

Practical Approach in Developing Desirable Peel–Seal and Clear Lidding Films Based on Poly(Lactic Acid) and Poly(Butylene Adipate-Co-Terephthalate) Blends

By Pitcha Liewchirakorn,¹  Duangdao Aht-Ong^{1,2} and Wannee Chinsirikul^{3,4†*}

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

²Center of Excellence on Petrochemical and Materials Technology, Bangkok 10330, Thailand

³National Metal and Materials Technology Center, National Science and Technology Development Agency, Pathum Thani 12120, Thailand

⁴National Nanotechnology Center National Science and Technology Development Agency, Pathum Thani 12120, Thailand

In this research, poly(lactic acid) (PLA) blend with poly(butylene adipate-co-terephthalate) (PBAT) were selected to fabricate peelable lidding films. In general, blending PLA with PBAT results in hazy films; however, desirable low haze films (<10%) could be achieved in this study by designing proper blend composition and cast film process under optimum conditions. Based on various blends containing PBAT ranging from 15 to 30% by weight, it could be seen that a PBAT/PLA blend of 20/80 showed desired optical and peel–seal property, which had a haze of <10% and low peel strength in an easy-peel characteristic. It was also observed that not only the blend composition but also the film thickness could influence both optical and peel–seal behaviours because the bulk morphology and surface irregularities of the films could vary by changing films' thicknesses. Thus, cast extruded pristine and PBAT/PLA (20/80) blend films of three different thicknesses (20, 35 and 50 μm) were studied. Peel–seal behaviour and optical properties of these films were examined. An I-peel test (180°) of films sealed on PLA sheet (thickness of $\sim 350\ \mu\text{m}$) with different interfacial sealing temperature illustrated failure mechanism of four types, i.e. tearing, partial tearing, cohesive and adhesive failure. Based on this study, the PBAT/PLA of 20/80 wt% films with thickness of 20 μm can be used as easy-peel lidding film sealed with PLA container. Such PBAT/PLA blend films possess a low haze of $\sim 4\%$ and a low peel strength of 8–10 N/15 mm at a broad range of interfacial sealing temperature of 76–105°C. Copyright © 2017 John Wiley & Sons, Ltd.

Received 10 January 2017; Revised 17 April 2017; Accepted 27 April 2017

KEY WORDS: poly(lactic acid); poly(butylene adipate-co-terephthalate); peel failure; peelable lidding film; haze

INTRODUCTION

Currently, the growth of world consumer on food packaging has increased every year. The experts from World Packaging Organization forecast that packaging will reach a \$1 trillion business by 2020 (Market Statistics and Future Trends in Global Packaging). The use of food packaging is increasing, resulting in increasing of packaging waste. Some biocompostable plastics should be used instead of commodity or non-compostable plastics to overcome this problem. There are many types of bioplastics that can be a good candidate for flexible packaging, especially packaging films, such as poly(lactic acid) (PLA),^{1–5} poly-3-hydroxybutyrate^{6,7} and cellulose.⁶ PLA is an aliphatic

*Correspondence to: W. Chinsirikul, National Metal and Materials Technology Center, National Science and Technology Development Agency, Pathum Thani 12120, Thailand.

E-mail: wannee@nanotec.or.th

†Present address: National Nanotechnology Center, National Science and Technology Development Agency, Pathum Thani 12120, Thailand.

thermoplastic obtained from renewable plant resources such as corn, cassava and sugar cane.⁸ Its properties on high transparency and biocompostability are interesting for packaging application. In addition, PLA is considered an economic bioplastic and readily available in the market⁹ as compared with other mentioned bioplastics. Apparently, there are many types of packaging for food and consumer products. One widely used form for fresh-cut and ready-to-eat products is lidding film sealed on container. To fabricate the proper lidding films, there are several key considerations. For example, lidding films have to provide hermetic seal on various containers, high clarity for product visibility, user friendly as easy to open and suitable gas permeation for quality retention and shelf-life extension.¹⁰ Considering PLA properties, there are some limitations for using it as a lidding film sealed on PLA container. PLA lidding film can provide good seal on PLA container, but it tends to tear or break during peeling process due to its brittleness. It is desirable that PLA lidding films must contain appropriate peel–seal ability (although it has a low seal initiation temperature of $\sim 80^{\circ}\text{C}$ ¹¹) and also still maintain good optical properties.

Peelable lidding film means that lidding film can be peeled off from the container, while easy-peel definition was explained by Zhang *et al.*¹² that the peel strength of peelable films in easy-peel range is between 2.75 and 7.95 N/15 mm. Some film manufacturers define easy peel in a range of 6–10 N/15 mm. Peelable lidding film is classified by a peeling mechanism (peeling failure) that is considered at the container surface. Peel failures of multilayer films were studied by Poisson *et al.*¹³. They revealed that three layers of polyethylene (PE)/binder/polyamide films sealed with itself highlighted differences in peel failures upon changing sealing temperatures. At low sealing temperatures, the film failed by separation (adhesive), and at high temperatures, the film failed by tearing (non-peelable). Using intermediate temperatures, the failure occurred by cohesive mechanism. Up to date, some peelable resins and films have been available in the market; however, only a few products are PLA-based materials. Dupont Co., Ltd. commercialized Appeel[®], the lidding sealant resins, which can offer excellent peelable seal characteristics to various substrates such as polypropylene (PP), PE, polystyrene, polyvinylchloride and PE terephthalate. Appeel[®] is polymer blend based on ethylene vinyl acetate (EVA), ethylene-methyl acrylate and modified polyolefin. The Dow chemical company has produced Sealution[™] peel polymer that can seal itself, PP and PE substrates. Considering commercially available peelable PLA lidding films, Innovia Films is the very first company producing PLA-based lidding film, coating PLA solution on cellophane film, called Natureflex[™]. Another peelable PLA film in the market is BioPeel[™] produced by Rockwell Solutions Ltd. In general, given technical information indicates that the available PLA films can be sealed to various types of tray materials, and each application should be tested for compatibility and performance. When considering the scientific and application viewpoints, one can see that the seal/peel performance of polymeric film on selected substrate and its controlling mechanism can be affected by several variables, including the film's properties (e.g. structure and thickness) and sealing and peeling conditions (e.g. seal temperature and peel angle). It is important to have such relevant basic understandings and practical data on the seal-peel characteristics of PLA films. This point thus leads to a major scope of this work to fabricate peelable PLA-based films, using practical and systematic manufacturing approach, and at the same time to establish key knowledge on the correlation of films' structure with peel–seal characteristics and optical property.

To add peelable function into PLA lidding film, some researchers revealed that PLA film can be modified by blending with other biopolymers. One research by Tabasi *et al.*¹⁴ studied the peel–seal properties of PLA blend films containing polycaprolactone (PCL) from 20 to 80 wt%. They found that each composition showed different seal strength and the addition of PCL to the PLA significantly improved seal properties by decreasing seal initiation temperatures of more than 15°C , as compared with the neat PLA. A patent of Chapalain¹⁵ explained that peelable PLA films can be achieved by dissolving PLA resin with at least one biopolymer such as PCL and some additives. All ingredients were mixed and dissolved in solvent prior to coating on cellophane film. Adhesive failure was found to be a peel mechanism in this work. Busch *et al.*¹⁶ fabricated biaxial PLA in two-layer or multilayer peelable films where the seal layer consisted of PLA (grade: PLA4060D) blended with other biopolymers such as poly(butylene adipate-co-terephthalate) (PBAT) with compositions between 30/70 and 80/20 (ratios of PLA/PBAT). This patent revealed that the peel mechanism of films was a cohesive failure. Another patent by Narita and co-worker¹⁷ demonstrated that peelable PLA can be

achieved by blending PLA with other biopolymers such as PBAT, PCL, poly(butylene succinate adipate) and additives. Obviously, the information in aforementioned patents did not explain the details of peel and seal behaviour thoroughly, including factors that affect optical properties of PLA lidding films.

For packaging with lidding film application, clarity of the film is of prime importance to visualize the packaged product. High clarity reflects to low haze films. Haze is used as a quality control property in the film production. Haze of lidding films typically lower than 10% is desirable for commercial use. Haze can be caused by light scattering in the bulk or from the surface. Internal light scattering from the bulk can come from materials' structure such as grain boundaries, crystalline structure¹⁸ and different phases of materials.¹⁹ Internal or bulk scattering is mostly due to changes of refractive indices that take place on the scale of the wavelength of light (400–700 nm).²⁰ Apart from the internal factor, light can scatter at the surface, interface between air and film. This implies that surface haze is also affected by surface roughness.^{21–27} Roughness on the surface of most lidding films can be caused by properties of materials and processing conditions. Extrusion or flow-induced haze and crystallization-induced haze are two main types of haze affecting surface roughness of polyolefin,^{24,28} such as PE and PP lidding films. The key property of olefin polymers influencing flow-induced irregularities haze is melt elasticity,^{26,29–32} while crystallization-induced haze is caused by type of material and also processing parameters.³³ In the present study, to fabricate practical lidding film, attentions were then paid on both the peel–seal property and clarity of the developed films.

A study by Poisson *et al.*¹³ investigated both peel–seal and optical properties of polyolefins by changing the blend compositions of two polymers. It was found that blending LDPE with EVA in the seal layer of LDPE (low-density PE)/binder/polyamide films could expand temperature range for sealing process. Besides, high content of EVA in LDPE could decrease crystallinity and surface roughness, resulting in low haze.

Some researchers revealed that there is a relation between thickness of films and peel strength.³⁴ The results from this study indicated that thickness of films might play an important role affecting light transmission and scattering of immiscible blend films. Therefore, the thickness of films should be controlled and optimized in order to obtain desired optical and peel–seal properties for peelable lidding films.

Based on the review mentioned in the preceding texts, optical and peel–seal properties of films have been mostly studied for polyolefins due to their extensive use in packaging applications. PLA and PLA blend films are obviously still in the developing stage, and basic understandings on what control their properties are considered a major challenge. This point addresses a need for a systematic study of PLA-based films, especially under the increasing demands of bioplastics as alternative to commodity plastics. Clearly, peel–seal and optical properties are key features for lidding film application. These two critical properties are affected by type of polymer used or blend compositions and film's structure and thickness as a result of processing conditions. Thus, the aims of this work were to demonstrate a practical means to develop peelable lidding films based on PLA blends with PBAT. Major attempts were made to create useful understandings on the peel–seal behaviour and optical properties of the developed single layer of PLA/PBAT blend films utilizing a common extrusion process. Optimum cast film extrusion as well as proper interfacial seal temperature to achieve easy peel, clear PLA blend films were also demonstrated. Developed films were characterized, and correlation between films' structure with peel–seal characteristics and optical property were discussed.

MATERIALS AND METHODS

Materials

The general purpose PLA4043D is an aliphatic thermoplastic having melt flow index (MFI) and density of 6 g/10 min (210°C/2.16 kg) and 1.24 g/cm³, respectively. It was obtained from Nature Works LLC. PBAT ecoflex® F Blend C1200 (MFI of 3.8 g/10 min at 190°C/2.16 kg and density of 1.25 g/cm³) is a biodegradable, aliphatic-aromatic copolyester supplied by Badische Anilin-und Soda-Fabrik.

Film fabrication

Poly(lactic acid), PBAT and PBAT/PLA blend films were prepared by extrusion through sheet die. Prior to cast extrusion, both PBAT and PLA pellets were dried at 60°C for 12 h until moisture content was lower than 250 p.p.m. by using a portable dehumidifying dryer [MJ3-50A, Matsui (Asia) Co., Ltd., Samutprakarn, Thailand]. In this work, dry blend technique was used to mix PBAT and PLA together before conveying the blend into extrusion's hopper. A single screw extrusion (HAAKE Rheomix 252, Thermo Electron Corporation, Karlsruhe, Germany) with standard metering screw (3:1), 19.05 mm diameter and L/D ratio of 25 was used to fabricate films. The die gap and air gap for all blends were set at 1.2 mm and 120 ± 10 mm, respectively. The temperature profile of the extruder from hopper to die was set at 165°C for all zones. Screw speed used was 18 r.p.m. Setting chill roll's temperature was $19 \pm 1^\circ\text{C}$ for providing high rate of cooling. To vary thickness of films, chill roll's velocities of 1.5, 2, 3 and 5 m/min were used for producing films of 60, 50, 35 and 20 μm , respectively.

Poly(lactic acid) sheet as a substrate was prepared for investigating peel–seal behaviour. Sheet thickness was 350 ± 15 μm .

Heat seal, peel test and peel surface analysis

Heat sealing process involves sealing temperature, pressure and dwell time. Sealing process parameters used were shown in Table 1. Upper and lower platen temperatures, sealing pressure and dwell time were directly set on heat sealing machine (Lako Tool SL2, Lako Tool and Manufacturing, Inc., Ohio, USA). Measured sealing temperatures between film and substrate (interfacial temperature) were analysed by contacting a thermocouple type K diameter of 0.3 mm (Fluke 568 Infrared and Contact Thermometer, Fluke Corporation, Washington, USA) with repeated five measurements.

In this system, all films were sealed on a PLA thick substrate of 350 ± 15 μm to mimic the container rim or the tray's lip. The measured sealing temperatures shown in Table 2 are between 76 and 105°C. This measurement reveals that temperature increases about 5°C with increasing upper platen temperature of 10°C from 60 to 120°C, for all films' thicknesses.

The peel strength of single-layer films was characterized by the 180°, I-peel tests according to American Society for Testing and Materials standard method F88 with a cross-head speed of 200 mm/min. Heat sealed rectangular samples with a size of 50 × 15 mm were cut along machine direction (MD). The Instron machine (5943, Instron European Headquarters, Buckinghamshire, UK) was used to record the applied force as a function of displacement. All the I-peel tests were performed at room temperature, $23 \pm 1^\circ\text{C}$, $50 \pm 2\%$ relative humidity with a load cell of 100 N. Grip distance was 25 mm. Peel strength values were the average values of five repeated measurements.

Haze, clarity and light transmission measurement

A haze metre (Haze-Gard Plus, BYK-Gardner GmbH, USA) was used in this test in accordance with American Society for Testing and Materials D-1003. Three pieces of specimen were tested to ensure reproducibility. Then, silicone oil was applied on each surface of both sides of film specimens for examining the film's internal or bulk properties by minimizing surface irregularities.

Film's morphology analysis

The morphology of the films (along MD) was investigated by using scanning electron microscopy, model: FE-SEM, Hitachi SU8030. The films were sealed between PLA sheets at 80°C of upper and

Table 1. Heat seal conditions.

Sealing parameter	Constant setting	Variation setting
Sealing time (s)	3	–
Sealing pressure (MPa)	0.102	–
Upper sealing temperature (°C)	–	60–120
Lower sealing temperature (°C)	110	–

Table 2. Settings of upper platen and lower platen temperatures versus measured sealing temperatures.

Upper platen/lower platen temperature (°C)	Measured interfacial temperature (°C)
60/110	75.9 ± 1.3
70/110	80.6 ± 0.9
80/110	84.9 ± 0.6
90/110	90.4 ± 0.8
100/110	95.1 ± 1.0
110/110	100.0 ± 1.1
120/110	105.3 ± 1.3

lower platen temperatures, pressure of 0.102 MPa and time of 3 s. The specimens were dipped into liquid N₂ for 15–20 min before fractured. The cryofractured specimens were coated with a proper thin gold layer (coating time ~45 s) to improve the sample conductivity. They were scanned under an accelerating voltage of 2.0 kV with magnification of 5000 times.

Viscosity ratio and melt density measurement

Capillary rheometer (Rosand RH2000, Malvern Instruments Ltd., Malvern, UK) was used to analyse the shear viscosity of PLA and PBAT at 165°C. Shear rate was set between ~20 and 2000 s⁻¹. Die diameter was 1 and 18 mm long.

A basic review of equations used to measure the shear stress (τ), shear rate ($\dot{\gamma}$) and shear viscosity (η) in a capillary rheometer is given in the succeeding texts:

$$\tau = \frac{\Delta P \cdot R}{2L} \quad (1)$$

$$\dot{\gamma} = \frac{4Q}{\pi R^3} \quad (2)$$

$$\eta = \frac{\tau}{\dot{\gamma}} \quad (3)$$

where ΔP is pressure drop, R is radius of capillary, L is capillary length, and Q is volumetric flow rate.

Another factor used to calculate the viscosity ratio of 20PBAT/80PLA was melt density. Melt densities of PLA and PBAT were measured by melt flow indexer (MFI-10, Lloyd Instruments Ltd, AMETEK Lloyd Instruments Ltd., West Sussex, UK). The temperature used was 165°C, compressed load of 5 kg, cut off time 60 s for PLA and 15 s for PBAT.

RESULTS AND DISCUSSION

Effect of PBAT/PLA compositions on peel strength and haze

In this research, lidding films were fabricated from PBAT/PLA blends to develop peelable lidding films in easy-peel range. Blend ratios were selected between 15/85 and 30/70 wt% beyond the ratio of aforementioned literatures. Film thickness, apparently, affected peel–seal and optical properties. At first, a film thickness of 60 μ m was selected to study the effect of PBAT/PLA compositions on peel–seal and optical properties. Then, the thickness of proper composition was varied and analysed systematically.

The peel strength of the film sealed on PLA sheet refers to the interaction between film and PLA substrate.^{35,36} The results of the I-peel test of all PBAT/PLA blend films with a thickness of 60 μ m are demonstrated in Figure 1. It can be seen that the blend ratios of PBAT/PLA affect peel strength. Peel strength tends to decrease with increasing PBAT contents. It might be because PBAT minor

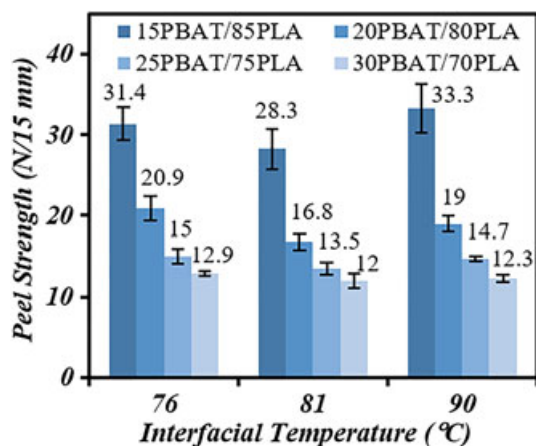


Figure 1. Peel strength of poly(butylene adipate-co-terephthalate)/poly(lactic acid) blend films with thickness of 60 μm .

phases have low interaction with PLA sheet. Interestingly, a large difference in peel strength between the blend ratio of 15/85 and 20/80 occurs, whereas the peel strength gradually decreases from the blend ratio of 20/80 to 30/70. A large drop in the peel strength of 20PBAT/80PLA film as compared with that of 15PBAT/85PLA indicates a significant influence of such 5 wt% increase of PBAT, possibly in the structure of blend films and in terms of decreased interaction between the two polymer surfaces. A very high peel strength of the 15PBAT/85PLA film about 30 N/15 mm is not as proper as that of peelable lidding film. Even though peel strengths of 20/80 and 30/70 blend films of around 20 and 15 N/15 mm, respectively, have a tendency to be used as peelable film, these values are considerably too high for easy-peel films. These results lead to another attempt (to be described in part II) to reduce the peel strength of the developed blend films by examining various film thicknesses that may affect the morphology of the film and hence its interaction to the PLA substrate.

Another property of interest is haze, and the results are shown in Figure 2. Haze or total haze increases with increasing PBAT contents. PBAT contents affect both surface and bulk haze. The proportion of surface haze can be calculated by (surface haze/total haze) * 100. The results reveal about 10–12% contribution from the surface haze, for PBAT/PLA blend films whose composition ratios are between 15/85 and 25/75. However, this effect might arise from increased melt elasticity with increasing PBAT content resulting in an increase of surface roughness. Therefore, this effect is obviously strong in a 30PBAT/70PLA film that has a proportion of surface haze approximately

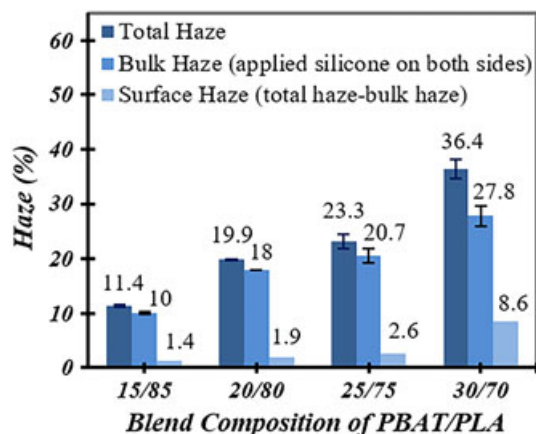


Figure 2. Transmission haze of poly(butylene adipate-co-terephthalate)/poly(lactic acid) blend films with thickness of 60 μm .

24%. For bulk haze, the major effect possibly comes from the structural irregularity or inhomogeneity based on the blends of PLA and PBAT phases.^{20,31} It is clear that the main effect of haze for PBAT/PLA blend films comes from bulk haze rather than surface haze.

The PBAT/PLA blend film of 15/85 has the lowest haze but the highest peel strength. Therefore, the proper selection of blend ratio for further investigation to improve peelable properties or reducing peel strength to achieve the desirable low easy-peel range should be 20PBAT/80PLA composition. In the following part, film thicknesses were optimized in order to obtain seal strength and % haze in desired range.

Peel-seal behaviour of different film thicknesses

All film types (PLA, PBAT and 20PBAT/80PLA) provide different peel-seal behaviours upon peeling film from the thick PLA substrates. Four types of the film's failure observed in this study are schematically illustrated in Figure 3a to d. Peel strengths of all films sealed on the PLA thick sheet (350 μm) at different seal or measured interfacial temperatures were reported in Table 3. In Table 3, PLA films demonstrate tearing and partial tearing failures (partial tearing failure at sealing temperature lower than 90°C and tearing failure at higher temperature of 90–105°C), whereas the 20PBAT/80PLA films exhibit mainly cohesive failure for a wide range of seal/interfacial temperatures. It was found that when the 20PBAT/80PLA films were sealed at higher interfacial temperatures of 100–105°C, cohesive failure changed to complete tear. In the case of PBAT film series, only adhesive failure occurred during peeling process. Tearing failure means no peel during peeling process due to high seal strength between film and substrate (or seal strength is higher than the inherent tensile strength of the film). This results in film breakage. Tearing failure shows the highest peel strength that is close to the film's breaking strength. For partial tearing failure, the samples mostly exhibit slightly lower peel strengths than those of the tearing failure. Partial tearing failure starts with peel at the beginning followed by tearing of the peel arm. For cohesive failure, the film tends to peel off from the substrate and peel advances during the test. The peel strengths of these cohesively failed samples are then averaged between 20 and 80% displacements. The peel strength is lower than those for both tearing and partial tearing failures. In this study, cohesive failure of the samples took place within the film counterpart. Another type of failure is adhesive failure, which was found in all sealed PBAT film/PLA substrate. This failure is located at the interface between the film and substrate resulting in the lowest peel strength values. There is no residue observed on both peeled surfaces of PLA thick substrate and PBAT films.

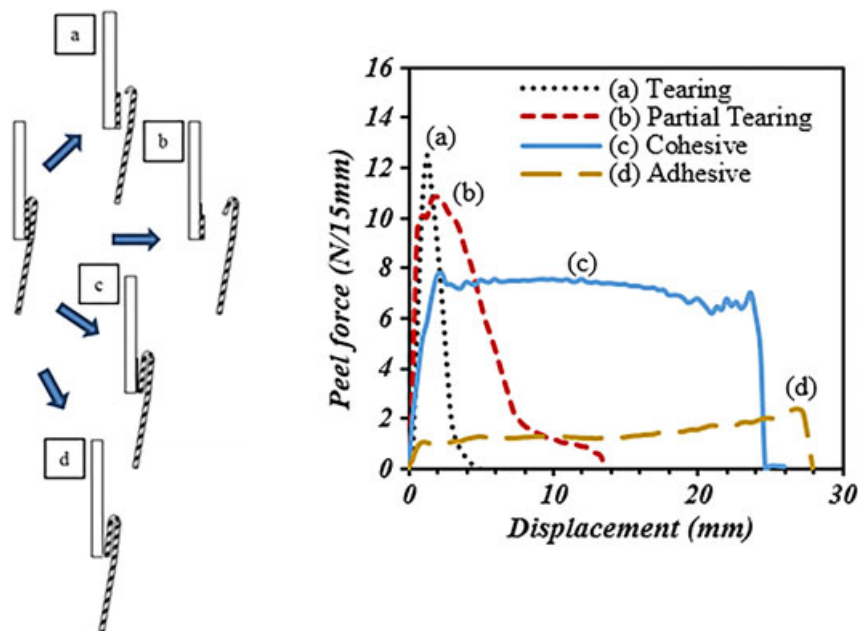


Figure 3. Typical peel failure curve (a) tearing, (b) partial tearing, (c) cohesive and (d) adhesive.

Table 3. Peel strength of PLA, 20PBAT/80PLA and PBAT films sealed on PLA sheet.

Film	Thickness (μm)	Peel Strength (N/15 mm) at Various Measured Interfacial Temperature ($^{\circ}\text{C}$)						
		75.9 $\pm$ 1.3	80.6 $\pm$ 0.9	84.9 $\pm$ 0.6	90.4 $\pm$ 0.8	95.1 $\pm$ 1.0	100.0 $\pm$ 1.1	105.3 $\pm$ 1.3
PLA	50	22.4 $\pm$ 0.5	21.9 $\pm$ 1.1	22.2 $\pm$ 0.1	25.4 $\pm$ 1.2	24.8 $\pm$ 1.6	24.8 $\pm$ 1.0	26.4 $\pm$ 0.4
	35	18.9 $\pm$ 1.1	17.5 $\pm$ 0.4	16.5 $\pm$ 1.4	18.0 $\pm$ 0.7	16.3 $\pm$ 1.5	17.9 $\pm$ 1.1	18.8 $\pm$ 1.7
	20	11.5 $\pm$ 0.7	11.0 $\pm$ 0.7	12.1 $\pm$ 1.1	12.2 $\pm$ 0.4	12.0 $\pm$ 0.6	12.2 $\pm$ 0.3	12.7 $\pm$ 0.3
20PBAT/80PLA	50	19.0 $\pm$ 0.4	20.3 $\pm$ 0.5	20.0 $\pm$ 0.3	18.8 $\pm$ 1.1	19.8 $\pm$ 0.6	21.2 $\pm$ 0.4	21.5 $\pm$ 0.5
	35	14.6 $\pm$ 0.8	14.5 $\pm$ 0.9	15.0 $\pm$ 0.4	14.4 $\pm$ 0.8	15.2 $\pm$ 1.3	15.0 $\pm$ 0.4	19.0 $\pm$ 0.7
	20	7.9 $\pm$ 0.7	8.9 $\pm$ 0.7	9.0 $\pm$ 0.3	9.4 $\pm$ 0.8	8.4 $\pm$ 0.5	9.6 $\pm$ 0.8	10.0 $\pm$ 1.1
PBAT	50	10.4 $\pm$ 0.3	10.2 $\pm$ 0.2	10.5 $\pm$ 0.0	10.8 $\pm$ 0.6	10.2 $\pm$ 0.3	10.8 $\pm$ 0.6	11.0 $\pm$ 1.0
	35	7.2 $\pm$ 0.2	7.9 $\pm$ 0.2	7.9 $\pm$ 0.1	8.2 $\pm$ 0.1	8.1 $\pm$ 0.4	8.0 $\pm$ 0.6	8.1 $\pm$ 0.1
	20	1.8 $\pm$ 0.2	1.8 $\pm$ 0.2	1.9 $\pm$ 0.4	2.5 $\pm$ 0.3	3.0 $\pm$ 0.9	3.1 $\pm$ 1.1	3.7 $\pm$ 1.0

Note: Demonstrated different peel failures by different shades

Adhesive failure	Cohesive failure	Partial tearing failure	Tearing failure
------------------	------------------	-------------------------	-----------------

To view the effect of film thickness on peel strength, the results indicate that peel strength values of neat and blend films are thickness dependent (Table 3 and Figure 4a–c). As shown in Table 3, all PBAT films had relatively low peel strengths in the range of 10.4–11.0, 7.2–8.2 and 1.8–3.7 N/15 mm for thicknesses of 50, 35 and 20 μm , respectively. PLA films reveal high peel strength among 22.4–26.4 N/15 mm, 16.3–18.9 N/15 mm and 11.5–12.7 N/15 mm for thicknesses of 50, 35 and 20 μm , respectively. For a given film type, higher peel strengths were obtained in the case of thicker film samples. This is because thicker film possesses higher total energy, as compared with thinner films under peeling process. This phenomenon can be explained by using fracture mechanic theory as shown in the following equation (Equation 4)^{37,38}:

$$G = G_a + G_p \quad (4)$$

Peel strength is a function of fracture energy or energy to create new surfaces during the peeling process. Peel initiation starts at crack front or peel front (Figure 5). The total energy used for peeling film from the substrate (peel strength) is termed G . Peel strength is the combination of adhesive fracture energy (G_a) and plastic energy dissipated in the peel arm (G_p). Thus, increasing film or peel arm thickness can result in increased dissipated energy as well as energy to bend the peel arm. Total input energy or peel strength consequently increases.

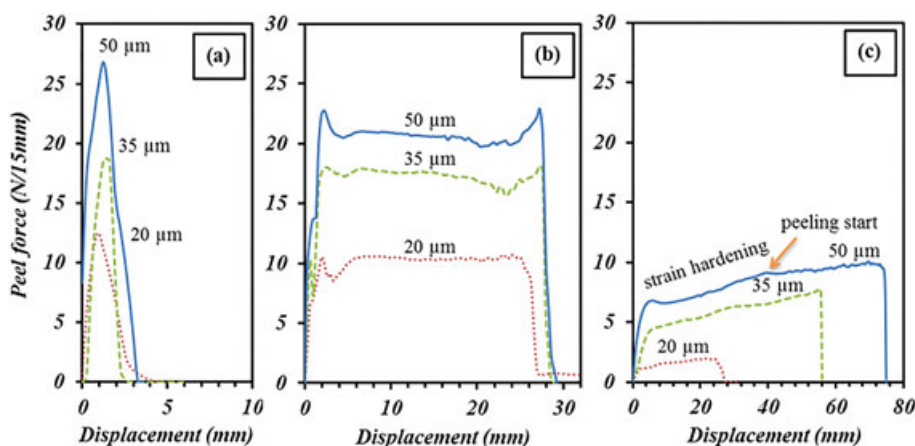


Figure 4. Peel curves of (a) poly(lactic acid) films at measured sealing temperature of $105.3 \pm 1.3^{\circ}\text{C}$ (tearing failure); (b) 20 poly(butylene adipate-co-terephthalate) (PBAT)/80 poly(lactic acid) film at measured sealing temperature is $80.6 \pm 0.9^{\circ}\text{C}$; (c) PBAT film at measured sealing temperature is $80.6 \pm 0.9^{\circ}\text{C}$.

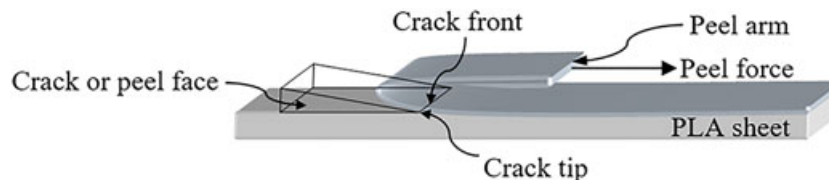


Figure 5. Schematic of 180° peeling process.

In the case of 20PBAT/80PLA films, obtained peel strength values are between those for PLA and PBAT, which are in the range of 18.8–21.5, 14.4–19 and 7.9–10 N/15 mm for thicknesses of 50, 35 and 20 μm , respectively. Incorporating PBAT into PLA apparently reduced plastic deformation energy and, in turn, decreased peel strengths of the blend film. Peel failure also changed from tearing to cohesive failure.

From peel curve results of the 20PBAT/80PLA and PBAT films having three different thicknesses and sealed with PLA thick sheet at a seal temperature of about 80°C (Figure 4 b and c), it can be seen that thicker films (50 and 35 μm) tend to be more elongated during peeling process than the thinner films (20 μm). This is possibly because thicker films have lower degree of molecular chain orientation during film fabrication process. Thus, films become more stretchable at the later peeling stage. Similarly, PBAT film with thickness of 50 μm fabricating at low take-up speed, peel arm shows high deformation, yielding and strain hardening before peeling occur because the deformation force requirement is lower than peel force. After a slight strain hardening (due to molecular orientation), PBAT films can be peeled when peel force is lower than film deformation or orientation force. PBAT films with thicknesses of 35 and 20 μm are peeled and strain hardened at the same time during peeling process. This might indicate that their peel force and deformation force are relatively equal at that time or upon a certain degree of film orientation.

Considering the effect of interfacial sealing temperature on peel strength, there is a tendency to increase peel strength by increasing sealing temperature. As illustrated in Table 3, at a given thickness, this influence is less pronounced on the tearing failure of neat PLA and becomes more pronounced for the cohesive failure of 20PBAT/80PLA films. Figures 6 and 7 highlight effect of increasing interfacial sealing temperatures on increased peel strength values of the 20PBAT/80PLA films (cohesive failure) and the PBAT film (adhesive failure) at the film thickness of 20 μm . The reasons why the heat seal strength (or peel strengths) increased with increasing sealing temperature for several types of polymers such as LDPE, LLDPE and PB-1 were studied and explained by several investigators.^{39–44} One

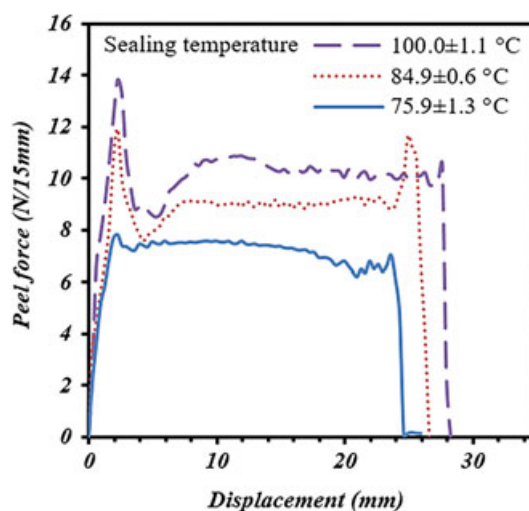


Figure 6. Peel curves of 20 poly(butylene adipate-co-terephthalate)/80 poly(lactic acid) (PLA) films (20 μm) on PLA sheet at different measured sealing temperature.

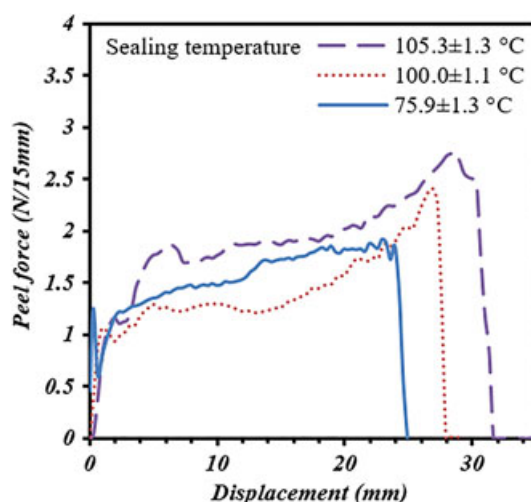


Figure 7. Peel curves of poly(butylene adipate-co-terephthalate) films (20 μm) on poly(lactic acid) sheet at different measured sealing temperature.

common explanation lies in the increase of thermal motion (micro-Brownian movement) of chain molecules with temperature resulting in deepening the original zone of diffusion causing greater peel strength.^{40,41,43,44} Additionally, some of the diffused molecules can entangle with nearby molecules; as a result, peel strength could be raised by higher degree of entanglement.^{40,43,44}

Optimized haze and morphology of 20PBAT/80PLA blend films

Haze or total haze of 20PBAT/80PLA films (as presented in Table 4) comes from bulk and surface haze. The bulk haze of the 20PBAT/80PLA blend films possibly relies on morphology of the films. In theory, the morphology of dispersed phase in polymer blends depends on many parameters, for example, draw ratio,^{45,46} viscosity ratio^{45–48} and interfacial tension between the two polymers.⁴⁶ Considering a viscosity ratio of the 20PBAT/80PLA blend, this value was calculated based on the specific composition (20% PBAT and 80% PLA by weight) and the film-processing temperature of 165°C, using a screw speed of 18 r.p.m. The obtained viscosity ratio (λ) for the 20PBAT/80PLA was ~ 1.2 , which was calculated under the specific strain rate of about 20–40 s^{-1} . In principle, this viscosity ratio tends to indicate the initial morphology of the dispersed phase (PBAT component) in the droplet form,⁴⁶ as evident in Figure 8. The original droplet structure of the minor phase in the blend can be largely affected by further drawing or extension in the film-forming process. Droplets can be altered to elongated structure (e.g. thin strips) depending upon the degree of drawing at a given process condition. It was observed by SEM that the morphology of the 20PBAT/80PLA blend films was changed upon varying draw ratios (by adjusting chill roll's speed) and correspondingly changed film thicknesses. The PBAT minor phase was deformed along with the PLA matrix upon drawing or under orientation process.³ Considering Figure 9a–c, all 20PBAT/80PLA blend films of three different thicknesses have similar morphology of oriented strips or layer-like structure along the MD. Thicker films of 50 μm contain PBAT minor phase of low aspect ratios (L/D ratio), as compared with the thinner films (35 and 20 μm). In other words, thicker films possess short and large strip/ribbon-like structures as compared with those observed in thinner films. With larger size or lower aspect ratio

Table 4. Transmission haze of 20PBAT/80PLA films with different thicknesses.

Film sample	Total haze, TH (%)	Bulk haze, BH (%)	Surface haze, SH (%)	BH/TH $\times 100$ (%)
20PBAT/80PLA 50 μm	17.2 $\pm$ 0.3	15.9 $\pm$ 0.7	1.3 $\pm$ 0.5	92.5
20PBAT/80PLA 35 μm	9.2 $\pm$ 0.4	7.8 $\pm$ 0.0	1.3 $\pm$ 0.4	85.6
20PBAT/80PLA 20 μm	4.0 $\pm$ 0.3	2.5 $\pm$ 0.3	1.5 $\pm$ 0.1	62.5

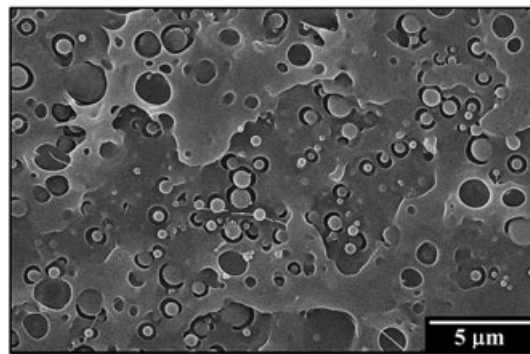


Figure 8. Morphology of 20 poly(butylene adipate-co-terephthalate)/80 poly(lactic acid) blend specimens by cryogenic fracture of melted extrudate before flowing through sheet die.

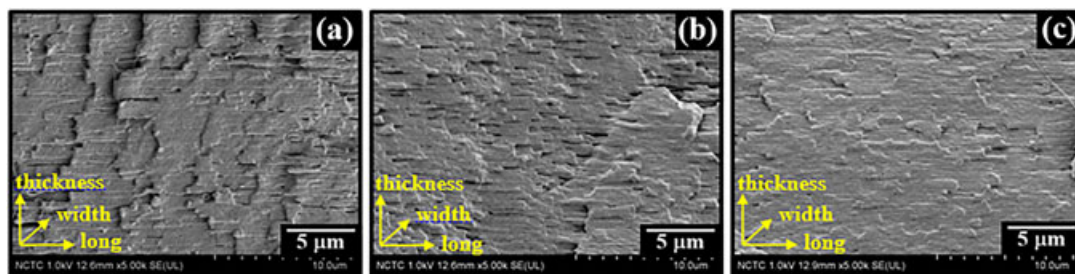


Figure 9. Morphology of films' cross-sectional along machine direction surface by cryogenic fracture of 20 poly(butylene adipate-co-terephthalate)/80 poly(lactic acid) films with different thicknesses of (a) 50 μm (b) 35 μm and (c) 20 μm .

of the minor phase in the blend film, there seems to be a higher tendency of light scattering in the system. Higher internal/bulk light scattering could give rise to higher bulk haze. Based on the current findings, there is a possibility to decrease bulk haze by reducing blend film thickness (increasing chill roll's speed in the film extrusion). By so doing, the PBAT minor phase could be elongated into fine strip-like structure, which results in lower light scattering. As illustrated in Table 4, the 20PBAT/80PLA films with thicknesses of 50, 35 and 20 μm show different total haze values of 17.2, 9.2 and 4%, respectively. The observed bulk haze is much higher in the thick blend films (15.9, 7.8 and 2.5% for film thicknesses of 50, 35 and 20 μm , respectively). Again, the resulting bulk haze correlates well with the observed morphology, revealed by SEM. Regarding the surface haze of the three 20PBAT/80PLA films of 50, 35 and 20 μm , the values are relatively the same in a range of 1.3–1.5%.

Table 5 demonstrates light transmission and clarity of the 20PBAT/80PLA blend films. Light transmission tends to increase with decreasing film's thickness. For clarity of 20PBAT/80PLA films, the results show no significant effect of thickness on film clarity.

Table 5. Light transmission and clarity of 20PBAT/80PLA films with different thicknesses.

Film sample	Light transmission, T (%)	Clarity, C (%)
20PBAT/80PLA 50 μm	83.4 ± 0.4	99.1 ± 0
20PBAT/80PLA 35 μm	88.7 ± 0.1	99.2 ± 0.1
20PBAT/80PLA 20 μm	92.1 ± 0.1	99.3 ± 0.1

CONCLUSION

Film with desired optical and peel–seal property for lidding film application could be achieved from the 20PBAT/80PLA blends. Developed films of such blend had desirable low haze of <10% and proper peel strength in an easy-peel characteristics. The haze of the 20PBAT/80PLA films had a profound effect from the internal or bulk structure as compared with the surface effect (e.g. surface irregularities). The bulk haze of the 20PBAT/80PLA blend films could be effectively reduced by decreasing film thickness. In practice, thin films could be fabricated at high chill roll's speed where all required market properties should be optimized. In fact, not only the blend compositions but also the films' structure and thickness played a vital role in controlling peel–seal behaviour and film clarity. Developed 20PBAT/80PLA film with a thickness of 20 μm exhibited practical properties, including a haze of ~4% and a peel strength of ~8–10 N/15 mm at a broad range of the interfacial sealing temperature of 76–105°C. This blend film had PBAT minor phase in the fine, oriented, strip-like structure, as revealed by SEM. When the sealed 20 μm film of 20PBAT/80PLA was peeled from the PLA substrate, cohesive failure was apparent during the peeling process.

ACKNOWLEDGEMENTS

The authors gratefully acknowledge financial support for this research from the Thailand Research Fund (TRF) (fund no. PHD56I0020). A part of the support was provided by AJ Plast Public Company Limited. The authors are also thankful for technical supports through uses of instruments and characterization facilities at National Metal and Materials Technology Center (MTEC), National Science and Technology Development Agency (NSTDA) and Ministry of Science and Technology, Thailand.

REFERENCES

1. Mistriotis A, Briassoulis D, Giannoulis A, D'Aquino S. Design of biodegradable bio-based equilibrium modified atmosphere packaging (EMAP) for fresh fruits and vegetables by using micro-perforated poly-lactic acid (PLA) films. *Postharvest Biology and Technology* 2016; **111**: 380–389 <https://doi.org/10.1016/j.postharvbio.2015.09.022>.
2. Ahmed J, Varshney SK. Polylactides—chemistry, properties and green packaging technology: a review. *International Journal of Food Properties* 2011; **14**: 37–58 <https://doi.org/10.1080/10942910903125284>.
3. Chinsirikul W, Rojsatean J, Hararak B et al. Flexible and tough poly(lactic acid) films for packaging applications: property and processability improvement by effective reactive blending. *Packaging Technology and Science* 2015; **28**: 741–759 <https://doi.org/10.1002/pts.2141>.
4. Ferri JM, Fenollar O, Jorda-Vilaplana A, García-Sanoguera D, Balart R. Effect of miscibility on mechanical and thermal properties of poly(lactic acid)/ polycaprolactone blends. *Polymer International* 2016; **65**: 453–463 <https://doi.org/10.1002/pi.5079>.
5. Plackett DV, Holm VK, Johansen P, Ndoni S, Nielsen PV, Sipilainen-Malm, et al. Characterization of l-poly(lactide) and l-poly(lactide)–polycaprolactone co-polymer films for use in cheese-packaging applications. *Packaging Technology and Science* 2006; **19**(1), 1–24. <https://doi.org/10.1002/pts.704>.
6. Seoane IT, Fortunati E, Puglia D, Cyrus VP, Manfredi LB. Development and characterization of bionanocomposites based on poly(3-hydroxybutyrate) and cellulose nanocrystals for packaging applications. *Polymer International* 2016; **65**: 1046–1053 <https://doi.org/10.1002/pi.5150>.
7. Modi SJ, Cornish K, Koelling K, Vodovotz Y. Fabrication and improved performance of poly(3-hydroxybutyrate-co-3-hydroxyvalerate) for packaging by addition of high molecular weight natural rubber. *Journal of Applied Polymer Science* 2016; **133**: n/a–n/a <https://doi.org/10.1002/app.43937>.
8. Lahtinen K, Kotkamo S, Koskinen T, Auvinen S, Kuusipalo J. Characterization for water vapour barrier and heat sealability properties of heat-treated paperboard/polylactide structure. *Packaging Technology and Science* 2009; **22**(8): 451–460 <https://doi.org/10.1002/pts.869>.
9. Kale G, Auras R, Singh SP. Comparison of the degradability of poly(lactide) packages in composting and ambient exposure conditions. *Packaging Technology and Science* 2007; **20**(1): 49–70 <https://doi.org/10.1002/pts.742>.
10. Pettersen MK, Bardet S, Nilsen J, Fredriksen SB. Evaluation and suitability of biomaterials for modified atmosphere packaging of fresh salmon fillets. *Packaging Technology and Science* 2011; **24**(4): 237–248 <https://doi.org/10.1002/pts.931>.
11. Jamshidian M, Tehrani EA, Imran M, Jacquot M, Desobry S. Poly-lactic acid: Production, applications, nanocomposites, and release studies. *Comprehensive Reviews in Food Science and Food Safety* 2010; **9**: 552–571 <https://doi.org/10.1111/j.1541-4337.2010.00126.x>.
12. Zhang J, Manias E, Polizos G et al. Tailored polyethylene nanocomposite sealants: Broad-range peelable heat-seals through designed filler/polymer interfaces. *Journal of Adhesion Science and Technology* 2009; **23**: 709–737.

13. Poisson C, Hervais V, Lacrampe MF, Krawczak P. Optimization of pe/binder/pa extrusion blow-molded films. I. Heat sealing ability improvement using pe/eva blends. *Journal of Applied Polymer Science* 2006; **99**: 974–985. <https://doi.org/10.1002/app.22405>.
14. Tabasi RY, Najarzadeh Z, Ajji A. Development of high performance sealable films based on biodegradable/compostable blends. *Industrial Crops and Products* 2015; **72**: 206–213 <https://doi.org/10.1016/j.indcrop.2014.11.021>.
15. Chapalain F. Packaging article. US Patent; 2012.
16. Busch D, Klein D, Schmitz B. Peelable sealable PLA film. US Patent; 2009.
17. Narita J, Sawai T, Takeishi I, Itoh S. Aliphatic polyester compositions, film made thereof and laminates thereof. US Patent; 2003.
18. Nalwa HS. *Handbook of Nanostructured Materials and Nanotechnology: Synthesis and Processing. Volume 1*. Academic Press: London, 2000; 161.
19. Johnson MB, Wilkes GL, Sukhadia AM, Rohlfing DC. Optical properties of blown and cast polyethylene films: surface versus bulk structural considerations. *Journal of Applied Polymer Science* 2000; **77**: 2845–2864 [https://doi.org/10.1002/1097-4628\(20000923\)77:13<2845::AID-APP6>3.0.CO;2-7](https://doi.org/10.1002/1097-4628(20000923)77:13<2845::AID-APP6>3.0.CO;2-7).
20. International A, Lampman S. Characterization and failure analysis of plastics: A S M International, 2003; 43–44.
21. Wang L, Kamal MR, Rey AD. Light transmission and haze of polyethylene blown thin films. *Polymer Engineering & Science* 2001; **41**: 358–372 <https://doi.org/10.1002/pen.10734>.
22. Patel R, Ratta V, Saavedra P, Li J. Surface haze and surface morphology of blown film compositions. *Journal of Plastic Film & Sheeting* 2005; **21**: 217–231 <https://doi.org/10.1177/8756087905057247>.
23. Smith PF, Chun I, Liu G et al. Studies of optical haze and surface morphology of blown polyethylene films using atomic force microscopy. *Polymer Engineering & Science* 1996; **36**: 2129–2134 <https://doi.org/10.1002/pen.10609>.
24. Khan MB, Keener C. An investigation of the spatial and optical behavior of biaxially drawn pet film. *Polymer Engineering & Science* 1996; **36**: 1290–1299 <https://doi.org/10.1002/pen.10523>.
25. Chen X-Y, Xiang M. Formulating of a novel polyolefin hazy film and the origins of haze thereof. *Polymer Bulletin* 2010; **64**: 925–937 <https://doi.org/10.1007/s00289-009-0229-6>.
26. Sukhadia AM, Rohlfing DC, Johnson MB, Wilkes GL. A comprehensive investigation of the origins of surface roughness and haze in polyethylene blown films. *Journal of Applied Polymer Science* 2002; **85**: 2396–2411 <https://doi.org/10.1002/app.10874>.
27. Lin YJ, Dias P, Chum S, Hiltner A, Baer E. Surface roughness and light transmission of biaxially oriented polypropylene films. *Polymer Engineering & Science* 2007; **47**: 1658–1665 <https://doi.org/10.1002/pen.20850>.
28. Andreassen E, Larsen Å, Nord-Varhaug K, Skar M, Øysæd H. Haze of polyethylene films—effects of material parameters and clarifying agents. *Polymer Engineering & Science* 2002; **42**: 1082–1097 <https://doi.org/10.1002/pen.11014>.
29. Huck ND, Clegg PL. The effect of extrusion variables on the fundamental properties of tubular polythene film. *Polymer Engineering & Science* 1961; **1**: 121–132 <https://doi.org/10.1002/pen.760010307>.
30. Bafna A, Beaucauge G, Mirabella F, Skillas G, Sukumaran S. Optical properties and orientation in polyethylene blown films. *Journal of Polymer Science Part B: Polymer Physics* 2001; **39**: 2923–2936 <https://doi.org/10.1002/polb.10049>.
31. Bheda JH, Spruiell JE. The effect of process and polymer variables on the light transmission properties of polypropylene tubular blown films. *Polymer Engineering & Science* 1986; **26**: 736–745 <https://doi.org/10.1002/pen.760261105>.
32. Pucci MS, Shroff RN. Correlation of blown film optical properties with resin properties. *Polymer Engineering & Science* 1986; **26**: 569–575 <https://doi.org/10.1002/pen.760260808>.
33. Ashizawa H, Spruiell JE, White JL. An investigation of optical clarity and crystalline orientation in polyethylene tubular film. *Polymer Engineering & Science* 1984; **24**: 1035–1042 <https://doi.org/10.1002/pen.760241307>.
34. Nase M, Großmann L, Rennert M, Langer B, Grellmann W. Adhesive properties of heat-sealed EVAc/PE films in dependence on recipe, processing, and sealing parameters. *Journal of Adhesion Science and Technology* 2014; **28**: 1149–1166 <https://doi.org/10.1080/01694243.2014.889431>.
35. Wagner JR. *Multilayer Flexible Packaging: Technology and Applications for the Food, Personal Care, and Over-the-Counter Pharmaceutical Industries*, Vol. 10. Elsevier: Oxford, 2009.
36. Bailey WJ. *Contemporary Topics in Polymer Science*. Springer Science & Business Media: New York, 2012.
37. da Silva LFM, Öchsner A, Adams RD. *Handbook of Adhesion Technology*. Springer: Berlin Heidelberg, 2011; 493–494.
38. Ebnesajjad S. *Plastic films in food packaging: materials, technology and applications*. Elsevier: Boston, 2012; 34–35.
39. Pocius AV, Dillard D. *Adhesion science and engineering: surfaces, chemistry and applications*. Elsevier: Amsterdam, 2002; 509.
40. Mueller C, Capaccio G, Hiltner A, Baer E. Heat sealing of lldpe: relationships to melting and interdiffusion. *Journal of Applied Polymer Science* 1998; **70**: 2021–2030 [https://doi.org/10.1002/\(SICI\)1097-4628\(19981205\)70:10<2021::AID-APP18>3.0.CO;2-A](https://doi.org/10.1002/(SICI)1097-4628(19981205)70:10<2021::AID-APP18>3.0.CO;2-A).
41. Nicastro LC, Paik JS, Keown RW, Metzner AB. Change in crystallinity during heat sealing of cast polypropylene film. *Journal of Plastic Film & Sheeting* 1993; **9**: 159–167 <https://doi.org/10.1177/875608799300900207>.
42. Nase M, Großmann L, Rennert M, Langer B, Grellmann W. Adhesive properties of heat-sealed evac/pe films in dependence on recipe, processing, and sealing parameters. *Journal of Adhesion Science and Technology* 2014; **28**: 1149–1166.
43. Su JF, Wang XY, Huang Z et al. Heat-sealing properties of soy protein isolate/poly(vinyl alcohol) blend films: effect of the heat-sealing temperature. *Journal of Applied Polymer Science* 2010; **115**: 1901–1911 <https://doi.org/10.1002/app.31150>.
44. Yuan CS, Hassan A, Ghazali MIH, Ismail AF. Heat sealability of laminated films with lldpe and ldpe as the sealant materials in bar sealing application. *Journal of Applied Polymer Science* 2007; **104**: 3736–3745 <https://doi.org/10.1002/app.25863>.
45. Gonzalez-Nunez R, Favis BD, Carreau PJ, Lavallée C. Factors influencing the formation of elongated morphologies in immiscible polymer blends during melt processing. *Polymer Engineering & Science* 1993; **33**: 851–859 <https://doi.org/10.1002/pen.760331310>.

46. Thomas S, Grohens Y, Jyotishkumar P. *Characterization of Polymer Blends: Miscibility, Morphology and Interfaces*. Wiley: Weinheim, 2014.
47. Heino MT, Hietaoja PT, Vainio TP, Seppälä JV. Effect of viscosity ratio and processing conditions on the morphology of blends of liquid crystalline polymer and polypropylene. *Journal of Applied Polymer Science* 1994; **51**: 259–270 <https://doi.org/10.1002/app.1994.070510208>.
48. Antunes CF, van Duin M, Machado AV. Morphology and phase inversion of EPDM/PP blends—effect of viscosity and elasticity. *Polymer Testing* 2011; **30**: 907–915 <https://doi.org/https://doi.org/10.1016/j.polymertesting.2011.08.013>.