Evaluation of the Influence of Processing Conditions on the Peel Properties of PE-LD/iPB-1 Peel Systems

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Abstract. The specific peel system low-density polyethylene/isotactic polybutene-1 (PE-LD/iPB-1) was investigated in this study. The required peel force to open the peel film package depends, among others, on the processing conditions of the blowing process. Commercial products have a constant amount of iPB-1. Thus, a defined peel force can be adjusted by varying the processing conditions. The standard peel system PE-LD with 6 m.-% iPB-1 was investigated in dependence on the processing conditions. The most important processing parameters of the processing line (extruder, die head, tube forming area) were varied and their influence on the peel properties, which can be considered as a special kind of fracture properties, were examined. The new developed parameter *time of solidification* was used to characterize the cooling behavior.

Introduction

A common method to produce films is the production by blowing process. It is already known from different studies about polyethylene films over the past 40 years, that the processing conditions have a strong influence on the material properties [1–14]. The extruder temperatures influence the flow of the melt within the extruder and, therefore, they have to be considered for any kind of blend compositions [4]. Another important processing parameter is the size of the die gap, which changes indirectly the draw-down ratio (DDR) in case of constant film thickness [2,5,9,10,13]. It can be stated that the haze [13] and the dart impact [9] increase with increasing size of the die gap. The DDR indicates the drawing in machine direction (MD), whereas the blow-up ratio (BUR) indicates the drawing in transversal direction (TD) of the film bubble [1,2,5,9,12]. The DDR can be changed using different take-up speeds (TUS) [13]. The TUS, in turn, influences the frost line hight (FLH), at which the molten polymer solidifies by crystallization. Note, that the most important processing parameter is the cooling of the blown film up to the frost line [2–6,8,13]. Rapid crystallization will provide smaller crystalls, and so, the film will be clearer, apparent in a low haze, and have a smoother surface, apparent in a high gloss [11]. The FLH is often used to get a quantification of the cooling behavior. However, in case of different BURs (BUR 1 > BUR 2), the FLH could be at the same level due to varying of different processing conditions and, nevertheless, the cooling behavior is not the same. In case of BUR 1, the partially molten film needs more time to solidify, because of the larger silhouette, than in case of BUR 2.

Regarding the fact, that the processing conditions influence the material properties, it is expected that they also influence the peel properties. The literature is very rare concerning this content [12,15,16] and, even more, for the peel system PE-LD/iPB-1 [17]. A slight increase of the peel force with enhanced BUR can be obsereved for linear PE-LD/polyisobutylene films [12]. An increased peel force with increased thickness of the film is pointed out for the peel system PE-LD/iPB-1 [17].

The present study focus on the specific peel system PE-LD/iPB-1. PE-LD/iPB-1 peel systems are often used in peel films for flexible packages in the foodstuff sector, as well as in the medical sector [17,18]. In practice, a defined seal area between two films is produced by application of heat and pressure using a sealing device, to close the package. These peel film packages can be easily re-opened by hand in a safe and comfortable manner. The easy-opening of the polyethylene/ polybutene-1 peel system is based on the breakdown of the interface between the incompatible thermoplastics polyethylene and polybutene-1 within the seal area. The reqired peel force to open the peel film package depends on the processing conditions of the sealing device [17], on the composition and properties of the used materials [19–21], and on the processing conditions of the blowing process, which were only marginally investigated [17]. Thus, the literature contains no profound results to the dependence of the peel force on processing conditions of the blowing process (herein after referred to as *processing conditions*) for the peel system PE-LD/iPB-1.
In the present study, it is intended to investigate the influence of several processing conditions along the processing line on the fracture/peel properties of the specific peel system PE-LD/iPB-1, in particular on the peel force with respect to the supermolecular structure of the blown film. Finally, morphology-property correlations will be established for this peel system.

Experimental

Materials. Peel films of PE-LD with 6 m.-% iPB-1 were investigated in the present study. This blend composition can be considered as optimum concerning reproducibility and economics of the peel process, as mentioned before [19,20]. The blend components PE-LD and iPB-1 were commercial polymers, provided by LyondellBasell (Germany). The density of the PE-LD is 0.923 g cm^{-3}, and the melt-flow index is 0.73 g $(10\ min)^{-1}$. The melt-flow index of the iPB-1 is 1 g $(10\ min)^{-1}$. The polymers were blended in a single-screw extruder (Collin, Germany) and processed to films by blowing process at Orbita-Film GmbH (Germany) using lab-equipment.
The films were produced using a set of standard processing conditions, which were listed in table 1.

Table 1: Standard processing conditions of peel film production and limitations of their variations.

	Processing Conditions	Standard Values	Variation Limits
Extruder	mix and shear element	with maddock mix and shear element	with and without
	screw speed	100 min^{-1}	60 … 120 min^{-1}
	temperatures of the extruder zones	140-160-180-180-180 °C	130-150-170-170-170 °C … 180-200-220-220-220 °C
Die Head	temperatures of the die head zones	180-180-180-180 °C	160-160-160-160 °C … 220-220-220-220 °C
	die gap size	0.8 mm	0.8 … 1.8 mm
Tube Forming Area	time of solidification	1.8 s	1.2 … 7.1 s
	blow-up ratio	1:2	1:1.5 … 1:3
	draw-down ratio	1:7.85	1:1.9 … 1:19.6

The processing conditions were grouped in "extruder", "die head" and "tube forming area" (along the processing line) for better structuring. It was tried to vary only one processing condition in each case while keeping the rest constant. The limits of the variations of each processing condition can be also seen in table 1.
A new developed parameter of the blowing process is the "time of solidification", which characterizes the cooling behavior of the molten film. The time of solidification is that time, which needs the molten film to solidify after exit of the die gap. In contrast to the FLH, the time of solidification considers the silhouette of the film bubble, in particular noticeable in case of different BURs. For this reason, only the time of solidification t_S is used to quantify the cooling behavior of the peel films in the present study. It was calculated following equation 1, where s_S is the length of the film silhouette from the die gap to the frost line, and v_B is the velocity of the film bubble in the same range.

$$t_S = \frac{s_S}{v_B} \qquad (1)$$

The length of the film silhouette from the die gap to the frost line s_S was measured by optical analysis of the film bubble. The velocity of the film bubble up to the frost line is not constant. The exit speed of the melt out of the die gap, and also the TUS (beeing constant from FLH) is known. The velocity of the film bubble between die gap and FLH was approximated by a linear law, since a measure of this value by other scientists evidences a nearly linear dependence [6].
After production by blowing process, the films were sealed at a temperature of 140 °C (seal area: length = 15mm, width = 5.5 mm), for a period of time of 2 s, and, subsequently, the sealed films were cooled to ambient temperature.

Instrumentation. The T-peel test according to ASTM D 1876 [22] was applied to investigate the peel behavior of the peel films produced with variable processing conditions. A Zwicki tensile-testing machine (Zwick, Germany) was used for this test. The initial distance between the clamps was 50 mm, and the standard peel rate was 100 mm/min. The recorded data, force as a function of elongation, were used to determine the peel force, which is defined as average force between 20% and 80% of the elongation at break (the plateau-like part of the curve progression).
The morphology of selected peel films was analyzed by transmission electron microscopy (TEM), to clarify the structural reason behind occuring differences of the peel behavior. The samples were oriented such that the images show the structure of the MD-ND cross-section, with MD being the machine direction, i.e., the direction of extrusion, and ND being the direction of the normal of the surface of the blown film. MD is indicated by the long axis of the bright iPB-1 particles.

Results and discussion

The processing conditions were varied in logical steps along the processing line. At first, the processing conditions of the extruder were varied. The films were produced using mix and shear elements, and in a second step, the production occurs without mix and shear elements. It can be stated that the absence of mix and shear elements (usually Maddock mix and shear elements) leads to a non peelable film. Thus, the homogeneous dispersion of the iPB-1 within the PE-LD matrix is necessary to achieve a peelable film. The next processing conditions, which were varied, were the screw speed and the temperature profile of the extruder zones (resulting in a specific measured temperature of the melt within the extruder). The screw speed has no significant influence on the peel force, because the degree of dispersion is independent from the screw speed. If the screw speed gets increased, the shear deformation also increases, however, the time of residence of the melt within the extruder decreases. So, these are contrary processes using moderate limitations of the screw speed. Fig. 1a is a plot of the peel force in dependence on the temperature of the melt within the extruder for standard screw speed. An increase of the peel force by about 13% with increasing temperature of the melt within the extruder can be observed. It can be assumed, that due to the higher degree of mobility in the molten state for the temperature of the melt of 210 °C the physical conjunctions between PE-LD and iPB-1, which occur in spite of their chemical incompatibility, become more stronger than for the temperature of the melt of 165 °C. In fact, the PE-LD lamellae grow deeper into the iPB-1 particles for 210 °C (visualized in Fig. 1b by the white arrows) than for 165 °C (Fig. 1c). Therefore, the physical conjunction between PE-LD and iPB-1 is stronger and, consequently, the peel force is higher for larger temperatures of the melt within the extruder.

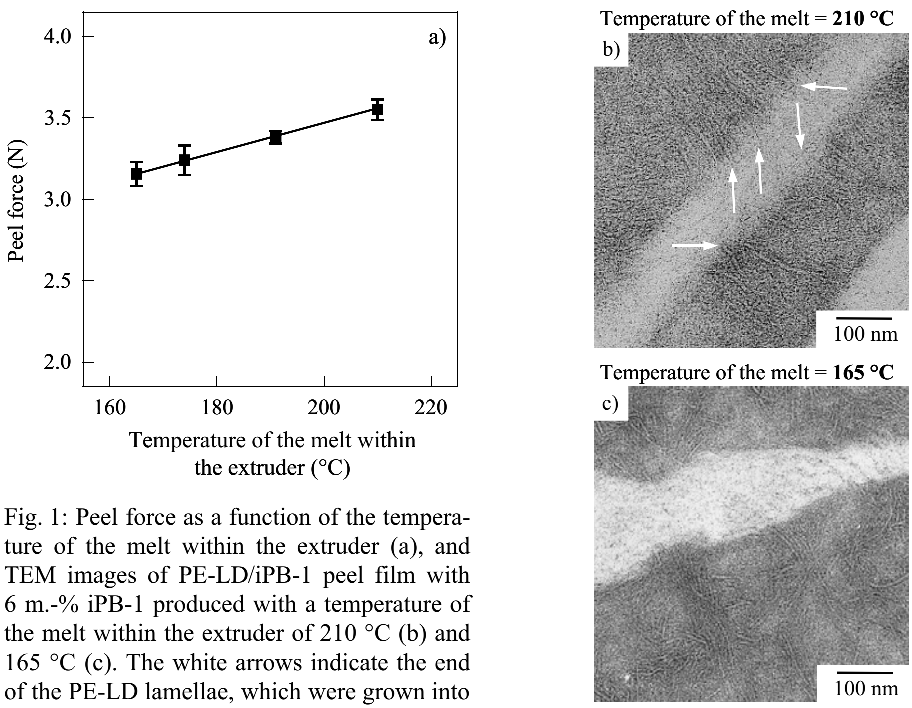
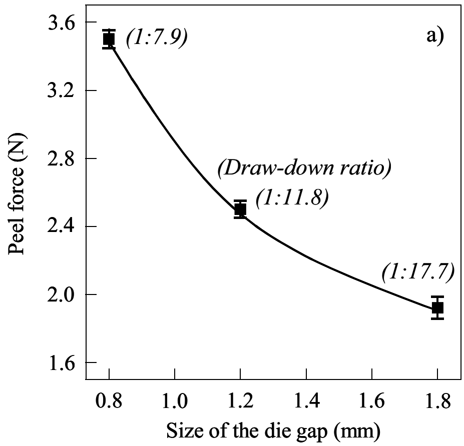


Fig. 1: Peel force as a function of the temperature of the melt within the extruder (a), and TEM images of PE-LD/iPB-1 peel film with 6 m.-% iPB-1 produced with a temperature of the melt within the extruder of 210 °C (b) and 165 °C (c). The white arrows indicate the end of the PE-LD lamellae, which were grown into the iPB-1 particle.

In a further step, the influence of the processing conditions of the die head on the peel properties were analyzed. For these purposes the temperature profile of the die head zones (resulting in a specific measured temperature of the melt within the die head) and the size of the die gap were varied. Note, that the temperature of the melt within the die head has an identical influence on the peel properties like the temperature of the melt within the extruder. Furthermore, the dependence of the peel force on the die gap size is visualized in Fig. 2a. The peel force decreases distinctly by about 40% with increasing size of the die gap. Now, it is intend to clarify the morphological reason behind this peel force decrease. The TEM images of the peel films produced with a die gap of 0.8 mm, and a die gap of 1.2 mm are shown in Fig. 2b and Fig. 2c, respectively. The iPB-1 particles are smaller and also more numerously for a die gap of 1.2 mm. In other words, the particles are well dispersed in the PE-LD matrix, using a die gap of 1.2 mm in comparison to the usage of a die gap



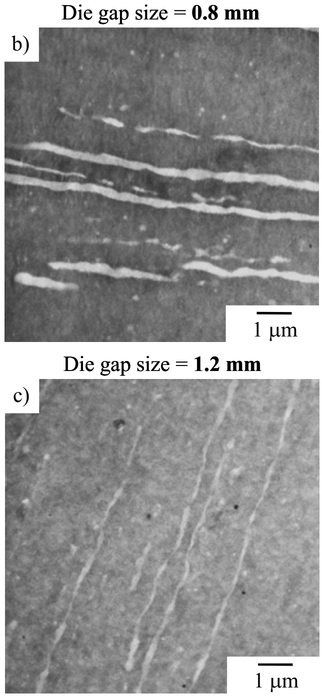
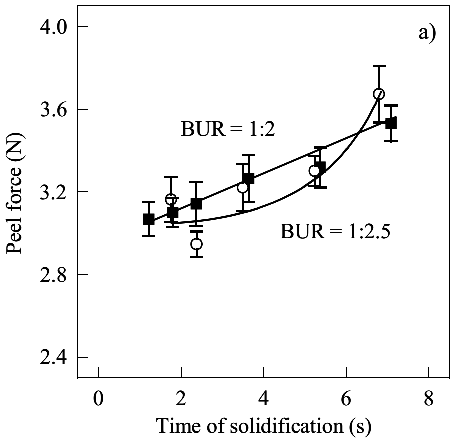
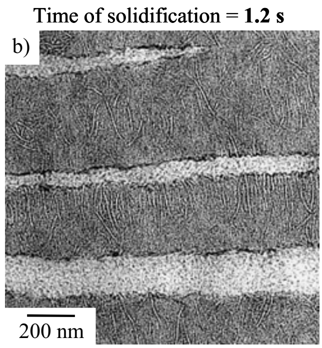


Fig. 2: Peel force in dependence on the size of the die gap and, consequently, variing DDRs (a), and TEM images of PE-LD/iPB-1 peel film with 6 m.-% iPB-1 produced with a die gap size of 0.8 mm (b) and 1.2 mm (c).

of 0.8 mm, what leads in an amazing change of the peel force. The smaller dimension and also better dispersion of the iPB-1 particles for a larger die gap can be due to the increasing DDR in case of increasing size of the die gap using a constant film thickness of 50 µm (cf. Fig. 2a). The final evidence could be achieved by analyzing the influence of the DDR on the peel properties as follows in the final part of this study.

The last step was the investigation of the processing conditions of the tube forming area in dependence on the peel properties. The tube forming area was directly influenced by the time of solidification, the BUR and the DDR. In practice, the cooling of the film bubble is one of the most important processing conditions. Therefore, an investigation of the cooling behavior in detail proves as necessary, using the new developed parameter *time of solidification*. Fig. 3a visualizes the dependence of the peel force on the time of solidification for two selected BURs of 1:2 and 1:2.5. The peel force increases by about 15–17% with increasing time of solidification for both BURs. An observation of the silhouette of the film bubble for a small and a large time of solidification reveals two completely different appearences. Thus, only the time of solidification, which includes the silhouette of the film bubble, leads to correct results in contrast to the FLH. The morphology of two peel films with different cooling behavior is shown in Fig. 3b,c for a BUR of 1:2. The top image represents a film with 1.2 s time of solidification, and the bottom image represents a film with 7.1 s time of solidification. Generally, it can be stated that the PE-LD lamellae grows slightly into the iPB-1 particles, so that physical interactions between PE-LD and iPB-1 can be occur within a small interfacial zone. Such a crystal growth is possible regarding the distinctly lower crystallization temperature of iPB-1 in comparison to PE-LD. So, the PE-LD lamellae can grow into the already molten/non-crystallized iPB-1 particles. The TEM images reveal a strong difference of the alignment of the PE-LD lamellae. A high degree of structuring and a





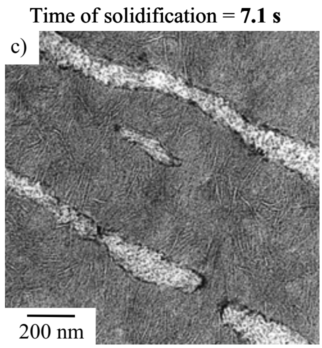
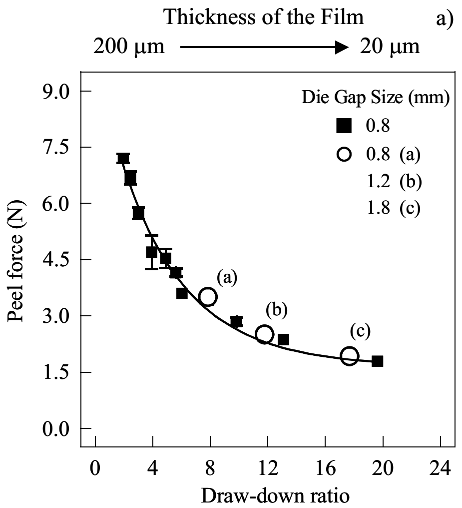


Fig. 3: Peel force as a function of the time of solidification for two different BURs of 1:2 and 1:2.5 (a), and TEM images of PE-LD/ iPB-1 film with 6 m.-% iPB-1 produced with a time of solidification of 1.2 s (b) and 7.1 s (c).

comb-like alignment of the PE-LD lamellae on the iPB-1 particles can be observed for a time of solidification of 1.2 s (Fig. 3b). In contrast to this, an isotropic alignment of the PE-LD lamellae with no preferred orientation is shown for a time of solidification of 7.1 s (Fig. 3c). Apparently, the high degree of structuring of the lamellea acts as pre-determined breaking-point on mechanical loading and supports an easy *crack growth* along the interface between PE-LD and iPB-1. In contrast to this, the random growth of the PE-LD lamellae into the iPB-1 particles acts as crack-delayed. Furthermore, it can be stated that the blow-up ratio has no significant influence on the peel force as also seen in Fig. 3a. Among the blow-up ratio, the draw-down ratio has a large part in the mechanical properties of the film. Fig. 4a shows the peel force as a function of the DDR for constant size of the die gap. Because of the constant die gap size, the thickness of the film decreases with increasing DDR. The peel force decreases with increasing DDR by about 80% by an exponential law. Fig. 4a also visualizes the three peel force values (cf. Fig. 2a) of the variation of the size of the die gap with consequently varying DDR for constant thicknesses of 50 μm (open circles). The pairs of variates of the die gap variation are in excellent agreement with the dependence of the DDR on the peel force by changing the film thickness. So, it can be stated that the DDR influences directly the peel force, independent from the way of changing the DDR, (1) due to different film thicknesses, or (2) due to different sizes of the die gap. Fig. 4b,c show the morphology of two selected films of different DDRs of 1:2.45 and 1:7.8, and also different film thicknesses of 160 μm and 50 μm, respectively. The strongly different dimensions of the iPB-1 particles are visible. Thus, a better dispersion of the iPB-1 particles due to drawing in MD (as pointed out in the part of varying processing conditions of the die head) leads to lower peel force. A further explanation, which also takes part on the peel force increase with decreasing DDR, is the simultaneously increase of the radius of curvature of the peel film. The higher the radius of



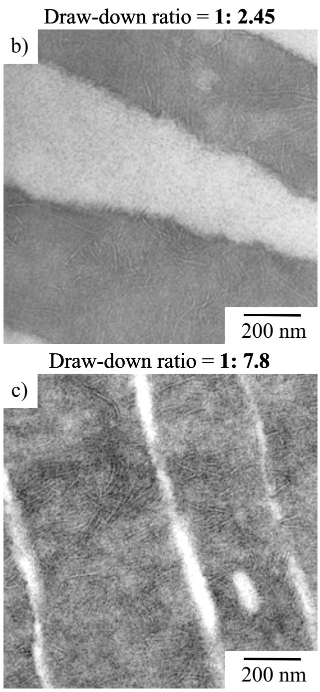


Fig. 4: Peel force as a function of the draw-down ratio (a). Additionally three pairs of variates with different die gap sizes but equal film thickness are visualized. TEM images of PE-LD/iPB-1 film with 6 m.-% iPB-1 produced with DDR 1:2.45 (b) and 1:7.8 (c).

curvature, the higher the total strength within the peel arm near the peel front. Such a higher strength can lead to a local change from interlaminar to translaminar crack propagation, which is accompanied by an increase of the energy-input [23].

Conclusions

The experimental investigation of the influence of processing conditions on the peel properties, in particular on the peel force of sealed PE-LD/iPB-1 films with 6 m.-% iPB-1 was reported in this study. The variations take place in logical steps along the processing line.
The usage of mix/shear elements is necessary to achieve a peelable film. The peel properties are independent of screw speed. The extruder temperatures have a small influence on the peel force. The die head temperature influences the peel force in a similar manner like the extruder temperature. The higher mobility of the two involved blend components for higher temperatures of the melt within the die head leads to a stronger adhesion within a small interfacial zone between PE-LD and iPB-1. Furthermore, the peel force depends strongly on the size of the die gap. In case of constant film thickness, a large die gap results in a large DDR, which leads to a fine dispersion of iPB-1.
The cooling behavior is of great practical importance. Thus, the new developed parameter *time of solidification*, which considers the film silhouette in contrast to the FLH, was used to quantify the cooling behavior of the peel films. The peel force decreases with decreasing time of solidification, due to a high oriented, comb-like alignment of the PE-LD lamellae on to the iPB-1 particles, which supports the crack growth along the PE-LD/iPB-1 interface. Among the cooling behavior, the BUR influences slightly and the DDR influences strongly the peel force. It can be stated that the DDR influences directly the peel force, independent from the way of changing the DDR, due to different film thicknesses, or due to different sizes of the die gap. The established correlations enable a defined adjustment of the peel force for a constant amount of iPB-1 of the present peel system.

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