Biodegradable fibers from poly (lactic acid)/poly (butylene succinate) blends

Kanokpun Panichsombat¹, Wisawachit Panbangpong¹, Nutthapong Poompiew¹, Pranut Potiyaraj¹

¹Department of Materials Science, Faculty of Science, Chulalongkorn University, Bangkok, 10330, Thailand

Abstract. PLA and PBS have more attention from many industries including textiles industry because there are biopolymer, which have eco-friendly. This research aims at fabricating biodegradable fibers from poly (lactic acid) (PLA) blended with poly (butylene succinate) (PBS). PLA and PBS were blended in a twin-screw extruder at various PBS contents 0 to 20 wt%. The blended fibers were then prepared by the melt spinning using a multifilament spinneret. The mechanical, thermal, and morphological properties were characterized by tensile testing, differential scanning calorimetry, and scanning electron microscopy. The results indicate that, at PBS content in PLA of 10 wt%, the obtained fiber has the optimal tensile properties. At this ratio, the blends also possesses the highest crystallinity among the blended fibers. Although these two biodegradable polymers are immiscible, but at the optimal ratio the PBS phase is well-dispersed in the PLA matrix phase.

1. Introduction

The polymer industry is growing steadily as polymers in various forms have been used as raw materials for fabrications of variety products. Despite its unique and customizable properties, the problems of plastic wastes and microplastics being accumulated in the environment due to its slow biodegradation of polymers raise a lot of concerns recently. One of their applications with high production capacities and consumer demands is for textile products which can be made from several types of polymers, especially synthetic fibers. Polyethylene terephthalate is the most consumed synthetic polymer which is used in textile products such as garments, home textiles, and automotive textiles. However, its non-biodegradability leads to a serious environmental problem. Consequently, biodegradable polymers get more and more attentions from textile industries. One of the most attractive biodegradable polymers is poly(lactic acid) (PLA) which is either chemically-synthesized from a biological materials or entirely bio-synthesized by living organisms [1]. PLA has been developed for food packaging applications as well as biomedical uses because it is non-toxic to human and also has superior mechanical properties and good thermal stability. However, PLA shows quite high glass transition temperature (Tg) that resulting in rigid and brittle fibers [2]. Thus, this study aims at reducing the rigidity and brittleness of PLA fiber via the polymer blending technique. Poly(butylene succinate) (PBS) was chosen to blend with PLA due to the fact that PBS possesses lower Tg than that of PLA thus illustrates more flexibility. PBS was blended with PLA at various ratios and successfully
fabricated into multifilament fibers. The mechanical, thermal, and morphological properties of the blended fibers were then investigated. These properties should be tuned in order to expand the utilization of biodegradable polymers in textile applications.

2. Experimental

2.1. Materials
General purpose film grade PLA pellets were supplied by NatureWorks (Ingeo™ Biopolymer, 4043D) [3]. General purpose injection grade PBS pellets were supplied by Mitsubishi Chemical Performance Polymers (BioPBS™, FZ91PB) [4].

2.2. Preparation of PLA/PBS blend fibers
PLA and PBS pellets were dried at 60 °C for 24 hours. The pellets were compounded in a twin-screw extruder at PLA: PBS various ratios (100:0, 95:5, 90:10, 85:15, 80:20). The barrel temperatures were 150 – 160 °C from feed zone to metering zone with the screw speed of 32 rpm. The obtained blend pellets were kept drying at 60°C for 24 hours. Fibers were prepared by the melt-spinning technique using a single screw extruder with barrel temperatures of 170 – 190 °C and the screw speed of 4 rpm. The molten polymers were extruded through a multifilament spinneret and were collected by a roller [1].

2.3. Characterizations
The characteristic functional groups of the samples were examined by Fourier transform infrared spectroscopy (FT-IR) with a Thermo scientific Nicolet 6700 (Tokyo, Japan). Polypyrrole coated zinc oxide nanoparticles were dried and coated with 10 nm of gold before being observed the surfaces using scanning electron microscopy (SEM: JSM-6480LV) at an acceleration voltage of 15 keV.

2.4. Testing methods
The mechanical properties of blended fibers are represented by the tensile testing where ASTM D3822 was followed using a Universal Testing Machine equipped with 5 N load cell at a crosshead speed of 5 mm/min. Samples were prepared as a single strand on C-shape paper which the length × width were 4.3 × 3.3 cm and the length of a single strand on C-shape paper was 2.3 cm. The thermal properties of blended fibers were tested by a differential scanning calorimeter (DSC) between 30 to 200 °C with the heating and cooling rate of 5 °C/min under N₂ atmosphere. The fractured cross-sectional surface morphology of the blended extrudates were analyzed by a scanning electron microscope (SEM) with an acceleration voltage of 15 kV. The samples were prepared by breaking the extrudates after immersing in liquid nitrogen for 10 min.

3. Results and discussion
According to the tensile properties of neat PLA and PLA/PBS blends shown in Figure 1, the amount of PBS affects tensile properties significantly. The addition of PBS into PLA matrix actually reduces the maximum stress of the fibers but the increase in elongation at break is found higher than neat PLA and reaching the maximum when the percentage of PBS is 10 wt.%. Thus, the addition of PBS could reduce the brittleness of PLA [5]. However, at 5 wt.% of PBS, the elongation at break decreases significantly resulting from the incompatibility between PLA and PBS phases. When the amount of PBS increases beyond 10 wt%, the elongation at break decreases probably because of PBS agglomeration.
Figure 1. (a) maximum stress (N/m²) and (b) elongations percentage of various PLA:PBS

To study the influence of incorporating PBS into PLA on morphology, DSC was employed to measure the crystallinity difference between neat PLA and PLA/PBS blends. The \( T_g \) for neat PLA and PLA/PBS blends does not change significantly while the melting temperature (\( T_m \)) slightly shift lower when PBS content increase (Figure 2.). From Table 1, the crystallinity percentage of PLA/PBS blends is reduced comparing with neat PLA because of the interruption of PLA chain rearrangement. However, at 10 wt% PBS showed more crystallinity than other bended fibers. This affects the tensile properties as explained earlier.

Table 1. Thermal properties of neat PLA and PLA/PBS blends

| Sample PLA:PBS | \( T_g \) (°C) | \( T_c \) (°C) | \( T_m \) (°C) | \( X_c \) (%) |
|---------------|---------------|---------------|---------------|--------------|
| 100:0         | 63.53         | 115.32        | 175.69        | 33.26        |
| 95:5          | 62.38         | 106.39        | 166.08        | 12.42        |
| 90:10         | 58.88         | 100.38        | 160.41        | 18.19        |
| 85:15         | 60.54         | 97.53         | 158.18        | 10.19        |
| 80:20         | 58.14         | 97.54         | 157.66        | 10.24        |
The compatibility of PBS in the PLA matrix was characterized by scanning electron microscopy (SEM). The results indicate that PBS phase obviously separates from PLA phase because the incompatibility of PLA and PBS (Figure 3). At 5 wt.% of PBS, poor distribution and agglomeration of PBS are observed while at 10 wt.% of PBS, PBS phase has regularly size and is well distributed in PLA phase. When the amount of PBS increases, PBS phase becomes larger and agglomeration is pertinent thus blocking PLA chain to rearrange causing the low crystallinity percentage and lower mechanical properties. In figure 4, the fibers surface of 5 wt.% of PBS are rough more than others because this ratio have high incompatibility. The high incompatibility and poor distribution are cause of very low mechanical properties of fibers.

Figure 3. SEM micrographs of the cross section of PLA/PBS blends (A) neat.PLA (B) 95:5 blends (C) 90:10 blends (D) 85:15 blends and (E) 80:20 blends at 5000x and scale bar is 5 μm

Figure 4. SEM micrographs of the surface of PLA/PBS fibers (A) neat.PLA (B) 95:5 blends (C) 90:10 blends (D) 85:15 blends and (E) 80:20 blends at 5000x and scale bar is 5 μm
4. Conclusion
The mechanical, thermal, and morphological properties of PLA/PBS blended fibers were thoroughly
investigated. It can be concluded that PLA/PBS blended fiber at the ratio of 90:10 gives the most
suitable properties for textile fiber application. While the blends have lower maximum stress than neat
PLA, the elongation at break increases at approximately 20% when the amount of PBS in PLA is 10 wt.%.
The DSC measurement indicates that the presence of PBS does have a remarkable impact on the
crystallinity of the PLA/PBS blends fibers. In addition, the SEM confirms that, when the amount PBS
is 10 wt.%, the PBS phase is well-distributed giving a high crystallinity percentage as well as good
mechanical properties suitable for the textile fiber application. In the future, we would to develop
mechanical and thermal properties to the blends with graphene particles [6].

References
[1] Jompang L, Thumsorn S, Wong-On J, Surin P, Apawet C, Chaichalermwong T, Kaabbuathong
 N, O-Charoen N and Srisawat N. Poly(lactic acid) and Poly(butylene succinate) Blend Fibers
 Prepared by Melt Spinning Technique. Energy Procedia. 2013; 34: 493-499
[2] Rasal RM, Janorkar AV, Hirt DE. Poly(lactic acid) modifications. Progress in Polymer Science.
 2010; 35: 338-356
[3] Ingéo™ Biopolymer on http://www.natureworksllc.com
[4] BioPBS™ on http://www.pttmcc.com
[5] Valapa R, Hussain S, Iyer PK, Pugazhenthi G and Katiyar V. Influence of graphene on thermal
degradation and crystallization kinetics behavior of poly(lactic acid). Journal of Polymer
  Research. 2015; 22 189-175
[6] Wang X, Yang H, Song L, Hu Y, Xing W and Lu H. Morphology, mechanical and thermal
properties of graphene reinforced poly(butylene succinate) nanocomposites. Composites
Science and Technology. 2011; 72 6-1