POLYMER BLENDS

Effect of Polypropylene Percentage on Morphological and Mechanical Properties of Polybutylene Terephthalate/Polypropylene Blends

Pham Thi Hong Nga^{*a*,*} and Nguyen Thanh Tan^{*a*}

 ^aHo Chi Minh City University of Technology and Education, Thu Duc City, Ho Chi Minh City, 71307 Vietnam
 *e-mail: hongnga@hcmute.edu.vn
 Received August 21, 2023; revised March 25, 2024; accepted March 26, 2024

Abstract—Polybutylene terephthalate (PBT) has a strong resistance to abrasion, deformation, and chemical resistance. However, how to further improve the impact toughness of PBT is still an issue that receives the attention of many researchers. This investigation aims to enhance its mechanical properties by mixing it with polypropylene (PP). The tensile strength, impact strength, flexural strength, and microstructure are accorded to ASTM D638, ASTM D256, and ASTM D790. In this analysis, PP was mixed with PBT with different ratios of 95/5, 90/10, 85/15, 80/20, and 75/25. Use injection molding technique to mold specimens for mechanical testing. The results show that the impact strength is 3.7, 6.6, 5.63, 5.2, 5.1, and 4.9 kJ/m², corresponding to 0, 5, 10, 15, 20, and 25% PP. The impact strength of PBT/PP blends is higher than neat PBT. Especially in the 5% PP sample, the impact strength increase can reach the highest level of 80.3% compared to neat PBT. The flexural strength tends to decrease with increasing PP content. After measuring, we get 77.9, 65.6, 62.2, 58.4, 58.2, and 54.0 MPa, corresponding to 0, 5, 10, 15, 20, and 25% PP. The microstructure of the blends showed that spherical PP particles were interwoven into the PBT substrate, but no interphase adhesion was observed. In general, selecting a suitable PBT/PP blend could improve the impact strength while preserving the tensile strength, allowing it to be applied widely.

DOI: 10.1134/S0965545X2460042X

INTRODUCTION

Waste from toothbrush bristles is of little interest to scientists. This waste, with the primary material being polybutylene terephthalate (PBT), often has brittle properties. When recycled, it is frequently challenging to inject mold to create products. After recycling, products often have low mechanical properties, especially low impact toughness. Therefore, making recycled toothbrush bristles with high impact toughness while retaining other properties such as tensile strength and flexural strength remains a big challenge.

Neat polymers usually have some limits related to their homogeneous structure. Therefore, to improve the characteristics of neat polymers, these polymers are blended to create new materials with more advanced properties [1-4]. PBT is a semi-crystalline thermoplastic polymer with excellent mechanical properties and strong resistance to abrasion, deformation, and chemical resistance. It is widely used in the automotive industry, electrical and electronic industries, and other fields [5]. However, some disadvantages, such as low impact strength, high shrinkage, and low strain temperature, limit its applications [6]. Recently, many researchers have aimed to improve PBT characteristics by mixing it with other polymers or fillers such as polycarbonate (PC), Polyethylene terephthalate (PET), Polyamide PA6, etc. Kim et al. [7] investigated the function of maleic anhydride (MAH) as a compatibilizer for the PBT/EVA blends. The results showed that the impact strength of the PBT/EVA-g-MAH (80/20) increased about three times compared to that of the PBT/EVA (80/20). This phenomenon is because adding MAH to the blends improves surface adhesion, increasing impact strength. Tan et al. [8] investigated the mixing of PC and PBT with ratios of 100/0, 40/60, 30/70, and 20/80, demonstrating that as the PC ratio increases, so do the impact strength and bending durability.

Polypropylene (PP) is today's cheapest and most commonly used plastics. It is a thermoplastic polymer produced from the monomer propylene. PP is widely employed in various industrial fields such as auto parts, home appliances, furniture, packaging, etc. [9]. Many studies have been conducted in recent years to improve the mechanical properties of PBT through copolymerization or plasticization [10, 11]. In these

 Table 1. Compositions of the PBT/PP blend samples (wt %)

Sample	100PBT	5PP	10PP	15PP	20PP	25PP
PBT, wt %	100	95	90	85	80	75
PP, wt %	0	5	10	15	20	25

 Table 2. Molding conditions of the testing samples

Molding parameters	Unit	Value
Melt temperature	°C	235
Injection pressure	MPa	120
Injection time	S	8
Holding time	S	8
Holding pressure	MPa	60
Skew speed	rpm	100

studies, the most commonly used method is to add suitable, compatible substances to improve the properties of the blends [12]. For example, Hietaoja et al. [13] investigated the function of oxazoline as a compatibilizer for PBT/PP blends. The results demonstrated that this composite significantly improved the blends' toughness while maintaining strength and hardness. Larsen Børve et al. [14] surveyed PBT/PP isotactic blends containing 10-30 wt % of PBT. According to the report, the viscosity ratio, compatibilizer content, and PBT content all play essential roles in the final blend characteristics. Tsai et al. [15] used ethylene-co-glycidyl methacrylate as a compatibilizer in PBT/PP blends to reduce surface tension during fusion, resulting in finer dispersed phase morphology and improved mechanical properties.

This study aimed to improve the impact toughness of PBT while preserving its durability. We examined the effects of PP on the tensile strength, flexural strength, and impact strength properties of PBT/PP blends with different PP ratios.

EXPERIMENTAL

This study used PBT purchased from Toan Dai Hung Trading Service Co., Ltd., a plastic company with product grade TACOMA TC BN60 originating from India/China of Lanxess plastic company (Germany). PP was obtained from Tuan Ngoc company with the product code PP 1100N, originating from Saudi Arabia. The melt indexes of the PBT and PP polymers are 60 g/10 min (at 250°C) and 12 g/10 min, respectively. We tested the MFI of the blends at 250°C using the 6MPCA Advanced Melt Flow Indexer machine, focusing on the lowest PP of 5% and the highest PP of 25% among these blends. The other blends have the MFI index value around these values. Because of the limited conditions, other blends are not tested for the MFI index. The MFI value of the sample PBT/5% PP is 55.8 g/10 min, while the sample PBT/25% PP has the MFI value of 35.1 g/10 min. With other blends, the MFI values could range from 55.8 g/10 min to 35.1 g/10 min. The injection molding machine is a Haitian MA 1200 III machine. The PBT/PP mixture was mixed evenly by a plastic granule mixing device according to the ratios shown in Table 1 and then dried at 80°C for 5 hours. After drying, the mixture is injected to generate samples at 235°C. During the injection process, the PBT/PP blend was heated through five stages, initially at 215°C and increased by 5°C through each step, keeping the outlet temperature at 235°C before injection into the mold. The injection molding conditions are presented in Table 2.

Samples were prepared for tensile strength with sizes according to ASTM D638 standards, using an AG-X plus Shimadzu universal tensile machine. Each piece of 6 ratios must meet the conditions of no warping, burr, or lack of spray and be measured in a laboratory with a temperature of $23 \pm 2^{\circ}$ C. The samples are pulled at 5 mm/s until rupture.

We measure flexural strength according to ASTM D790 using the Shimadzu Autograph AG-X Plus. Before beginning the experiment, we ensure the experimental environment is at a temperature of $23 \pm 2^{\circ}$ C and a humidity of $50 \pm 5\%$. In addition, samples must be stored in the laboratory for at least 4 hours. The model was mounted on the machine and measured at a 1.56 mm/min speed.

The impact strength samples are tested in an environment with a temperature of $23 \pm 2^{\circ}$ C and a humidity of $50 \pm 5\%$, measured in an Izod Tinius Olsen IT504 impact tester. The microstructure was tested using a Hitachi TM4000Plus electron microscope.

RESULTS AND DISCUSSION

Sample after Injection

Figure 1 shows the tensile, flexural, and impact strength samples after injection. The injection molding samples of PBT/PP mixture with the ratios of 100/0, 95/5, 90/10, 85/15, and 80/20 are all very similar in shape, size, and color. In pressing samples with a higher percentage of PP, the shrinkage is more significant, leading to easy mold jamming, swarf, welds, and underfilling, making injection molding difficult. By adjusting the appropriate parameters, the samples are created perfectly [16, 17].



Fig. 1. Sample after injecting.



Fig. 2. Tensile stress-strain curves of (a) 100% PBT and PBT/PP blends: (1) 5PP, (2) 10PP, (3) 15PP, (4) 20PP, (5) 25PP.

Tensile Strength

Figure 2 depicts the tensile stress-strain diagrams of the blend samples. The variations of the tensile stress with the tensile strain from the figure point out the differences between the models by a large percentage of variation. The tensile stress of the pieces at the beginning of the tensile increases gradually and decreases as the samples fracture. In general, the fracture mechanism of the examples is a brittle fracture with low strain values [18].

The average tensile strength comparisons of different PBT/PP blends are displayed in Fig. 3. The results show that the tensile strength decreases from 60.1 MPa to 42.5, 39.0, 35.5, 34.3, 31.9, and 30.7 MPa when the PP ratio gradually increases from 0% to 5, 10, 15, 20, 25, and 100%. Therefore, adding PP will reduce the tensile strength of neat PBT because the two substances, PBT/PP, are incompatible due to the significant difference in polarity [19]. That incompatibility will reduce the tensile strength. The incompatibility of the blends reduces the intermolecular bonds in the combinations [20].

Figure 4 shows the average elongation at break comparisons of different PBT/PP blends. The average elongation at break values is 48.0, 21.2, 20.8, 6.2, 5.6,

and 4.7%, corresponding to 0, 5, 10, 15, 20, and 25% PP. The results show that adding PP to the blend significantly reduces the elongation value due to the poor compatibility between PP and PBT. Specifically, at 5% PP blend, elongation at break and tensile modulus values of the sample are 21.2% and 3310 MPa. In contrast, the pure PBT sample has elongation at break and tensile modulus values of 48% and 520 MPa. Therefore, adding 5% PP to PBT leads to a reduction in elongation value. The reason is the poor compatibility between PP and PBT. However, the tensile modulus increases when adding 5% PP to the blends due to the well-dispersed and fine structure of the PP phase in the PBT matrix.

Flexural Strength

Figure 5 provides an overview of the influence of PP on the flexural strength of the PBT/PP composite. It can be seen that neat PBT has the highest flexural strength. The curves of the specimens were mainly of the same shape, and none of the models broke suddenly during the test. This result shows that the models have high flexural strength, but adding more PP decreases the flexural strength of the mixture.



Fig. 3. Average tensile strength of PBT/PP blends.



Fig. 4. Average elongation at break of PBT/PP blends.

The flexural strength of the PBT/PP composite exhibits properties quite similar to the tensile strength (Fig. 6). The initial flexural strength of the 100% PBT sample was 77.9 MPa and gradually decreased to 54.0 MPa when PP was mixed at 25%. Similar to tensile strength, flexural strength decreases with increasing PP content. The weak association between PP and PBT phases can explain this. A similar result was also reported by Ignaczak et al. [12]. The results show that when mixing the PBT/PP mixture with the respective ratios of 60/40, 50/50, and 40/60, the percentage increase of PP in the mix significantly reduces the flexural strength.

The modulus of elasticity when bending samples of the PBT/PP composite decreased rapidly from 2105.9 MPa (100% PBT sample) to 1792.8 MPa (PBT/15%PP sample), then increased but not much to 1825.0 MPa at 20% PP ratio and continued to decrease 1723.2 MPa (sample PBT/25%PP) (Fig. 7).



Fig. 5. Flexural stress-strain curves of (*1*) 100% PBT and PBT/PP blends: (*2*) 5PP, (*3*) 10PP, (*4*) 15PP, (*5*) 20PP, (*6*) 25PP.

The results show that the flexural modulus decreases with increasing PP content. This effect is similar to that reported by other studies [12, 21]. The reduction in strength and modulus values when mixing more PP into the blends is due to the weaker mechanical properties of the PP compared to the PBT polymers. According to the supplier, the pure PP polymer has a UTS tensile strength of 35 MPa, while the pure PBT value is 60.11 MPa. Therefore, adding more PP to the PBT matrix could lead to a reduction in strength and modulus. Moreover, the impact toughness of the pure PBT sample is lower than that of the other blends, which differs from the strength and modulus of elasticity in bending. The reason could be the crystallinity and multiphase system of the blends, which help rapidly improve the impact toughness of the blends. According to Zare et al. [22], more crystallinity and a higher cold-crystallization temperature of the PP matrix were two signs of improved crystallization behavior in the toughened blends compared to the raw PP resin. The impact strength of a hardened multiphase system depends on the size, distribution, dispersion state, and degree of phase adhesion between the dispersed particles and matrix.

Impact Strength

Figure 8 shows the average impact strength diagram of different PBT/PP blends. The results indicate that the impact strength of PBT/PP blends is higher than neat PBT. The impact strength of the 100PBT sample has the lowest impact strength of 3.7 kJ/m^2 . Adding 5% PP blends, the impact strength increased by 80.3% compared to the neat PBT sample, which is the highest value compared to other combinations. Adding more PP to the blends leads to a reduction in the tensile and flexure strengths. However, the impact strength increases when adding PP to the blends. The impact strength improvement is significant and proves the benefits of PP in the mixes. Increasing the PP ratio from 5 to 25% results in a drop in the impact strength from 6.6 to 4.9 kJ/m². The PP particles are well dispersed in the PBT substrate, thereby improving the impact resistance of the mixture. This result is evident in the 5PP sample with the highest impact strength, showing that 5PP has the most stable crystallinity and crystal structure. However, at a higher PP percentage,







Fig. 7. Average flexural modulus of PBT/PP blend.



Fig. 8. Average impact strength of PBT/PP blends.

due to the poor compatibility between PP and PBT, adding more PP leads to a reduction in the impact strength.

Generally, the neat PP sample has the highest strength and modulus values in the bending test. Adding PP to the blend will reduce the flexure strength and flexure modulus because the two substances, PBT/PP, are incompatible due to the significant difference in polarity [19]. The incompatibility of the blends affects the crystallinity and PP molecules,

test, the results are different from those in the bending test. Addgth and stances, ing PP to the blends leads to a higher impact strength than the pure one. The PP particles are well dispersed in the PBT substrate, thereby improving the impact resistance of the mixture. The poor compatibility

reducing the intermolecular bonds in the combina-

tions [20]. Moreover, the decrease in tensile strength

could also be attributed to the difference in the vitrification temperatures of PBT and PP. In the impact



Fig. 9. XRD diagrams of (1) 100% PBT and PP/PBT blends: (2) 5PP, (3) 10PP, (4) 15PP, (5) 20PP, (6) 25PP.

between PP and PBT reduces the strength and modulus values. However, the high impact resistance of PP is an improvement in the impact strength of the blend. when increasing the PP content. In future work, we will find a suitable compatibilizer for the blends to improve the bonding between PP and PBT.

XRD and SEM Microstructures Analysis

Figure 9 presents the XRD diagrams of PP/PBT blend polymers with different PP percentages. The neat PBT sample has some high peaks from 15° to 25° , similar to the Deshpande et al. [16] report. Adding PP from 5 to 25% results in 14.1°, 16.8°, 18.6°, 21.1°, and a peak that belongs to the PP structure [17, 23]. Moreover, from 15 to 25% PP, the peaks become sharper, indicating a higher rate of crystalline. The reason for this phenomenon is the higher crystallinity of the PP compared to the PBT polymer. Typically, the crystalline rate of PP varies around 30–60%, while this value of PBT is only 13-30%. Therefore, adding more PP mostly leads to an improvement in the crystalline rate of the blend. Remarkably, the XRD peak system at 10% PP sample is lower than the other samples, indicating a lower crystalline or higher amorphous structure rate. These results could be explained by SEM figures presented in Fig. 10.

Figure 10 shows the structure of fine PP particles that scatter evenly on the PBT matrix. Adding more PP leads to its coarser size in the blends. Notably, the boundary between the PP and PBT phases is clear. This result indicated poor compatibility between them

CONCLUSIONS

This study tries to improve PBT mechanical properties by mixing it with 5-25% PP. The report reveals that the impact strength values of the PBT/PP blends are 3.7, 6.6, 5.6, 5.2, 5.1, and 4.9 kJ/m², corresponding to 0, 5, 10, 15, 20, and 25% PP. The results show that the impact strength of PBT/PP blends is higher than neat PBT. Compared to neat PBT, the impact strength in the 5% PP sample is at its highest level of 80.3%. The tensile strength values of the blended samples are 60.1, 42.5, 39.0, 35.5, 34.3, and 31.9 MPa, corresponding to 0, 5, 10, 15, 20, and 25% PP. Furthermore, as the PP content increases, the flexural strength decreases. The microstructure of the blends reveals that spherical PP particles were interwoven into the PBT substrate, but there is no evidence of interphase adhesion. The microstructure of the blends reveals that the crystal structure is most stable at 5%, and the more PP, the more unstable the structure is. PBT/PP blends with 5% PP significantly improve impact strength compared to neat PBT while conserving good tensile strength, allowing them to be used



PBT/20%PP

PBT/25%PP

Fig. 10. SEM microstructure of PP/PBT blends: (a) PBT/5%PP, (b) PBT/10%PP, (c) PBT/15%PP, (d) PBT/20%PP, (e) PBT/25%PP.

widely. The above study found that the PBT/5%PP blend has the best mechanical properties and meets the initial goal.

ACKNOWLEDGMENTS

We acknowledge the Ho Chi Minh City University of Technology and Education, and Material Testing Labora-

POLYMER SCIENCE, SERIES A Vol. 66 No. 1 2024

tory. They gave our research team an opportunity to join

FUNDING

their team and access the laboratory and research machines.

This work was supported by ongoing institutional funding. No additional grants to carry out or direct this particular research were obtained.

CONFLICT OF INTEREST

The authors of this work declare that they have no conflicts of interest.

REFERENCES

- S. Krause, "Polymer–Polymer Compatibility," in *Polymer Blends*, Ed. by D. R. Paul and S. Newman (Academic Press, 1978).
- 2. N. T.-H. Pham, Polym. Sci., Ser. A 63, 800 (2021)
- 3. N.-T. Tran and N. T.-H. Pham, Int. J. Polym. Sci. 2021, 7635048 (2021)
- C. Koning, M. Van Duin, C. Pagnoulle, and R. Jerome, Prog. Polym. Sci. 23, 4 (1998).
- B. Mu, Z. Y. Jiang, B. P. Yang, J. F. Cui, X. Wang, J. H. Guo, X. M. Bao, and L. Chen, Polym. Eng. Sci. 59, 3 (2019).
- X. Y. Yao, X. Y. Tian, X. Zhang, K. Zheng, J. Zheng, R. X. Wang, S. H. Kang, and P. Cui, Polym. Eng. Sci. 49, 4 (2009).
- S.-J. Kim, B.-S. Shin, J.-L. Hong, W.-J. Cho, and C.-S. Ha, Polymer 42, 9 (2001).
- Y. Tan, X. Wang, and D. Wu, J. Reinf. Plast. Compos. 34, 21 (2015).
- D. G. Seong, C. Kang, S. Y. Pak, C. H. Kim, and Y. S. Song, Composites, Part B 168, 218 (2019).
- 10. H. Schmalz, V. Abetz, R. Lange, and M. Soliman, Macromolecules **34**, 4 (2001).
- 11. E. Gubbels, L. Jasinska-Walc, D. H. Merino, H. Goossens, and C. Koning, Macromolecules **46**, 10 (2013).

- 12. W. Ignaczak, K. Wiśniewska, J. Janik, and M. El Fray, Polish J. Chem. Technol. **17**, 3 (2015).
- 13. P. Hietaoja, M. Heino, T. Vainio, and J. Seppl, Polym. Bull. **37**, 3 (1996).
- K. Larsen Børve, H. K. Kotlar, and C.-G. Gustafson, J. Appl. Polym. Sci. 75, 3 (2000).
- 15. C.-H. Tsai and F.-C. Chang, J. Appl. Polym. Sci. **61**, 2 (1996).
- R. Deshpande, G. Naik, S. Chopra, K. A. Deshmukh, A. D. Deshmukh, and D. R. Peshwe, IOP Conf. Ser.: Mater. Sci. Eng. 346, 012004 (2018).
- 17. T. N. Moja, N. Bunekar, S. B. Mishra, T. Y. Tsai, S. S. Hwang, and A. K. Mishra. Sci. Rep. **10**, 1 (2020).
- N. T.-H. Pham and V.-T. Nguyen, Adv. Mater. Sci. Eng. 2020, 8890551 (2020).
- Y.-T. Shieh, T.-N. Liao, and F.-C. Chang, J. Appl. Polym. Sci. 79, 12 (2001).
- K. Balani, V. Verma, A. Agarwal, and R. Narayan, Biosurfaces: A Materials Science and Engineering Perspective: Physical, Thermal, and Mechanical Properties of Polymers (Wiley-American Ceramic Society, 2015).
- 21. A. Hajibaba, M. Masoomi, and H. Nazockdast, Iran. Polym. J. **25**, 2 (2016).
- 22. L. Zare, A. Arefazar, and O. M. Jazani, Iran. Polym. J. **30**, 1181–1200 (2021).
- 23. X. Wang, S. Hu, Y. Guo, G. Li, and R. Xu, Polymers 11, 12 (2019).

Publisher's Note. Pleiades Publishing remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.