



10th Eco-Energy and Materials Science and Engineering
(EMSES2012)

Poly(lactic acid) and Poly(butylene succinate) Blend Fibers Prepared by Melt Spinning Technique

Lalita Jompang^a, Supaphorn Thumsorn^{a*}, Jessada Wong On^b, Prayoon Surin^b,
Chiyaprek Apawet^b, Tirapong Chaichalermwong^a, Narin Kaabbuathong^c,
Narongchai O-Charoen^a, and Natee Srisawat^a

^aFaculty of Engineering, Rajamangala University of Technology Thanyaburi, Pathumthani, 12110, Thailand

^bFaculty of Engineering, Pathumwan Institute of Technology, Wongmai, Pathumwan, Bangkok 10330, Thailand

^cPTT Research and Technology Institute, Phra Nakhon Si Ayutthaya 13170, Thailand

Abstract

Poly(lactic acid) (PLA) and poly(butylene succinate) (PBS) were blended in a twin screw extruder at various contents of PBS from 0-50 wt%. PLA/PBS blends were melt spun using a single screw extruder equipped with multifilaments spinnerette. The effect of PBS contents on morphology, thermal and mechanical properties of PLA/PBS blend fiber was investigated SEM micrographs indicated that the addition of PBS at 10 wt% was miscible with PLA while the other contents of PBS exhibited phase separation in the blends. The incorporation of PBS affected on the declination of crystallinity in the blends. It can be noted that the addition of PBS could enhance the elasticity of PLA/PBS blend fibers.

© 2013 The Authors. Published by Elsevier B.V. Open access under [CC BY-NC-ND license](https://creativecommons.org/licenses/by-nc-nd/4.0/).

Selection and peer-review under responsibility of COE of Sustainable Energy System, Rajamangala University of Technology Thanyaburi (RMUTT)

Keywords: poly(lactic acid); poly(butylene succinate); melt spinning; miscibility; crystallinity

1. Introduction

In the present day, biodegradable polymers have been promoting for production due to our consideration of the environment. Poly(lactic acid) (PLA) is a biodegradable polymer, which derived from renewable resources. It has now been prompt for using in plastic industries. However, there is less information for applying in textile industry. PLA is generally known as a brittle material and low crystallization polymer [1-3]. The blending with elastomeric or ductility polymers would enhance PLA

* Corresponding author. Tel.: +66 2 549 3450; fax:+66 2 549 3452.

E-mail address: gibza_p45@hotmail.com, supaphorn.t@en.rmutt.ac.th.

deformation [4-7], especially for fiber spinning. Binary and ternary blends of PLA with thermoplastic starch (TPS) and poly(butylene adipate-co-terephthalate) (PBAT) were researched by Ren et al [4]. This research showed that the mechanical properties, especially ductility of the blends exhibited dramatic improvement when increasing PBAT contents. Su et al [5] enhanced the ductility of PLA by blending with glycidyl methacrylate grafted poly(ethylene octane)

Poly(butylene succinate) (PBS) is biodegradable polymer, which derived from petroleum resources. PBS is good in elasticity and well in crystallization. The incorporation of PBS would improve PLA properties. Harada et al [6] studied on improving impact strength of PLA/PBS blends by using a reactive processing agent. Yokohara et al [7] focused on miscibility and crystallization of PLA and PBS blend. This research presented that PLA and PBS were immiscible. However, the crystallinity of PLA could enhance by nucleating ability of PBS.

PLA fiber spinning was researched by Schmuck et al [8]. They used a high speed spinning process and studied the effect of melt spinning conditions on the development of fibers structure, which further used for textile applications. Takasaki et al [9] also used high speed melt spinning process for polylactides fiber and focused on the structure and orientation of the fibers. Xu et al [10] prepared chitosan/PLA blend micro- and nano- fibers by electrospinning technique. PLA was blended with polypropylene (PP) for improved the hydrolysis resistance biodegradation and dyeability of PLA. This research was clarified by Reddy et al [11].

The aim of this research was prepared PLA/PBS blends compounding for fiber spinning in textile application. The ratio of PLA:PBS was varied from 90:10, 80:20, 70:30, 60:40 and 50:50. Morphology, thermal properties and mechanical performance of PLA/PBS blends were summarized for fabrication PLA:PBS blends fibers.

2. Experimental

2.1. Materials

Poly(lactic acid) (PLA) (grade 3051D) was supplied from NatureWorks LLC, USA. Poly(butylene succinate) (PBS) was supplied from Mitsubishi Chemical, Japan.

2.2. Compounding and Fiber Preparation

PLA and PBS were dried in an oven at temperature of 80 °C for at least 12 hours before compounding in a twin screw extruder. The barrel temperature was set at 140-165 °C with screw speed of 80 rpm. The ratios of PLA and PBS (PLA:PBS) by weight ratio in the blends were 90:10, 80:20, 70:30, 60:40 and 50:50. The neat PLA and PBS were also prepared as controls. PLA/PBS blends were compression-molded to dumbbell specimens at temperature set of 190 °C for mechanical and physical testing. The specimen designation will be referred to the content of PLA and PBS. For example, PLA50:PBS50 is PLA 50 wt% and PBS 50 wt%.

The blends were dried in the oven at temperature of 80 °C for at least 12 hours. After that the blends were melt-spun by a single screw extruder equipped with multifilaments spinnerette at barrel temperature of 190-210 °C with screw speed of 8 rpm. The melt spun PLA/PBS blends were collected.

2.3. Characterization

Thermal properties of the blend was carried out by differential scanning calorimetry (NETZSCH, DSC 200 F3) at 30-200 °C at heating and cooling rate of 10 °C/min and 5 °C/min, respectively.

Morphological analysis of fibers was performed by using a scanning electron microscope (SEM) (JEOL/JSM-5410LV) set at 15 kV. Gold was sputtered onto the specimens for electron conductivity.

Tensile Properties was investigated by an INSTRON universal testing machine (INSTRON5560). The gauge length was 100 mm. The extension rate was 10 mm/min. Five specimens were tested for each condition.

3. Results and Discussion

3.1. Thermal Properties of PLA/PBS

Blends Fig. 1 (a) shows DSC melting thermograms of PLA/PBS blends and neat materials. PLA and PBS thermograms exhibit endothermic peak around 171 °C and 113 °C, which indicated the melting temperature (T_m) of PLA and PBS, respectively. The thermograms of PLA/PBS blends present two distinguish endothermic peaks around 169-172 °C and 110-113 °C, which represented the melting temperature of PLA and PBS, respectively. From the results, it was indicated that PLA and PBS were immiscible. In addition, it can be seen that there were two melting peaks of PLA in PLA:PBS blend at 50:50 ratio. It was due to melting behavior of difference crystal size in this blend [12].

Crystallization behavior of neat polymers and the blends is shown in Fig. 1 (b). PLA was generally known in low crystallization characteristic, which can be seen a low intensity of exothermic peak of PLA as presented in Fig. 1 (b). The crystallization temperature (T_c) of PLA was around 92.8 °C. On the other hand, PBS is crystallized easier than the PLA, which was clearly observed of exothermic peak in DSC cooling cycle of PBS. The crystallization temperature (T_c) of PLA was shown at 68.3 °C. There were two exothermic peaks in PLA:PBS blends at PBS contents of 30, 40 and 50 wt% while there was only one exothermic peak of low PBS contents of 10 and 20 wt%, which the intensity of PLA crystallization peak in these blends were higher than neat PLA. It could be considered that PBS would aid in PLA crystallization. However, the intensity of PBS exothermic peak in the blends was decreased when PLA contents increased. It was attributed to PBS crystallized retardation by PLA.

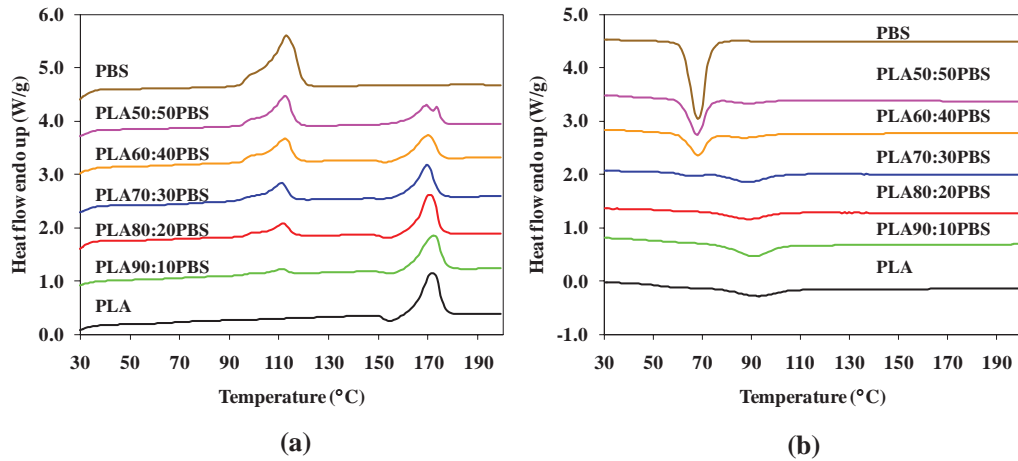


Fig 1. DSC thermograms of PLA, PBS and the blends
(a) melting endotherms (b) crystallization exotherms.

Thermal properties of neat polymers and the blends are summarized in Tables 1 and 2. Crystallinity of PLA in the blends increased when increasing PBS contents. On the contrary, crystallinity of PBS was suppressed when adding high PLA contents. From the results, it was considered that PBS would act as nucleation site for PLA in order to crystallization while PLA inhibited PBS crystallization

Table 1. Thermal properties of PLA in the blends.

PLA:PBS blend	PLA		
	T_m (°C)	T_c (°C)	Crystallinity (%)
Neat PLA	171.5	92.8	40.7
90:10	172.2	90.9	42.6
80:20	170.6	88.7	45.2
70:30	169.4	88.7	47.4
60:40	170.0	87	40.5
50:50	169.3, 173.3	89	50.8

Table 2. Thermal properties of PBS in the blends.

PLA:PBS blend	PBS		
	T_m (°C)	T_c (°C)	Crystallinity (%)
100:0	-	-	-
90:10	111.1	-	49.3
80:20	111.8	-	-
70:30	110.7	66.1	56.1
60:40	112.4	68.2	58.2
50:50	112.5	67.8	53.7
0:100	113.0	68.3	63.8

3.2. Morphology of PLA:PBS blends

Fig. 2 presents SEM photographs of PLA:PBS blends. It can be seen brittle fractured surface of PLA and PLA/PBS blends. PLA and PBS were immiscible and exhibited phase separation in these blends. However, PBS was well dispersed in PLA matrix, which would enhanced toughness in the PLA:PBS blends.

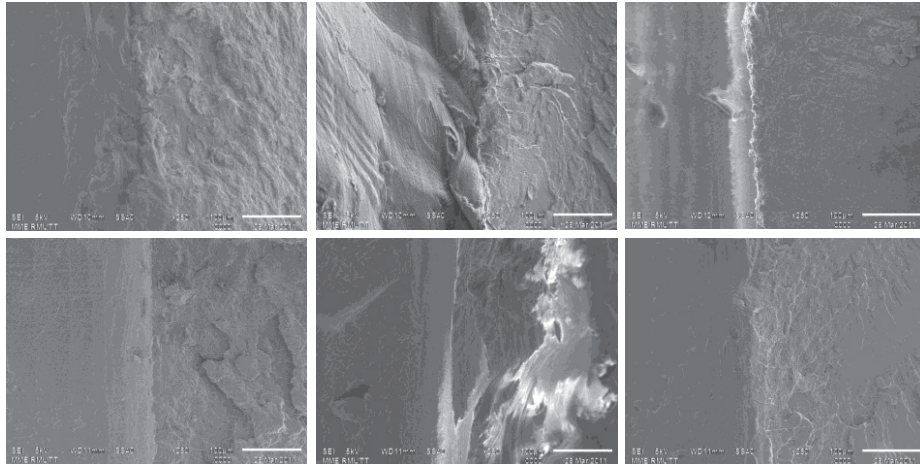


Fig 2. SEM photographs of (a) neat PLA (b) 90:10 blends (c) 80:20 blends (d) 70:30 blends (e) 60:40 blends (f) 50:50 blends (scale bar is 100 μm).

3.3. Mechanical properties of PLA:PBS blends

Fig 3. exhibits tensile modulus of PLA:PBS blends compression molded. PLA shows the highest modulus, which it generally known that PLA has been good in tensile properties [1]. The incorporation of PBS in the blends yielded the declination of tensile modulus of PLA:PBS blends due to low stiffness and ductility of PBS. Furthermore, it was attribute to less interfacial adhesion between PBS and PLA matrix, especially when increasing PBS contents. It can be note that poor adhesion between PLA and PBS as dispersed phase resulted in poor stress transferred during tensile loading [13-14]. Therefore, tensile strength of the PLA:PBS blends was also not improved as shown in Fig. 4.

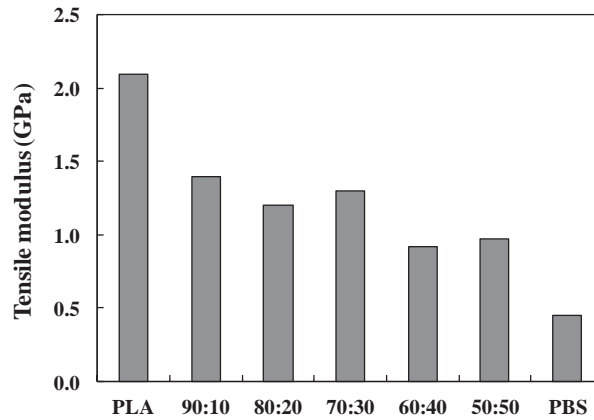


Fig.3. Tensile modulus of neat PLA, PBS and the blends.

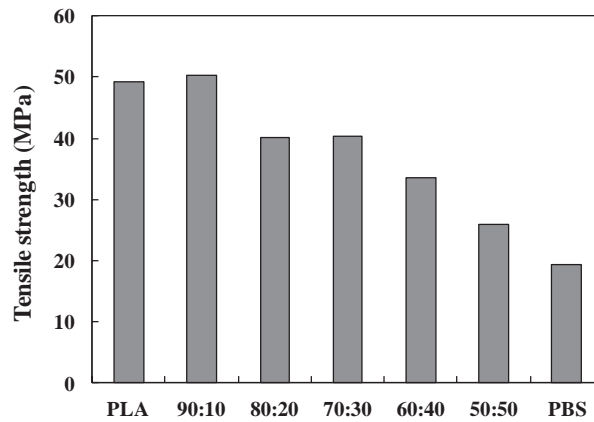


Fig 4. Tensile strength of neat PLA, PBS and the blends.

3.4.Characteristic of PLA:PBS blends fiber

Thermal properties and morphology results indicated that PLA and PBS were immiscible and exhibited phase separation. On the contrary, the tensile properties of the blend showed good tensile modulus and strength when adding PBS up to 30 wt%. Nevertheless all blends were melt-spun in a single screw extruder at take up speeds of 50, 100 and 150 m/min. Melt-spun PLA/PBS fiber could be collected at the take up speed less than 150 m/min. The blend fiber of PLA:PBS at 80:20 ratio could be spun but it

was discontinuous spinning. It was probably due to immiscible between PLA and PBS and the unfine of PBS dispersed sizes. Moreover, the increasing of PBS contents in the blends made it difficult to spinning. Therefore, PLA/PBS blends of 90:10 ratio could be spun and successfully collected at the take up speed of 50 m/min as shown in Fig. 5.



Fig 5. PLA:PBS blends fibers for melt spinning technique.

4. Conclusions

The blending of PBS in PLA enhanced the blends ductility but less stiffness, which resulting in the declination of tensile modulus of PLA:PBS blends. DSC and SEM results indicated that PLA was not miscible with PBS, especially there was a phase separation in the blends. PBS dispersed particles in the blends influenced on the discontinuous fiber spinning. It can be note that PLA:PBS blend fiber was successfully prepared from 90:10 PLA]PBS blend ratio, which would applied it for textile applications.

Acknowledgements

The authors are gratefully thanks PTT Research and Technology Institute, PTT Public Company Limited for supporting research fund.

References

- [1] Lim L-T, Auras R and Rubino M. Processing technologies for poly(lactic acid). *Prog Polym Sci* 2008; **33**: 820-52.
- [2] Garlotta D. A literature review of poly(lactic acid). *J Polym Environ* 2001; **9**: 63-84.
- [3] Ljungberg N and Wesslen B. Preparation and properties of plasticized poly(lactic acid) films. *Biomacromolecules* 2005; **6**:1789-96.
- [4] Ren J, Fu H, Ren T and Yuan W. Preparation, characterization and properties of binary and ternary blends with thermoplastic starch, poly(lactic acid) and poly(butylene adipate-co-terephthalate). *Carbo Polym* 2009; **77**:576-82.
- [5] Su Z, Li Q, Liu Y, Hu G-H and Wu C. Compatibility and phase structure of binary blends of poly(lactic acid) and glycidyl methacrylate grafted poly(ethylene octane). *Eur. Polm. J* 2009; **45**:2428-33.
- [6] Harada M, Ohya T, Iida K, Hayashi H, Hirano K and Fukuda H. Increase impact strength of biodegradable poly(lactic acid)/poly(butylene succinate) blend composites by using isocyanate as a reactive processing agent. *J. Appl. Polm. Sci.* 2007; **106**: 1813-20.
- [7] Yokohara T and Yamagushi M. Structure and properties for biomass-based polyester blends of PLA and PBS. *Eur. Polm. J.*, 2008; **44**: 677-85.
- [8] Schmack G, Jehnichen D, Vogel R, Tandler B, Beyreuther R, Jacobsen S and Fritz H-G. Biodegradable fibers spun from poly(lactide) generated by reactive extrusion. *J. Biotechnol* 2001; **86**: 151-60.
- [9] Takasaki M, Ito H and Kikutani T. Structure development of polylactides with various D-lactide contents in the high-speed melt spinning. *J.Macromol. Sci. Part B. Phys* 2003; **42**: 57-73.
- [10] Xu J, Zhang J, Gao W, Liang H, Wang H and Li J. Preparation of chitosan/PLA blend micro/nanofibers by electrospinning. *Mate. Lett.* 2009; **63**: 658-60.
- [11] Reddy N, Nama D and Yang Y. Polylactic acid/polypropylene polyblend fibers for better resistance to degradation. *Polm. Degrad. Stab* 2008; **93**: 233-41.
- [12] Park JW and Im SS. Phase behaviour and morphology in blends of poly(L-lactic acid) and poly(butylene succinate). *J. Appl. Polm. Sci* 2002; **86**: 647-55.
- [13] Huda MS, Drzal LT, Mohanty AK and Misra M. Chopped glass and recycled newspaper as reinforcement fibers in injection molded poly(lactic acid) (PLA) composites: A comparative study. *Comp. Sci. Tech* 2006; **66**: 1813-24.
- [14] Zhang JF and Sun X. Mechanical properties of poly(lactic acid)/starch composites compatibilized by maleic anhydride. *Biomacromolecules* 2004; **5**: 1446-51.