POLYMER BLENDS

A Study of Poly(butylene Terephthalate) and Thermoplastic Polyurethane

Van Tron Tran^{*a*}, Nga Thi-Hong Pham^{*a*,*}, Hoang-Khang Lu^{*a*}, Ngoc Tran-Nhu Nguyen^{*a*}, Huy Huynh-Nhat Do^{*a*}, Duy-Quang Doan^{*b*}, Tran Minh The Uyen^{*a*}, and Hoang Van Huong^{*a*}

^a Faculty of Mechanical Engineering, Ho Chi Minh City University of Technology and Education, Ho Chi Minh City 71307, Vietnam

^bFaculty of High Quality, Ho Chi Minh City University of Technology and Education, Ho Chi Minh City 71307, Vietnam

*e-mail: hongnga@hcmute.edu.vn

Received June 7, 2021; revised November 25, 2021; accepted December 25, 2021

Abstract—Considered a brittle material, poly(butylene terephthalate) is inappropriate for certain applications requiring high impact resistance. In this report, we investigate improved poly(butylene terephthalate) by its blending with thermoplastic polyurethane 10% by weight prepared by injection molding technique. Tensile strength, flexural strength, and impact strength samples were tested and compared with those of the neat thermoplastic polyurethane and poly(butylene terephthalate). It is found that impact toughness is higher, while tensile and flexural strengths are lower than for the neat poly(butylene terephthalate). This study provides more insight into the effect of thermoplastic polyurethane on the mechanical properties of the blend.

DOI: 10.1134/S0965545X22020110

INTRODUCTION

Poly(butylene terephthalate), PBT, produced by polycondensation of terephthalic acid or dimethyl terephthalate with 1,4-butanediol using special catalysts, has high tensile and flexural strength, high-temperature deviation, low moisture absorption, and good size stability. PBT does well with injection molding, blow molding, or extrusion processes. Its main applications include automotive exterior components, under-hood parts, electrical components, such as connectors and fuse covers, small appliances, and pump housings [1]. However, PBT is still considered a brittle material that exhibits low shock resistance and is unsuitable for specific applications requiring high impact strength.

To strengthen the toughness of PBT, many attempts have been made by mixing it with other polymers such as high-density polyethylene, polypropylene, thermoplastic polyurethane (TPU), etc. [2–9]. TPU is a prominent candidate for blending with PBT since it has good impact strength and lowtemperature flexibility among these polymers. W. Zou et al. found that TPU is a linear copolymer. The microstructure of TPU combines soft and hard segments. The soft segments often form an elastic matrix to ensure the elasticity and low-temperature performance of the TPU. In contrast, the hard segments are a multifunctional bonding point for bonding physical reinforcement and fillers [10]. In a prior report, Hao et al. demonstrated that supplementing TPU could improve the PBT/TPU mixture [7]. Moreover, the effect of TPU on the properties of PBT/TPU blends was also evaluated [8]. Recently, a substance including clay, CNT (carbon nanotubes), and CF (carbon fiber) has been used as a third component in PBT/TPU system to enhance its properties. For instance, Tehran and coworkers added clay nanoparticles to PBT/TPU blends [11]. It was found that the PBT/TPU/clay nanocomposite system (80/20/3) had a balance between tensile and impact properties. Adding CNT to the PBT/TPU mixture significantly improved tensile and flexural strength and modulus [12]. Thermodynamic analysis showed that incorporating CNT into the PBT/TPU, advanced storage module increased the glass transition temperature [12]. Another study was conducted by Jintao Huang et al. [13] with adding the CF content to the TPU/PBT mixture. The results showed that CF could be evenly distributed in the TPU/PBT matrix and did not show an agglomeration phenomenon typical of CF. Continuously increasing the CF content caused a decrease in the impact strength of the mixture. However, its tensile and flexural strength were significantly improved.

Although many types of research on PBT/TPU blend with or without a third component have been done, the effect of TPU on the mechanical properties of PBT/TPU blend needs to be studied further to pro-

 Table 1. Compositions of the samples

Sample	TPU, wt %	PBT, wt %
PBT	0	100
10TPU	10	90
TPU	100	0

vide a more fundamental understanding. In this report, we investigate the impact toughness and the tensile and flexural strength of a PBT/TPU blend, which is prepared by adding 10 wt % TPU to PBT. Subsequently, the mechanical properties of the PBT/10 wt % TPU blend are compared with those of the neat PBT and TPU.

EXPERIMENTAL

Materials

For the experiments, we used PBT with code PBT-POCAN B4225 of Lanxess (Germany), India/China, provided by Toan Dai Hung Trading Services Co., Ltd. TPU is provided by Hau Chem Company, code TPU-F-X9190, China. The various PBT/TPU blends were prepared by weight percent, as shown in Table 1. They were dried in air at 120°C for 6 to 8 h or at 150°C for 2 to 4 h to ensure their humidity which is less than 0.03%.

Methods

The samples are tested following the Tensile test ASTM D638 standard and the Flexural test ASTM D790, using AG-X plus Shimadzu universal testing machine, Japan. Before the tensile test, the speed of the pull head is determined, samples are clamped between the ends utilizing the support. The pull head is moved upwards at a constant rate until the test piece is broken or torn.

Charpy Impact Testing samples are tested following the Charpy ISO 179-1 standard on the impact tester Tinius Olsen IT504, UK. The microstructure of samples is observed by using a high-resolution SEM microscope HITACHI S-4800, Japan. The basic parameters: the resolution of secondary electronic image—1.0 and 1.4 nm, accelerated voltage reducer—2.0 nm (1 kV, WD = 1.5 nm, conventional model); magnification: LM 20–2000 times; high magnification HM 100–800000 times.

RESULTS AND DISCUSSION

Samples of PBT, 10TPU blend and TPU prepared by injection molding are opaque (PBT, 10TPU) or slightly transparent (TPU).

The SEM micrographs of these samples are shown in Fig. 1. The neat PBT (Fig. 1a) and the neat TPU (Fig. 1c) have homogenous structures. In the case of PBT sample, there is a sheet and ripple matrix, while the TPU sample has flat and glossy surface. However, the structure of the 10TPU blend (Fig. 1b) shows two distinct regions in which the TPU phase in the dispersed droplets separates from the PBT matrix. There is no evidence for the adhesion between TPU and BPT in the blend, as the surface of the TPU particles is apparent.

Figures 2a, 2b show tensile stress-strain curves of the compositions. It is found that the neat PBT exhibits the highest strength. However, its fracture strain is the lowest one. It is due to the brittle behavior of PBT. With increasing the TPU content, the tensile strength decreases, whereas the fracture strain dramatically increases. The average toughness, the average area under the tensile stress-strain curve of these compositions also increases along with an increase in TPU content (Figs. 2c, 2d). 10TPU sample exhibits the average toughness of 87.8 kJ/m³, around ~ 25% more than that (70.2 kJ/m³) of the neat PBT (Fig. 2c), while it dramatically increases up to 28000 kJ/m³ for TPU sample due to its extremely high stretchability (Fig. 2d).

Figure 3 shows the average tensile strength of the compositions. The tensile strength of the PBT, 10TPU and TPU are 37, 28, and 10 MPa, respectively. The PBT exhibits the highest strength at the



Fig. 1. The SEM micrographs of (a) PBT, (b) 10TPU blend, and (c) TPU which were observed on the surface of the impact test samples before the test.

POLYMER SCIENCE, SERIES A Vol. 63 Suppl. 1 2021



Fig. 2. (a) Tensile stress-strain curves of (1) PBT, (2) 10TPU blend, (b) tensile stress-strain curves of TPU, and (c, d) average toughness of these compositions.

fracture strain of ~ 0.3%. With adding 10 wt % TPU to 90 wt % PBT, the tensile strength is reduced more than 20%, while the fracture strain is twice as high as that of the neat PBT. The TPU has the lowest tensile strength with the highest fracture strain of $\sim 300\%$. The results are probably due to the different glass transition temperatures of PBT and TPU, which are 53.9°C (Fig. 4a) [14, 15] and -44°C [16, 17], respectively. Since the experiments were carried out at laboratory temperature, lower than the glass transition temperature of PBT, it has the highest rigidity and lowest elongation. By contrast, the neat TPU shows opposite properties at this temperature. When blending TPU with PBT, the glass transition temperature of the composition was 44.8°C (Fig. 4b) which is lower than PBT and closer to the laboratory temperature, so the material has properties of reducing hardness, increasing elongation.

The flexural stress-strain curves of materials are shown in Fig. 5. The neat PBT has the highest strength and lowest fracture strain. Similar to the tensile properties, an increase in TPU content causes decreasing flexural strength and increasing fracture strain of materials. Figure 6 shows the values of the average flexural strength of the compositions. The flexural strength of the 10TPU blend is 56 MPa, slightly lower than that (67 MPa) of the neat PBT. This decrease is probably due to the poor compatibility between TPU and PBT,



Fig. 3. The average tensile strength of test samples.

POLYMER SCIENCE, SERIES A Vol. 63 Suppl. 1 2021



Fig. 4. Glass transition temperature T_g of (a) PBT and (b) 10TPU blend determined using the DSC curves recorded at temperatures of 30 to 300°C at a heating rate of 10 K/min.



Fig. 5. Flexural stress-strain curves of (*1*) PBT, (*2*) 10TPU, (*3*) TPU.

accompanied by the lower glass transition temperature of the 10TPU sample. However, it is much higher than the flexural strength (3.6 MPa) of the neat TPU. The results indicate that adding a small amount of TPU to the PBT matrix causes a negligible decrease in the material's flexural strength, which is similar to the recent report [18].

Figure 7 indicates the average impact strength of compositions. The impact toughness of the PBT, 10TPU blend and TPU are 3.7, 4.2, and 34.6 kJ/m², respectively. It was found that increasing the TPU content increased the impact toughness of materials. For example, with adding only 10 wt % TPU to PBT, the impact toughness of the blend is ~14% more than



Fig. 6. The average flexural strength of test samples.

that of the neat PBT, while the impact toughness of the neat TPU is the highest one. Our results are consistent with the recent research [19]. This research indicated that increasing the TPU content of the mixture led to an increase in the impact toughness of the materials.

Since PBT behaves as a brittle material at laboratory temperature, its impact strength sample fractured in a brittle manner, which was determined in a recent report [20]. This study observed the fracture surface of the impact strength samples via SEM image (Fig. 8). It was found that the fracture surface morphology of the 10TPU composition (Fig. 8b) had a pattern similar



Fig. 7. The average impact strength of test samples.



Fig. 8. SEM micrographs of PBT and the blend's impact strength samples which observed on the fracture surface of (a) PBT and (b) 10TPU blend at high magnification.

to that of the neat PBT (Fig. 8a). This result is probably due to the less TPU content in a PBT matrix and the poor adhesion.

Due to having a glass transition temperature T_g of 53.9°C [14, 15], PBT is stiff and brittle and had low impact toughness at ambient conditions. At this temperature, it exhibits tensile strength and strain of 37 MPa and ~0.3%, respectively, the flexural strength of 67 MPa, and impact strength of 3.7 kJ/m². By contrast, TPU displays high flexibility, stretchability, and shock resistance at the same temperature condition with those values of 9.9 MPa and ~300%, 3.6 MPa, and 34.6 kJ/m², respectively. This result is due to its low T_g . Blending 10 wt % TPU into PBT leads to a decrease in T_g (44.8°C) of the obtained mixture, compared to the neat PBT. However, because of the poor adhesion of the TPU phase in the PBT matrix, the

blend exhibited the mechanical properties, which slightly increased in toughness and impact strength and modest decreased in tensile strength and flexural strength, compared to those of the neat PBT.

CONCLUSIONS

The obtained results indicated that at a concentration of 10 wt %, TPU had a moderate effect on the mechanical properties of the PBT/TPU blend, particularly:

-The SEM microstructure of the PBT/10 wt % TPU blend showed a biphasic heterogeneous of PBT and TPU. There was no evidence of adhesion between the TPU phase and the substrate, as the surface of the TPU particles was entirely transparent. The microstructure showed two distinct regions, the PBT matrix and the TPU dispersed droplets.

—The blend exhibited an average toughness of 87.8 kJ/m^3 and average impact strength of 4.2 kJ/m^2 ,

around ~ 25 and $\sim 14\%$ more than those of the neat PBT, respectively.

—The average tensile strength and flexural strength of the blend (28 MPa and 56 MPa, respectively) were slightly lower than those of the PBT.

ACKNOWLEDGMENTS

We acknowledge HCMC University of Technology and Education, Dong Nhan Phat Co., Ltd and Material Testing Laboratory (HCMUTE). They gave us an opportunity to join their team, accessed the laboratory and research machines. Without their appreciated support, it would not be possible to conduct this research.

CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

DATA AVAILABILITY

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

REFERENCES

- 1. N. P. Cheremisinoff, *Condensed Encyclopedia of Polymer Engineering Terms*, 1st ed. (Butterworth-Heinemann, Woburn, 2012).
- N. T.-H. Pham and V.-T. Nguyen, Adv. Mater. Sci. Eng. 2020, 8890551 (2020).
- 3. W. Ignaczak, K. Wiśniewska, J. Janik, and M. El Fray, Pol. J. Chem. Technol. **17** (3), 78 (2015).
- Handbook of Thermoplastic Polyesters: Homopolymers, Copolymers, Blends, and Composites, Ed. By S. Fakirov (Wiley-VCH, Weinheim, 2002), Chap. 2, pp. 59–103.

- G. S. Deshmukh, D. R. Peshwe, S. U. Pathak, and J. D. Ekhe, Trans. Indian Inst. Met. 64 (1, 2), 127 (2011).
- W. Hale, H. Keskkula, and D. R. Paul, Polymer 40, 3665 (1999).
- Y. Hao, H. Yang, H. Zhang, and Z. Mo, Fibers Polym. 19, 1 (2018).
- 8. K. Palanivelu, P. Sivaraman, and M. Dasaratha Reddy, Polym. Test. **21**, 345 (2002).
- F. Gribben, G. M. McNally, A. H. Clarke, W. R. Murphy, and T. McNally, Dev. Chem. Eng. Miner. Process. 12 (1, 2), 77 (2004).
- W. Zou, J. Huang, W. Zeng, and X. Lu, ES Energy Environ. 9, 67 (2020).
- A. Chalabi Tehran, K. Shelesh-Nezhad, P. Faraji Kalajahi, and A. Olad, Mech. Adv. Compos. Struct. 4 (3), 179 (2017).
- 12. A. C. Tehran, K. Shelesh-Nezhad, and F. J. Barazandeh, J. Thermoplast. Compos. Mater. **32**, 815 (2019).
- J. Huang, H. Liu, X. Lu, and J. Qu, AIP Conf. Proc. 1713, 120003 (2016).
- J. D. Ambrósio, L. A. Pessan, H. Otaguro, M. A. Chinelatto, and E. Hage, J. Mater. Res. 16, 1220 (2013).
- 15. K. Banik and G. Mennig, Mech. Time-Depend. Mater. 9, 247 (2006).
- S. Chuayjuljit and S. Ketthongmongkol, J. Thermoplast. Compos. Mater. 26, 923 (2012).
- A. Dorigato, D. Rigotti, and A. Pegoretti, Front. Mater. 5, 58 (2018).
- 18. J. Huang, X. Lu, G. Zhang, and J. Qu, Polym. Test. 36, 69 (2014).
- 19. N. A. Ahad, Mater. Sci. Eng. 957, 012045 (2020).
- P. Du, B. Xue, Y. Song, S. Lu, J. Yu, and Q. Zheng, Polym. Bull. 64, 185 (2009).