. At this ratio, we have not observed the dispersed phases of the core-shell structure.



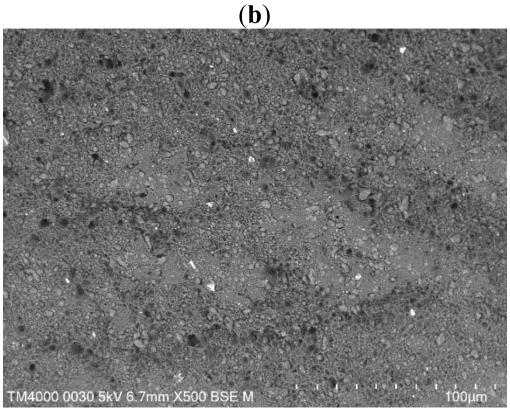
(**a**)



(**b**)

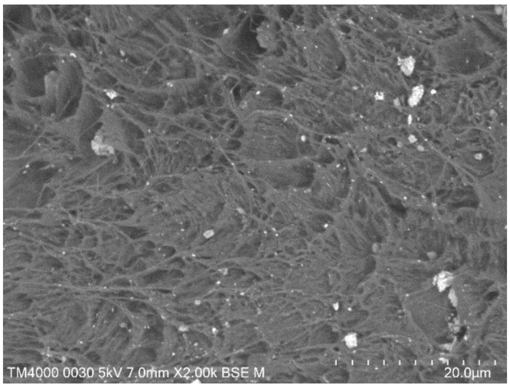


(**c**)

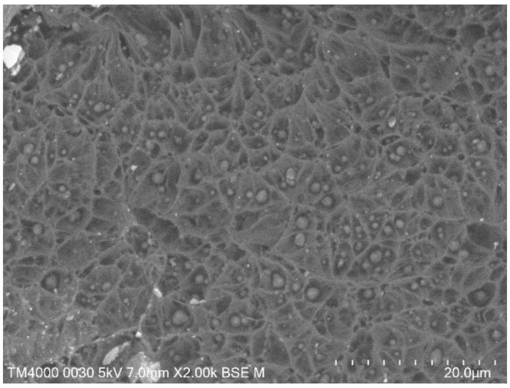


(**d**)

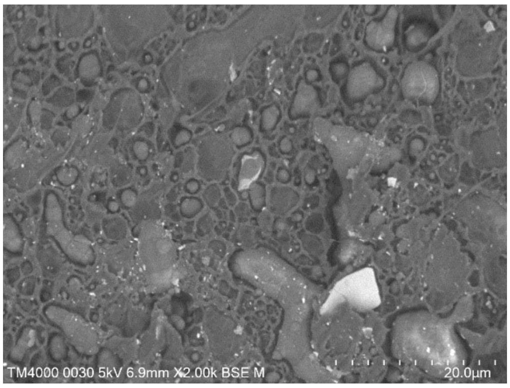
Figure 3. Microstructure image at 500× magnification. (**a**) 100HDPE; (**b**) 90HDPE; (**c**) 80HDPE; (**d**) 70HDPE.



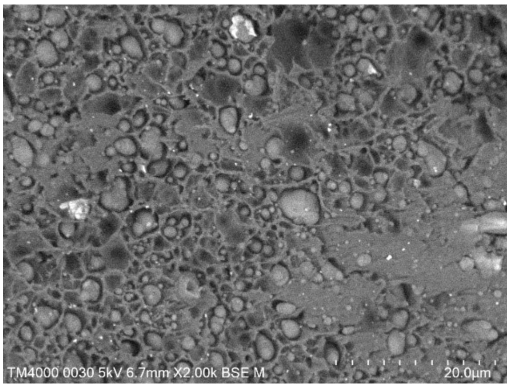
(**a**)



(**b**)



(**c**)



(**d**)

Figure 4. Microstructure at 2000× magnification. (**a**) 100HDPE; (**b**) 90HDPE; (**c**) 80HDPE; (**d**) 70HDPE.

Figures 3 and 4c,d show that when increasing the content of PBT/PA6, spherical particles with uneven sizes, scattered distribution, and concave surfaces begin to appear, forming voids with cracks, so the bonds are easily destroyed, leading to a decrease in impact toughness compared to the ratio of 90/5/5. At this time, the incompatibility of the two substances is shown, and the PBT/PA6 particles no longer play the role of a bridge in the HDPE matrix but are the cause of the decrease in the material's mechanical properties. At this ratio, when subjected to an impact force or a deformation stress, the core structure is stretched, and the PBT/PA6 particles are separated from the substrate due to crack propagation, leading to broken bonds. Figures 3 and 4c show that when the PBT/PA6 content exceeds the allowable level, the bonds are broken, creating more gaps, reducing the adhesion of the two types of plastic, causing many damaged spots, cracks on the surface, and brittleness of the plastic, causing the impact toughness to decrease significantly.

4. Discussion

The impact toughness of HDPE/PBT/PA6 blends is significantly influenced by the HDPE and PBT/PA6 content. As the HDPE content increases, the material becomes more rigid and flexible due to HDPE's high molecular density structure. However, the core-shell structure's effectiveness diminishes at higher PBT/PA6 ratios. Studies show that as the PBT/PA6 content exceeds 10%, the density of PBT/PA6 particles increases, causing a decrease in bonding with the HDPE matrix, leading to lower energy absorption and reduced impact toughness [8–12].

Moreover, research indicates that the dispersion of HDPE within the blend enhances mechanical properties by reducing plastic shrinkage and improving bonding between phases, thereby increasing toughness.

When the PBT/PA6 blend content increases to more than 10%, specifically at the ratio of 70/15/15, the core-shell structure is no longer effective because the density of PBT/PA6 particles increases, causing partitioning that hinders the bonding of HDPE matrix molecules. At this time, the strength of the dispersed phase is lower, and the energy absorption decreases during the stretching process, leading to a decrease in impact toughness. Impact toughness is also affected by the HDPE content. Zhou et al. have studied the mechanical properties of PBT/PA6 when the three substances are combined. In the study, Zhou et al. pointed out that the good stress transfer effect between the dispersed particles and the polymer matrix makes the polymer blend harder [11]. The dispersion density of HDPE helps the blend reduce plastic shrinkage and maintain the shape of the sample. However, higher PBT/PA6 content increases particle interactions, creating voids and stress concentration zones and diminishing the overall impact toughness. The balance between HDPE and PBT/PA6 content is crucial to maintaining optimal mechanical properties while minimizing brittleness [13,14].

The study highlights how the PBT/PA6 content influences the impact toughness of HDPE/PBT/PA6 blends. As the PBT/PA6 ratio increases, the mechanical properties degrade, especially when the ratio reaches its maximum at 100%, resulting in a significant decrease in impact toughness to 16.17 kJ/m^2. This result is attributed to reduced inter-particle spacing and poor bonding between phases, which causes particle breakage and void formation, leading to lower toughness. Incorporating PBT/PA6 into HDPE creates stress concentration zones that reduce energy absorption, further decreasing impact toughness.

In the study, integrating of HDPE into PBT/PA6 blends significantly influences the material's mechanical properties. As the HDPE content increases, the core-shell particles' interface strength and elastic modulus surpass those of pure PBT/PA6, mainly due to the high modulus of PA6 and covalent bonding between PA6 and HDPE. This results in better energy absorption during stretching, thereby enhancing impact toughness. The core-shell structure facilitates better stress distribution, improving the overall mechanical performance. Similarly, HDPE/PA6/PBT ternary blends show improved toughness due to core-shell dispersed particles [15–17].

SEM microstructure demonstrates the influence of HDPE on the microstructure of HDPE/PBT/PA6 blends. In blends containing HDPE, the dispersed phase retains its spherical shape after impact, indicating better phase stability. In contrast, blends with poor HDPE compatibility display visible gaps between the dispersed phase and the matrix, signifying weak interfacial bonding and reduced impact toughness. Additionally, SEM analysis identified banded spherulites within the HDPE microstructure, though artifacts caused by sample preparation methods like etching may appear in formulated compositions [18–21].

These findings highlight the importance of optimizing the HDPE content to balance the core-shell structure's effectiveness, ensuring superior impact resistance while maintaining compatibility between components.

5. Conclusions

- Recycled HDPE has demonstrated potential for various industrial applications. This study focused on the unnotched impact toughness properties of recycled HDPE blends, emphasizing the utility of recycled plastics as an alternative material for household and industrial products. The samples were prepared using extrusion and injection molding techniques;
- The HDPE and PBT/PA6 content ratio significantly influenced the blends' mechanical properties and microstructure. While a 90/5/5 ratio showed promising results with minimal defects and improved impact toughness, the 70/15/15 ratio exhibited numerous microstructural defects around the PBT/PA6 particles, attributed to weak interfacial adhesion between HDPE and PBT/PA6, leading to decreased tensile strength. These findings suggest that the reinforcement mechanism is multifaceted and relies on combined structural mechanisms;
- Although PBT/PA6 can alter the mechanical properties of HDPE blends, higher PBT/PA6 content reduces impact toughness and mechanical efficiency. To optimize mechanical properties, the PBT/PA6 content in the mixture should remain below 10%. At a 90/5/5 ratio, the blend achieved desirable mechanical properties, meeting performance criteria.

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Author Contributions

Conceptualization, P.T.H.N.; Methodology, T.H.N.; Software, B.H.D.; Validation, D.Q.T.; Formal Analysis, T.H.N., B.H.D. and D.Q.T.; Investigation, D.T.V.A.; Resources, N.C.T. and P.Q.A.; Data Curation, N.V.T.; Writing—Original Draft Preparation, T.H.N., B.H.D. and D.Q.T.; Writing—Review & Editing, P.T.H.N.; Visualization, P.T.H.N.; Supervision, P.T.H.N.; Project Administration, P.T.H.N. During the preparation of this work, the authors used Grammarly to improve readability and language. After using this tool/service, the authors reviewed and edited the content as needed and took full responsibility for the publication's content.

Ethics Statement

Not applicable.

Informed Consent Statement

Not applicable.

Data Availability Statement

The data used to support the findings of this study are available from the corresponding author upon request.

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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