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Bond strength of laminated films between poly(lactic acid) based cast films and metalized cast polypropylene films

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Abstract. This research was carried out to study the feasibility to use biodegradable poly(lactic acid) (PLA) in flexible packaging films, which they were usually laminated with metalized cast polypropylene (MCP) film in order to reduce oxygen permeability and could be microwaveable. Since PLA was too brittle, two types of polymers; poly(butylene adipate-co-terephthalate) (PBAT) and polyethylene glycol (PEG), were blended and extruded into cast films in the weight ratios of 5, 10, and 15 wt%. The prepared blend films were laminated with MCP films using a commercial polyurethane adhesive. Mechanical and thermal properties of the blend films were investigated by tensile testing and differential scanning calorimetry. The bond strength of the laminated films was analyzed. It was found that the glass transition temperatures of PLA in the PLA/PEG blends were dramatically reduced attributed to the plasticizing effect, however, they were not changed in the PLA/PBAT blends. As a result, tensile strength of the PLA/PEG films was reduced significantly with respect to PEG content. The laminated films of PLA/PBAT blend and MCP had higher bond strength than those of PLA and MCP or PLA/PEG and MCP. The transparent PLA/PBAT 95/5 wt% was the best formula that could replace poly(ethylene terephthalate) for laminated film.

1. Introduction

Typically, flexible packaging is produced from polyethylene (PE), polypropylene (PP), and poly(ethylene terephthalate) (PET). However, these flexible packaging films require to use as laminated films for specific end-use, such as oxygen barrier and light barrier for long-time shelf life. Metalized cast polypropylene (MCP) film [1] is usually laminated with other polymer substrates for this purpose with lower cost. Also, the laminated film with MCP can be microwaveable. These polymers are non-biodegradable, so that they will be accumulated as plastic waste to cause environment problem. Use of biodegradable polymer such as poly(lactic acid) [2] has been encouraged to use. Since PLA was too brittle, two types of polymers; poly(butylene adipate-co-terephthalate) (PBAT) and polyethylene glycol (PEG), was melt blended into PLA to produce cast films. The lamination was performed by using a commercial polyurethane adhesive to bind the blend films with the MCP films. Therefore, the bond strength or adhesive strength [3] was evaluated and compared with the use of pure PLA film.

2. Experimental



2.1 Materials

Poly(lactic acid) or PLA (Ingeo™ Biopolymer 4043D, density of 1.24 g/cm³, MFI of 6 g/10 min (210°C/2.16 kg)) was purchased from NatureWork LLC, USA. Poly(butylene adipate-co-terephthalate) or PBAT (Ecoflex® F BX 7011, a density of 1.25 g/cm³, MFI of 2.7-4.9 g/10 min (190°C/2.16 kg)) was purchased from BASF, Germany. Polyethylene glycol or PEG (M_n of 5000-7000, density of 1.13 g/cm³) was purchased from Sigma-Aldrich, USA. Polyurethane adhesive was a mixture between Loctite Liofol LA 7728 (isocyanate part) and Loctite Liofol LA 6028 (hydroxyl part) in the weight ratio of 100/80. They were kindly supported by Henkel (Thailand) Company, Thailand. Metalized cast polypropylene (MCP) with thickness of 25 µm was kindly provided by Huhtamaki (Thailand) Company, Thailand.

2.2 Melt blending and cast film extrusion

PLA and PBAT pellets were dried in an air-circulating oven (Model FD53, Binder, Germany) at 60°C for 8 hours. The melt blending was carried out in a co-rotating twin-screw extruder (SHJ-25, Yongteng, China) with the weight ratios of the PLA/PBAT or PLA/PEG blend were 100/0, 95/5, 90/10, and 85/15 wt%. The barrel/die temperature was 140-190°C, and the screw-speed was 40 rpm. The extrudate was cooled in a water bath and pelletized. After thoroughly drying, the pure PLA and the blend pellets were fabricated into films using a cast film extruder (LBCR-150, Labtech Engineering Co, Thailand). The barrel/die temperature for cast film extruder was 180-210°C. The chill-roll temperature was set at 28°C. The chill-roll speed was 1.1-1.3 m/min, and the pull-out speed was 2.2-2.6 m/min. For each blend ratio, the condition was justified to obtain the cast film that had the thickness between 60 to 90 µm.

2.3 Lamination between cast films and MCP films

Loctite Liofol LA 7728 (poly-isocyanates) and Loctite Liofol LA 6028 (polyols) in the weight ratio of 100/80 were mixed and warmed in a water bath at 40°C. Then, the adhesive was poured onto the substrate surface (prepared cast film), and the Meyer rod was rolled over manually with constant speed to obtain the adhesive thickness of about 5 µm. After that, the MCP film was placed on top of the adhesive layer. A rubber roller was rolled over the laminated films several times to ensure adhesion of the laminated films.

2.4 Tensile testing of cast films

The tensile testing of blend films was carried out in accordance to ASTM-D882 using a universal testing machine (Instron 5969, Instron Engineering Corporation, USA). The specimens were cut from cast films by a cutter in the machine direction to obtain the rectangular strip of 2.54 x 15.24 cm. The gauge length was 100 mm. The load cell was 5 kN. The crosshead speed was 10 mm/min. Five trials were tested, which the averages and standard deviations were reported graphically.

2.5 Thermal analysis by differential scanning calorimetry (DSC) of blend films

The pure PLA and the blend films were cut into small pieces (5-7 mg) for thermal property analysis by a differential scanning calorimeter (DSC Pyris 1, Perkin Elmer, USA). The tests were performed in a heat-cool-reheat mode from 25 to 250°C with a heating/cooling rate of 5°C/min. The glass transition temperature, the cold crystallization temperature, and the degree of crystallinity were reported.

2.6 Bond strength testing of laminated films

The bond strength was evaluated in accordance to ASTM-F904 (comparison of bond strength or ply adhesion of similar laminates made from flexible materials). The specimen was cut from laminated films in the dimension of 2.54 x 15.24 cm. The laminated films were set to perform the 180° peel test, which a gauge length was set at 50 mm. The peel speed was set at 300 mm/min. The average load required to separate the laminated was recorded and expressed as the bond strength in Newton (N) per 25 mm.

3. Results and discussion

3.1 Tensile properties of blend films

In this study, the prepared blend films were transparent but they were less transparent than the pure PLA film. Table 1 shows film thickness of the pure PLA film, the blend films, and the laminated films. The thickness of MCPP film was constant at $25.6 \pm 1.9 \mu\text{m}$. The thickness of the PLA/PEG 85/15 wt% blend films was significant thicker than other films. Since this blend ratio had the lowest melt strength during the cast film extrusion, the chill-roll speed had to be reduced to obtain the continuous films.

Table 1. Film thickness of pure PLA film, the blend films, and the laminated films.

	PLA100	PLA95/ PBAT5	PLA90/ PBAT10	PLA85/ PBAT15	PLA95/ PEG5	PLA90/ PEG10	PLA85/ PEG15
Cast film (μm)	86.0 \pm 0.013	63.0 \pm 0.003	71.0 \pm 0.005	77.0 \pm 0.004	90.0 \pm 0.007	81.0 \pm 0.008	119.0 \pm 0.011
Adhesive layer (μm)	9.0 \pm 0.007	7.0 \pm 0.005	8.0 \pm 0.006	8.0 \pm 0.009	6.0 \pm 0.005	8.0 \pm 0.004	7.0 \pm 0.004
Laminated film (μm)	120.0 \pm 0.01	95.0 \pm 0.005	104.0 \pm 0.005	110.0 \pm 0.006	121.0 \pm 0.002	114.0 \pm 0.005	151.0 \pm 0.011

Figure 1 presents tensile modulus and tensile strength of the PLA/PBAT blend and the PLA/PEG blend films in the blend weight ratios of 0, 5, 10, and 15 wt%. The pure PLA films had tensile modulus of $4160 \pm 363 \text{ MPa}$, and tensile strength of $37.0 \pm 1.9 \text{ MPa}$. Since PBAT was more flexible than PLA, blending it into PLA reduced rigidity of the blend films with respect to PBAT content. The tensile modulus of the PLA/PBAT blend films were 3788 ± 342 , 3575 ± 379 , and $3385 \pm 310 \text{ MPa}$ for the blend weight ratio of 5, 10, and 15 wt%, respectively. For blending with PEG, the tensile modulus of the blend films were 3357 ± 369 , 3457 ± 358 , and $902 \pm 96 \text{ MPa}$ for the blend ratios of 5, 10, and 15 wt%, respectively. Although the blend film with PEG of 15 wt% was the thickest film (due to the lowest melt strength), the tensile modulus was the lowest attributed to the plasticizing effect of PEG molecules. The reduction of tensile strength after blending PLA with PBAT or PEG was in the same fashion.

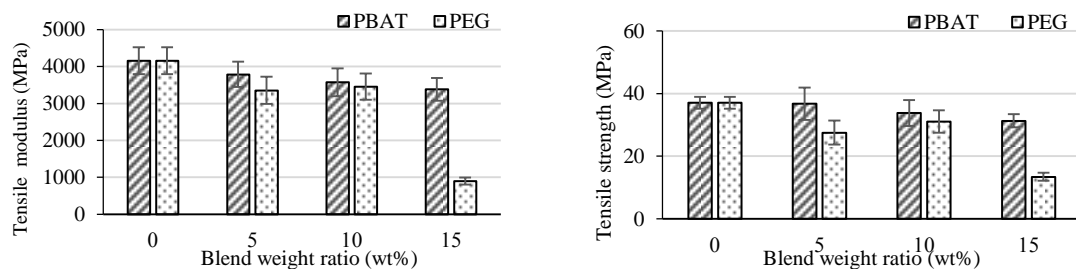


Figure 1. Tensile modulus and tensile strength of the PLA/PBAT and PLA/PEG blend films in the blend weight ratios of 0, 5, 10, and 15 wt%.

3.2 Thermal properties by differential scanning calorimetry

Figure 2 presents the glass transition temperature (T_g), the cold crystallization temperature (T_{cc}), and the degree of crystallinity (X_c) of the PLA/PBAT and PLA/PEG blend films in the blend ratios of 0, 5, 10, and 15 wt%. The blending PLA with PBAT did not change either T_g or T_{cc} and the X_c was increased slightly. In contrast, blending PLA with PEG reduced T_g significantly confirming the plasticizing effect of PEG molecules [4] that interacted with PLA molecules causing more free volume for easier molecular mobility. This influence reduced tensile modulus and tensile strength of the PLA/PEG blend films significantly. In addition, the mobile PLA molecules could recrystallize during the heating scan resulting to dramatically reduction of T_{cc} . As a result, the X_c of PLA was increased

from 8% to 29% when blending with PEG of 15 wt%. Despite of higher X_c , all blend films were still transparent.

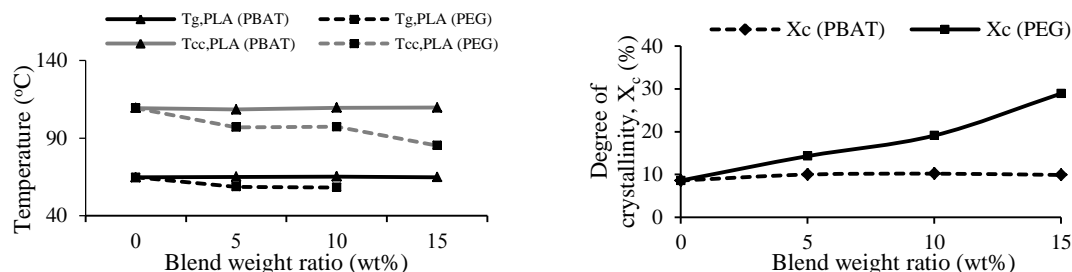


Figure 2. Glass transition temperatures (T_g), cold crystallization temperatures (T_{cc}), and the degree of crystallinity (X_c) of the PLA-based blend films in the blend weight ratios of 0, 5, 10, and 15 wt%.

3.3 Bond strength of laminated films

Figure 3 shows failure of the laminated films after the 180° peel testing. There was trace of aluminium on the cast film surface indicating that aluminium coating layer was peeled out from the MCPP film. The cast films were intact during the delamination process, while the relatively thin MCPP films were stretched and worn. Since the adhesive mixture consisted of poly-isocyanates, it could interact with the hydroxyl groups of the polyester cast films creating high bonding strength [5]. The laminated films of PLA/PEG blend and MCPP showed mostly substrate failure that the deposited aluminium layer was ripped off from the MCPP film. This implied that the bond strength was similar in all blending formula. On the other hand, the laminated films of PLA/PBAT blend and MCPP showed a mix of substrate failure (deposited aluminium layer from the MCPP film) and adhesion failure. The maximum bond strength was obtained in the laminated film between PLA/PBAT 95/5 wt% cast film and MCPP film.

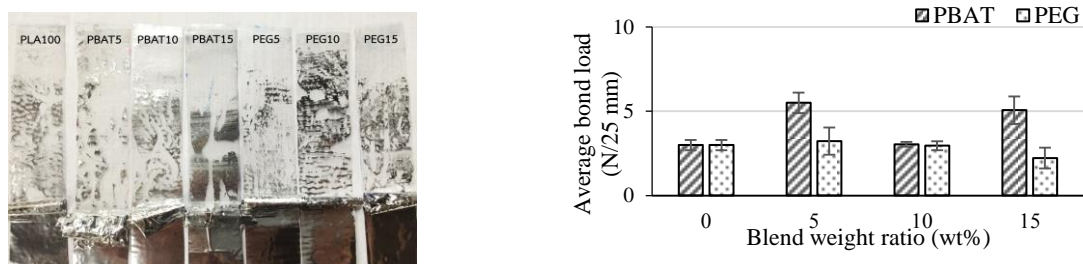


Figure 3. Images of film specimen after failure from the 180° peel test (left), and bond strength of the laminated films between PLA/PBAT or PLA/PEG and MCPP films (right).

4. Conclusion

The laminated films were successfully prepared between the PLA/PBAT or the PLA/PEG cast films and the MCPP films. By using the proper polyurethane adhesive, it was found that the transparent PLA/PBAT 95/5 wt% was the best formula that could replace PET for flexible packaging film.

5. References

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