

Evaluation and Simulation of the Peel Behavior of Polyethylene/Polybutene-1 Peel Systems

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ABSTRACT: The peel characteristics of sealed low-density polyethylene/isotactic polybutene-1 (PE-LD/iPB-1) films, with different contents of iPB-1 up to 20 m.-% (mass percentage), were evaluated and simulated in dependence on the iPB-1 content, and in dependence on the peel rate. Sealing involves close contact and localized melting of two films for a few seconds. The required force, to separate the local adhered films, is the peel force, which is influenced, among others, by the content of iPB-1. The peel force decreases exponentially with increasing iPB-1 content. Transmission electron microscopy studies reveal a favorable dispersion of the iPB-1 particles within the seal area, for iPB-1 concentrations ≥ 6 m.-%. Here, the iPB-1 particles form continuous belt-like structures, which lead to a stable and reproducible peel process. The investigation of the peel rate-dependency on the peel characteristics is of im-

portant interest for practical applications. The peel force increases with increasing peel rate by an exponential law. A numerical simulation of the present material system proves to be useful to comprehend the peel process, and to understand the peel behavior in further detail. Peel tests of different peel samples were simulated, using a two-dimensional finite element model, including cohesive zone elements. The established finite element model of the peel process was used to simulate the influence of the modulus of elasticity on the peel behavior. The peel force is independent of the modulus of elasticity, however, the peel initiation value increases with increasing modulus of elasticity. © 2008 Wiley Periodicals, Inc. *J Appl Polym Sci* 111: 363–370, 2009

Key words: polyethylene/polybutene-1 films; adhesion; peel test; interfaces; mechanical properties; modeling

INTRODUCTION

Peel films as well as peel systems are mainly used for packaging, for example in food and household packages. Peel systems are integrated in protective packagings for OI cutleries, catheters, and prostheses in the medical sector, since a fast and sterile handling is possible, using these medical devices. The peel properties, which influence the stability and reproducibility of the peel system, depend strongly, among others, on the processing conditions.¹ In practice, a sealing area between two peel films is produced by application of heat and/or pressure, using a seal device with two so-called seal plungers.

Peel systems can be realized by (1) adapting the geometry of the seal plungers in such a way, that a

discontinuous seal area like a dashed line is produced, or (2) modifying the polymer film matrix with another, mostly incompatible, component, which enables the peel process. The first case is well-known in nonaroma barrier packages. Such peel systems are not subject of scientific literature, however, only of commercial interest. In the second case, the peel system includes two or more polymer components. The present study focused on a two-component peel system. The peel system low-density polyethylene (PE-LD) combined with a small content of isotactic polybutene-1 (iPB-1), acting as peel component, is described in the literature.^{1,2} PE-LD and iPB-1 are immiscible at microscopic level. Because of this immiscibility and the dispersion of iPB-1 in the PE-LD matrix, zones of different adhesive strength are formed. The combination of PE-LD with PE-LD leads to a much higher adhesive strength, as the combination of PE-LD with iPB-1, during sealing. In case of combining iPB-1 with iPB-1, the iPB-1 particles become more aggregated. The interface between iPB-1 particles and PE-LD matrix acts as predetermined breaking point on mechanical loading. It can be stated that immiscibility and, consequently, phase

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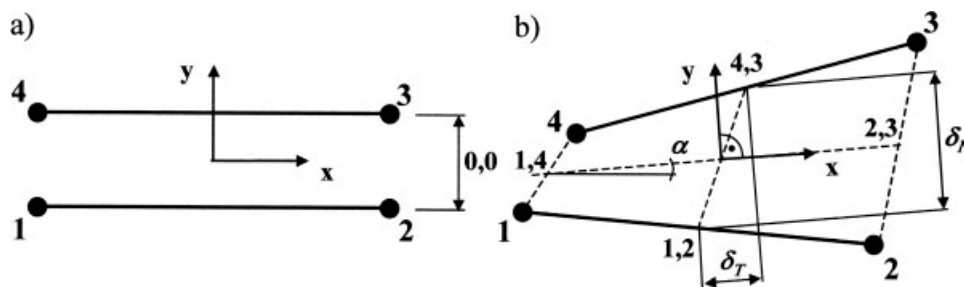


Figure 1 Undeformed (left) and deformed (right) cohesive zone element, respective to the local x - y coordinate system.

separation between PE-LD and iPB-1 is necessary to achieve acceptable peel properties. The required peel force to open the peel film package depends, among others, on the content of iPB-1, as well as on the peel rate, which is controlled by the customer.³

A numerical simulation proves to be useful to comprehend the peel process, and to understand the peel behavior in further detail. Such a finite element simulation and, consequently, finite element model of the peel process reveals an approach to nonexperimental parameter variations, which may help to limit the costs of experimental expenditure. The finite element model has to be created in such a way that a separation of the two sealed films is possible. The theoretical background for a numerical description of a crack-tip zone of a failure process, which is essential for simulated material separation, is given by the work of Dugdale and Barenblatt.^{4,5} Separation of two connected layers can be simulated, using a cohesive surface formulation, which, in turn, is part of the finite element method (FEM).^{6,7} Figure 1(a) shows a schematic cohesive element, which is described by congruent pairs of nodes (1-4 and 2-3). The undeformed initial state is of zero thickness. To describe the complete separation of two connected cohesive layers, a local x - y coordinate system is defined in the cohesive element, with respect to the virtual mid area. Now, it is possible to split the relative displacement δ in a normal δ_N and in a tangential δ_T part [Fig. 1(b)], using the x - y coordinate system, which depends on the current displacement state. In case of symmetric separation of two corresponding layers, the tangential part of the relative displacement becomes zero. Thus, the relative displacement in normal direction δ_N comply with the fracture mechanics crack-tip opening displacement, which is used to describe the toughness of a material in a specific manner.⁸ The simulation of the separation/peel process of two connected films or layers is possible, using a specific relation between cohesive strength and relative displacement (traction-separation law, TSL), which is defined by maximum strength and fracture energy of the cohesive zone, and some material parameters of the peel film.

In the present study, it is expected that the iPB-1 content has a strong influence on the peel properties

of the system PE-LD/iPB-1. Furthermore, it is found that the test rate influenced the mechanical properties of polymer materials.^{9,10} With respect to this fact, it is expected that the peel force depends on the peel rate in a similar way. Summarizing the scope of the present research, it is intended (1) to evaluate the effect of iPB-1, and the influence of the peel rate on the peel properties, especially on the peel force, and (2) to simulate the peel process, using cohesive zone elements and integrate the peel rate-dependency.

EXPERIMENTAL

Materials

The PE-LD and iPB-1 of the present study were commercial polymers, provided by Basell Polyolefins. The PE-LD used is Lupolen 2420FTM, which was designed for film production including blow-molding. The density is 0.923 g cm^{-3} , and the melt-flow index, determined at 503 K with a load of 2.16 kg, is $0.73 \text{ g (10 min)}^{-1}$. The iPB-1 of the present study was PB 8640MTM. It was a statistical copolymer with a low amount of ethylene, and for primary use as minority blend component for blown-film extrusion. The melt-flow index, also determined at 503 K with a load of 2.16 kg, is $1 \text{ g (10 min)}^{-1}$. Both polymers were blended in a single-screw extruder (Collin, Germany), using mix and shear elements and processed to films by blowing process at Orbita-Film GmbH, Germany. The iPB-1 content varied between 3 and 20 m.-%. The maximum concentration of iPB-1 in the PE-LD/iPB-1 blends was 20 m.-%, which can be considered as an upper limit for the use as peel system. The films were sealed at a temperature of 413 K, for a period of time of 2 s, and, subsequently, the sealed films were cooled to ambient temperature. The films have a thickness of 50 microns.

Instrumentation

Microscopy

The phase structure of selected blends was analyzed by transmission electron microscopy (TEM).

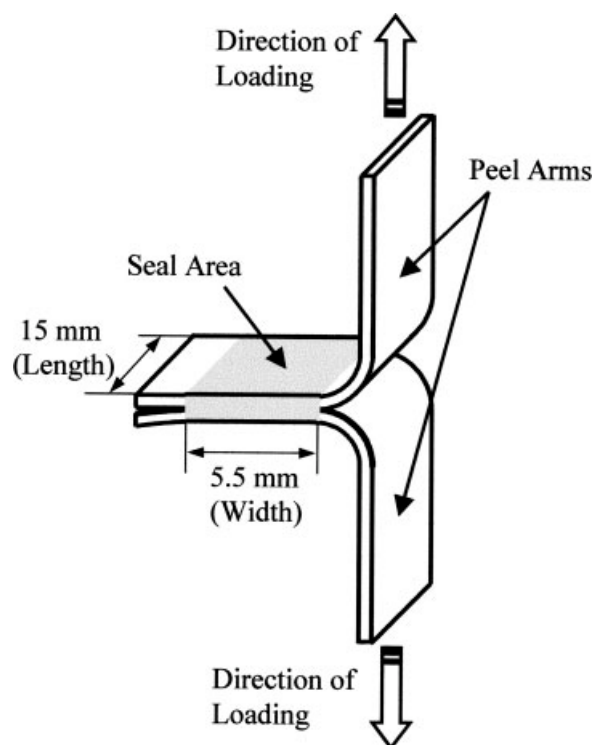


Figure 2 Scheme of the T-peel test. The gray area represents the seal area, which was sealed at 413 K, for a period of 2 s and subsequently cooled in air.

Therefore, a LEO 912 microscope (ZEISS, Germany) was used, which operated at 120 kV. Thin sections of thickness of about 80 nm were prepared using an Ultracut E (Reichard, Germany) microtome and, subsequently, stained with RuO_4 . 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.

T-peel test

The T-peel test was applied for investigation of the peel properties according to ASTM D 1876.¹¹ A scheme of the T-peel test including sample geometry, and direction of loading respective to sample orientation, is shown in Figure 2. A Zwick 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^{-1} . 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, relating to 15 mm length of the seal area. The peel rate varied between 1 and 500 mm min^{-1} for investigation of the peel rate-dependency.

Simulation

A two-dimensional model, including 6500 elements and about 14,500 translational degrees of freedom, was used for finite element simulations, which were performed at Institute for Structural Analysis in Dresden, with an in-house finite element program. The finite element model has different zones of element representation to use the elements in an efficient way. This fact leads to a rough element representation within the peel arms (element height/length = 1/5), and a fine element representation within the seal area (element height/length = 1/1), relating to 15 mm length of the seal area. A profound description of the accompanying parameter identification procedure is reported in a separate study.¹²

Analysis of the Data

A fit of the data was used in case of exponential dependency of two physical parameters following a general exponential function (eq. 1), with e being the Euler's constant, A being the difference between the initial and the end y -value of the curve, t being the reciprocal decay constant (of the parameter of the y -axis), and x_0 , y_0 being a shift of the curve in x - and in y -direction, respectively.

$$f(x) = y_0 \pm Ae^{-\frac{x-x_0}{t}} \quad (1)$$

In the present study, the reciprocal decay constant is used as a characteristic parameter to describe the appearance of the curve.

RESULTS AND DISCUSSION

The morphology of the seal area of the specific peel system PE-LD/iPB-1 was analyzed by TEM. Figure 3(a–d) shows TEM images of the seal area, i.e., a defined area between two films, which were sealed at 413 K and subsequently cooled in air, of a PE-LD/iPB-1 blend with iPB-1 contents of 3, 6, 10, and 15 m.-%. The images show the structure of the MD-ND cross section, with the bright areas representing the iPB-1 phase. MD is indicated by the orientation of iPB-1 domains. The images were taken to get an overview about the phase structure of the seal area of the PE-LD/iPB-1 blends, and to confirm immiscibility of the blend components at microscopic level, for these specific preparations. Note that immiscibility and consequently phase separation was required to get acceptable peel properties. The TEM images clearly show separation of PE-LD and iPB-1. The iPB-1 domains show a continuous belt-like geometry for iPB-1 contents ≥ 6 m.-% [Fig. 3(b–d)]. The smallest dimension is oriented parallel to ND, and, even more, in longitudinal direction of the blown-film. In

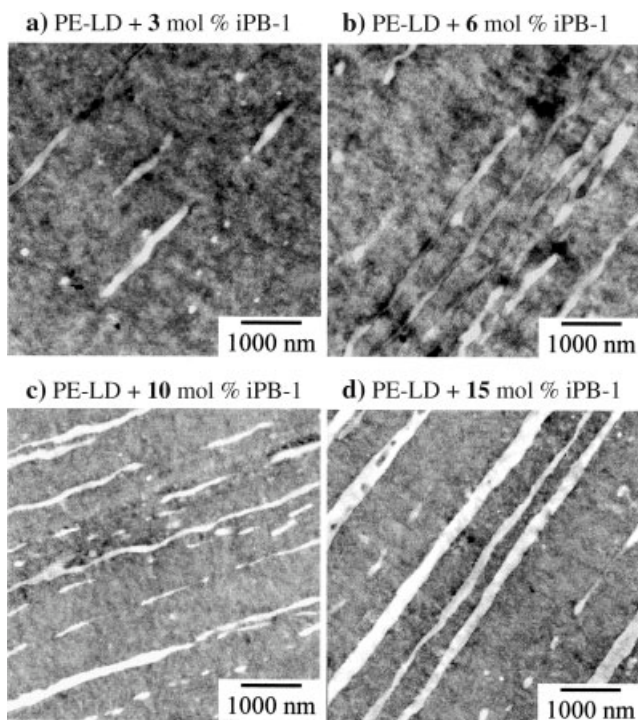


Figure 3 TEM images of PE-LD/iPB-1 blends with different iPB-1 content. The images show the phase structure of the MD-ND cross section of the seal area.

contrast to this, a discontinuous belt-like structure was observed for PE-LD with 3 m.-% iPB-1 [Fig. 3(a)]. Concerning the peel characteristics, the continuous belt-like geometry seems to be more stable for iPB-1 contents ≥ 6 m.-%, as the discontinuous belt-like geometry of the sample with 3 m.-% iPB-1. This fact can yield an inhomogeneous look of the already peeled seal area, combined with so-called stringing effects, because of the strong deformed PE-LD areas between the iPB-1 particles in the seal area, of a PE-LD/iPB-1 blend with 3 m.-% iPB-1. In addition, it could be observed that the small dimension of the belt-like iPB-1-structures increases with increasing iPB-1 content. In other words, the separated iPB-1 domains aggregate themselves with enhanced content of iPB-1. So, it is expected, that the peel force depends nonlinearly on the content of iPB-1, considering the change of the dispersion of the iPB-1 domains, from discontinuous belt-like to continuous belt-like structure to much larger belt-like structures.

The influence of the iPB-1 content on the peel force was now investigated to verify the assumption above. Therefore, force-elongation diagrams of different peel film samples were recorded. Figure 4 shows exemplary a typical peel curve, force as a function of elongation, of the blend PE-LD/iPB-1 with 6 m.-% iPB-1. The peel curve exhibits three characteristic zones. Raw-data show two local maxima, which were addressed to the geometry of the peel film sample. The partially molten polymer

moves to the border of the sample due to the inserted pressure of the two seal plungers of the seal device, during sealing at 413 K. Thus, the seal area becomes a concave cross section. This fact leads to the increased force in the border region of the seal area. The plateau-like curve progression between the two local maxima (between 20 and 80% of elongation at break) was used to calculate the peel force, i.e., the average force in this curve-range. The peel force is about 3.3 N, of the PE-LD/iPB-1 blend with 6 m.-% iPB-1, which is shown in the actual force-elongation diagram. The peel curve of Figure 4 is an example of a larger set of measurements for quantification of the dependency of the peel force on iPB-1 content. A plot of the peel force (filled squares) as a function of iPB-1 content is shown in Figure 5 to evaluate this dependency. The solid line represents a fit of the data, using a general exponential function, following eq. (1). The peel force, shown in Figure 5, decreases exponentially with a reciprocal decay constant of about 4.3 m.-%. This relation is of important interest, since different applications of peel films, like packages for food or medical devices, need an individual and defined peel force. If the peel force of a specific application, for example of a food package, is too large, then it is possible, that the protective film breaks without control, and not in a defined manner along the seal area. In the other case, if the peel force is too low, then it may happen that the seal area opens spontaneous, and the packaged substance is not safe. Now, it was possible to adjust the peel force exactly by varying the iPB-1 content, using the peel force-iPB-1 content relation. Additionally to the general dependency of the peel force on

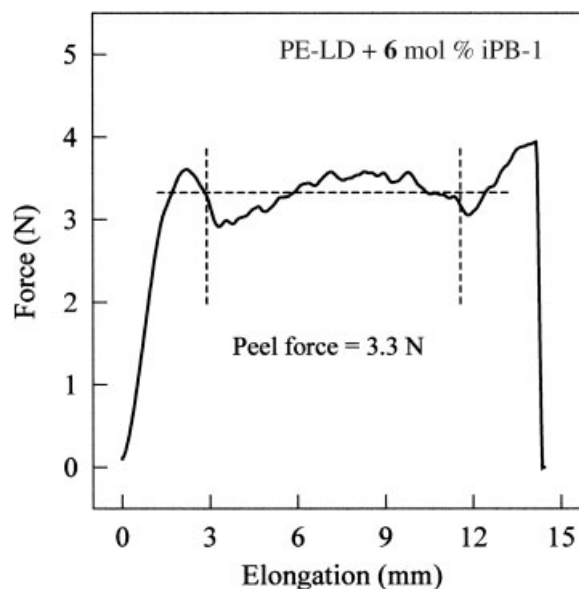


Figure 4 Force-elongation diagram of experimental data (peel curve) of the blend PE-LD/iPB-1 with 6 m.-% iPB-1.

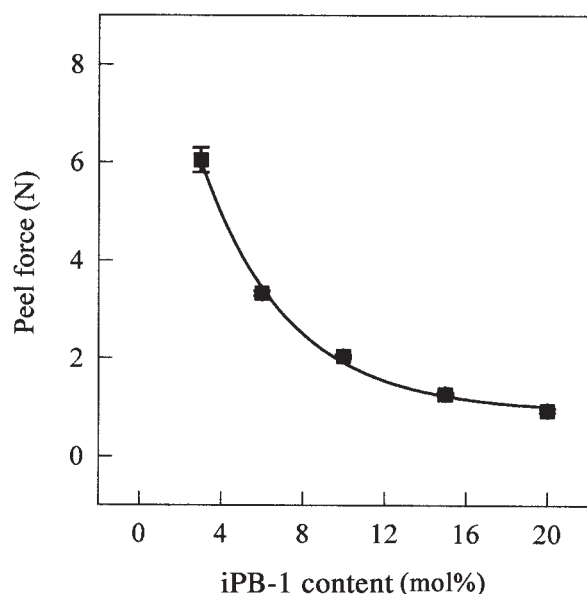


Figure 5 Peel force as a function of iPB-1 content, obtained by experimental data as the average force between 20 and 80% of elongation at break in the T-peel test. The solid line was achieved by fitting the experimental data using an exponential function.

the iPB-1 content, it could be pointed out that the peel system is only stable and reproducible for iPB-1 contents ≥ 6 m.-% as revealed by TEM measurements. Regarding the costs of the peel system PE-LD/iPB-1, it can be stated that the polymer iPB-1 is more expensive than the polymer PE-LD. So, an iPB-1 content of 6 m.-% proves as optimal, concerning costs and stability of the peel system. In other words, if it is necessary to produce peel films with different peel forces, it should be realized by varying the processing parameters, which also influence the peel force.¹³

One of the intentions of the present work is the development of the numerical simulation of the peel test by FEM, using cohesive zone elements. The finite element calculation needs some experimental input, in particular, the establishment of a TSL to provide a basis for the use of the cohesive zone elements. Therefore, it was necessary to determine the relation between cohesive strength and relative displacement (traction–separation relation) of the peel process of the sealed peel films with different content of iPB-1 for the numerical calculations within the finite element program. First step was the calculation of the separation energy Γ of the different peel processes, which corresponds to the area under the traction–separation diagram following eq. (2), with W being the total peel energy, which corresponds to the area under the force–elongation diagram, and A being the surface area of the already peeled seal area.

$$\Gamma = \frac{W}{2A} \quad (2)$$

Second step was the calculation of the maximum traction (maximum cohesive strength) T_{\max} of the different peel processes following eq. (3), with F_{peel} being the peel force, and B being the cross section of the corresponding film.

$$T_{\max} = \frac{F_{\text{peel}}}{B} \quad (3)$$

The separation energy Γ and the maximum traction T_{\max} were used to determine the traction–separation curves by fitting the data with a third-degree polynomial.^{6,14} Figure 6 shows the traction–separation curves of the peel films with different content of iPB-1. The separation energy Γ , the maximum traction T_{\max} , and also the maximum separation decrease with increasing iPB-1 content. The separation energy Γ has the strongest influence on the appearance of the traction–separation curves.¹² Now, the peel process could be simulated using the calculated traction–separation relations for the material behavior in the cohesive zone for each cohesive zone element, and some specific material parameters, whose identification was described in detail in a separate study about the numerical background of the finite element model of the peel process.¹² Figure 7 shows a peel curve based on simulated data, force versus elongation. In comparison with the experimental peel curve, some differences are obvious. The peel curve is smooth, due to the constant and perfect separation of the finite elements. The

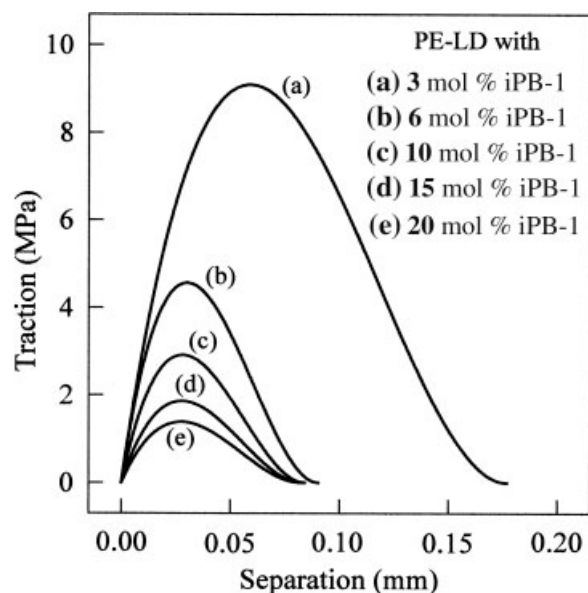


Figure 6 Traction–separation (cohesive strength–relative displacement) curves of PE-LD with different content of iPB-1.

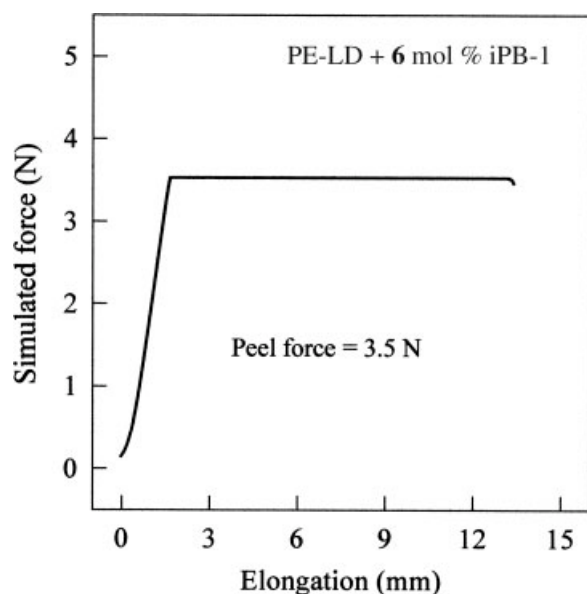


Figure 7 Force-elongation diagram of simulated data (peel curve) of the blend PE-LD/iPB-1 with 6 m.-% iPB-1.

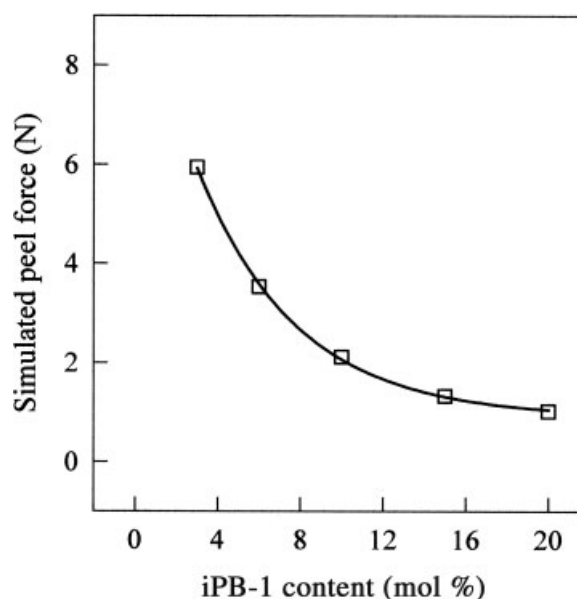


Figure 8 Simulated peel force as a function of iPB-1 content, obtained by simulation as the average force of the plateau-like peel curve progression. The solid line was achieved by fitting the experimental data using an exponential function.

thermomechanical history of the seal process (sealing at 413 K), which caused the concave cross section, was excluded, since the numerical results were based on an ideal, nondeformed seal area. Thus, the peel force could be read off easily in the force-elongation diagram. In this case, the peel force is 3.5 N for the blend PE-LD/iPB-1 with 6 m.-% iPB-1 and is in agreement with the experimental value, shown in Figure 4, which is about 3.3 N. Another important point is the different curve progression at the end of the peel process. When the finite element simulation ends, the two corresponding peel arms have unbounded displacements. Thus, the numerical calculation becomes unstable and has to be finished at this point. The peel force, obtained by simulated data, as a function of the iPB-1 content was shown in Figure 8. Raw-data was fitted by an exponential function, also following eq. (1). The reciprocal decay constant is about 4.7 m.-% in case of the simulated peel force of the blend PE-LD/iPB-1. This result is in accordance with the reciprocal decay constant of the experimental data, which was 4.3 m.-%. The minor difference between the two constants is addressed to the limited number of data points of the peel force-iPB-1 content plots.

Another intention of this study was the investigation of the dependency of the peel force on the peel rate, and their integration in the finite element model. Figure 9 shows with the filled squares the experimentally obtained peel force as a function of the peel rate, of the blend PE-LD/iPB-1 with 10 m.-% iPB-1. The fit of the data, represented by the solid line, reveals an exponential increase of the peel force with increasing peel rate, which is in agreement

with former literature concerning rate-dependency of mechanical properties.⁹ The reciprocal decay constant of the data-fit is about 33 mm min⁻¹, according to the analysis of the peel force dependence on iPB-1 content. The data show nearly constant and peel rate-independent peel forces for peel rates ≥ 100 mm

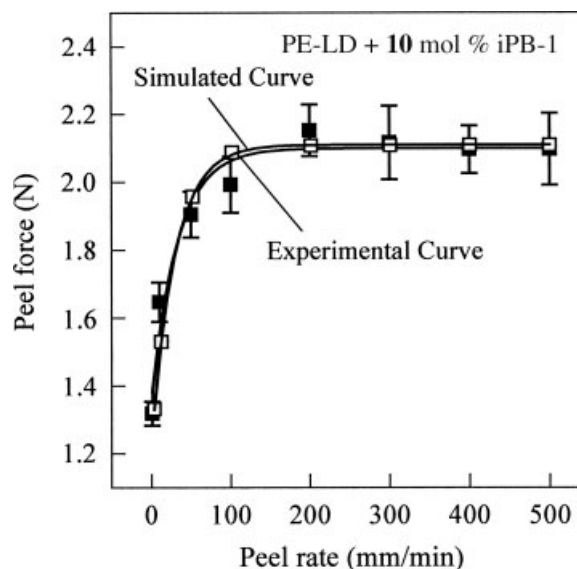


Figure 9 Experimentally obtained peel force (filled squares) and simulated peel force (open squares) as a function of the peel rate of the polymer blend PE-LD/iPB-1 with 10 m.-% iPB-1, using the T-peel test. The solid lines, which represents the fitted raw-data, follow an exponential law.

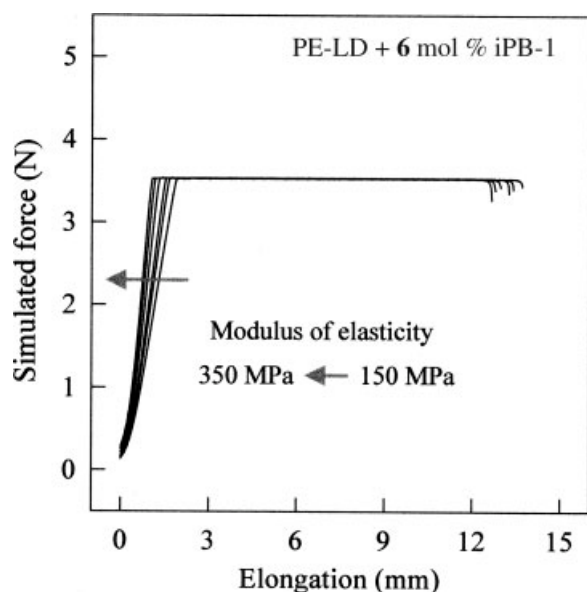


Figure 10 Set of peel curves, with different moduli of elasticity, of the blend PE-LD/iPB-1 with 6 m.-% iPB-1, based on simulated data.

min^{-1} , which are the most common in practice. This fact is of meaningful interest for practical applications, since it is impossible to specify a typical peel rate for the customers. In comparison to this, Figure 9 shows with the open squares the dependency of the peel force on the peel rate of the PE-LD/iPB-1 blend with 10 m.-% iPB-1, based on simulated data. The fit of the data is also of exponential character. A reciprocal decay constant of about 30 mm min^{-1} was calculated, which is in agreement with the experimental result. Thus, the finite element model of the peel test was extended by the peel rate-dependency.

Such a complex finite element model of the peel process can now be used to vary several parameters, and achieve simulated results, which extend the knowledge of the investigated material and help to limit the costs of experimental expenditure. The stiffness of polyolefine films is an important mechanical parameter concerning the further processing of the films, e.g., the laminating of films with some other materials. In this case, a defined modulus of elasticity is necessary and it is the result of the moduli of several film layers, which can be added to the peel film layer, and the real peel film layer.

So, the variation of the modulus of elasticity of the peel system is of practical interest, since it is mostly one of a specific number of film layers. Figure 10 shows a set of peel curves, with different modulus of elasticity, of the blend PE-LD/iPB-1 with 6 m.-% iPB-1. The right peel curve was obtained by a peel film sample with a low modulus of elasticity of 150 MPa, and forwards to the left, the modulus increases

up to 350 MPa. In spite of the variation of the modulus of elasticity in the range of 150 MPa up to 350 MPa, the peel force does not change. The plateau-like part of the peel curve progression has always the same force value. So, a defined modulus of elasticity can be realized without an unwanted change of the peel properties. However, the onset of the peel force plateau decreases exponentially with increasing modulus of elasticity (Fig. 11). Regarding the finite element representation within the seal area, it seems logic that the onset of the peel force plateau decreases with increasing modulus of elasticity, and, consequently, increasing numbers of elements, which were separated simultaneously. In other words, if the stiffness of the film increases, than the radius of curvature of the peel arm also increases. In case of a larger stiffness of the film, some more elements were loaded and separated, respectively.

CONCLUSIONS

The experimental investigation and numerical simulation of sealed PE-LD/iPB-1 films with different content of iPB-1 was reported in this study. Regarding costs and stability of the peel system PE-LD/iPB-1, it can be stated that an iPB-1 content of 6 m.-% is optimal. The mechanical properties of the peel system depend exponentially on the peel rate. However, the data show nearly constant and peel rate-independent peel forces for peel rates $\geq 100 \text{ mm min}^{-1}$, which are the most common in practice. The successful numerical simulation of the peel process gave an approach to nonexperimental parameter variations, for example the variation of the

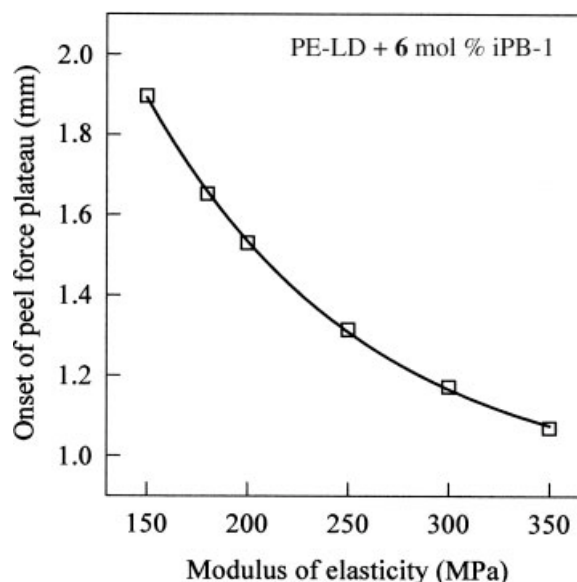


Figure 11 Onset of the peel force plateau as a function of the modulus of elasticity.

total modulus of elasticity. The numerical investigation of this dependency reveals an independency of the peel force on the modulus of elasticity. So, the peel system is multifunctionally applicable without change of the adjusted peel force. However, the onset of the peel force plateau decreases with increasing modulus of elasticity.

Further applications of the finite element model of the present peel systems are feasible in future work. Note that in case of a symmetric separation of the layers, the relative displacement in normal direction δ_N comply with the fracture mechanics crack-tip opening displacement. It is difficult to obtain the crack-tip opening displacement by experimental measurements of the films presented in this study. Thus, a favorable alternative could be the simulation of the peel process using the FEM and, consequently, get the crack-tip opening displacement as a numerical result.

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References

1. Hwo, C.C. *J Plast Film Sheeting* 1987, 3, 245.
2. Stober, P. *Kunststoffe* 2004, 6, 66.
3. Nase, M.; Androsch, R.; Langer, B.; Baumann, H. J.; Grellmann, W. *J Appl Polym Sci* 2008, 107, 3111.
4. Dugdale, D. S. *J Mech Phys Solids* 1960, 8, 100.
5. Barenblatt, G. I. *Adv Appl Mech* 1962, 7, 55.
6. Needleman, A. *J Appl Mech* 1987, 54, 525.
7. Rahul-Kumar, P.; Jagota, A.; Bennison, S.; Saigal, S.; Muralidhar, S. *Acta Mater* 1999, 47, 4161.
8. Grellmann, W.; Seidler, S., Eds.; *Deformation and Fracture Behaviour of Polymers*; Springer-Verlag: New York, 2001.
9. Schoßig, M.; Bierögel, C.; Grellmann, W.; Bardenheier, R.; Mecklenburg, T. In *Proceedings of the European Conference of Fracture (ECF 16)*, Alexandroupolis, Greece, 2006, p 1.
10. Spathis, G.; Kontou, E. *J Appl Polym Sci* 2004, 91, 3519.
11. ASTM D 1876. Standard Test Method for Peel Resistance of Adhesives (T-Peel Test); ASTM: West Conshohocken, PA, 2001.
12. Geißler, G.; Kaliske, M.; Grellmann, W.; Nase, M. *Eng Comp* 2007, 24, 586.
13. Nase, M.; Langer, B.; Baumann, H. J.; Grellmann, W. In *Proceedings of International Conference on "Deformation and Fracture Behavior of Polymers,"* Merseburg, Germany, 2007, p 283.
14. Tvergaard, V. *Mater Sci Eng A* 1990, 125, 203.