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Lamination of Polyethylene Composite Films by Ultrasonic Welding: Investigation of Peel Behavior and Identification of Optimum Welding Parameters

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ABSTRACT: The lamination of different polyethylene (PE) composite films by ultrasonic welding to fabricate peelable seals that open at defined burst pressures is investigated. The sealing time, pressure, and amplitude were varied within the range of 100–400 ms, 50–250 kPa, and 12–24 μm, respectively. T-peel tests and electron micrographs revealed four different peel regimes, depending on the parameter combination: (I) *Interlaminar peeling* at low-peel strength with uniform peeling along a weakly bonded PE lamination layer; (II) *transition tearing* at intermediate peel strength showing areas of interlaminar peeling and translaminar tearing; (III) *translaminar tearing* at high-peel strength showing tears through the entire film; and, (IV) *undefined tearing* at varying tear strength occurring when vibration effects during welding lead to insufficient contact of the films or high pressures lead to a displacement of PE. This study will allow the systematic adjustment of ultrasonic welding parameters for PE films. © 2013 Wiley Periodicals, Inc. J. Appl. Polym. Sci. **2014**, *131*, 40291.

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INTRODUCTION

Peelable seals are often used in manufacturing of easy-to-open packages. Laminates of PE composite films are extensively used in both food and pharmaceutical packaging.[1–4] The composite films of aluminum and polyethylene (PE) are typically employed in applications, where superior gas barrier properties are required.

Heat sealing is the most common method to seal plastic films. For the various heat sealing technologies described in literature,[5,6] the most influential parameters for bond strength are sealing pressure, sealing time, and temperature.[7–10] The influence of these parameters[11–13] and the influence of the polymer composition[13–16] on the peel mechanism have been well investigated. However, the heat uptake during thermal sealing can damage the heat-sensitive products, when packaged in small volumes. Ultrasonic welding may offer a preferable alternative,[17,18] where the thermal energy required for sealing is produced primarily in the interface of the layers. The sealing times for polymer films is typically in the range of 50–200 ms, which limits the thermal stress on the packaged product. In addition to pressure and time, the amplitude of a sonotrode also influences the peel strength.

Although there are relatively high numbers of scientific publications in the field of heat sealing of thin films, the prevalence of systematic studies on ultrasonic welding of thin PE films is limited. However, due to an increasing number of innovative packaging solutions in various fields of application, a deeper understanding of the relation between ultrasonic welding parameters and peel behavior is required. One example of such an innovative packaging approach is stickpackaging of *in vitro* diagnostic reagents in volumes as low as 80 μL, which is required for point-of-care diagnostic systems.[19] These stickpacks are manufactured from an aluminum–PE thin film and equipped with a peelable seal, which is created by an adjusted ultrasonic welding process. During use, the peelable seal of the stickpacks needs to open at a defined burst pressure in order to release the prestored reagents on demand.

This study investigates the influence of ultrasonic welding parameters on the peel behavior of laminated aluminum–PE films. Practical advices have been elaborated, which allow the

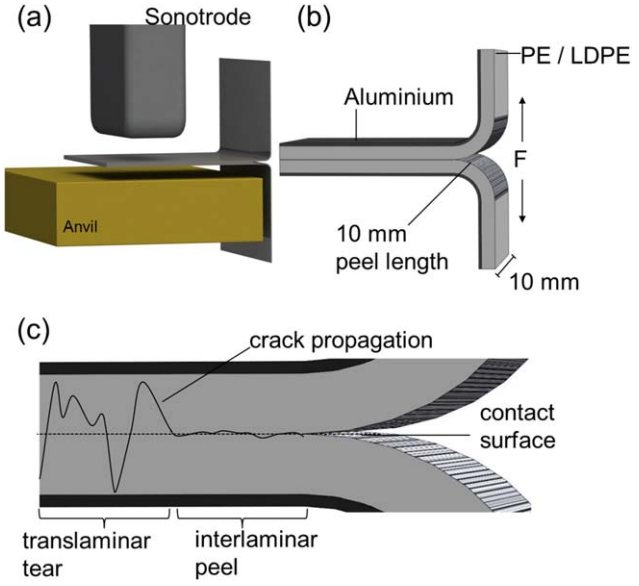

Figure 1. Peel testing: (**a**) setup of the ultrasonic welding, (**b**) scheme of the T-peel test and dimensions of the test strips, and (**c**) tear formation of the films. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

rapid selection of optimum welding parameters in order to achieve defined peel strengths.

EXPERIMENTAL

Materials

Three different commercially available composite (PET–)Aluminum–PE films were investigated: "Standard Peel" ("PET/Alu/PE 12/12/50 µm, no. 68121250JG102", referred to as *standard film,* and "Peel-Seal" PET/Alu/PE 12/9/50 µm no. 68120950JG101", referred to as *peel-seal film,* both supplied by JG Service AG, Hohenwart, Germany) with a 50 µm PE layer and the third investigated film is a commercially available extrusion-coated film ("MCE 38EE28" supplied by Constantia Teich, Weinburg, Austria) with 28 g m^{-2} PE on 30 µm aluminum, referred to as *LDPE film.* All the investigated seals were PE-sealed on PE (or LDPE on LDPE) of the same film.

Ultrasonic Welding

The peelable seals were sealed by a 35 kHz ultrasonic welder type "ECO 35" (SONOTRONIC Nagel, Karlsbad, Germany) with an unstructured sonotrode (type "35 kHz, Ti, RSMS 33/21/10, L1/2") by SONOTRONIC. The anvil was an unstructured brass block.

Peel Testing

T-peel tests were applied for investigation of the peel strength using a "Zwick Z010" testing machine (Zwick GmbH, Ulm, Germany). Test strips 10 mm wide were sealed and investigated with a peel rate of 50 mm min^{-1}. The initial distance of the clamps was 30 mm. The length of the peel seal was 10 mm, according to the dimensions of the sonotrode [Figure 1(a,b)]. The peel strength was determined by the average force between 20% and 80% of the elongation at break.[13,20] The error bars represent the standard deviations of a mean of four individual tests. The surface structure of peeled test strips is described as the *tear formation.* Two types of crack propagations are distinguished,[13,16] that is, the *interlaminar* (the delamination along the contact plane of the two test strips) and the *translaminar* (cracks also propagate through the contact plane of the films) [Figure 1(c)].

RESULTS AND DISCUSSION

Investigation of Peel Behavior

The most critical ultrasonic parameters, that is, sealing time, pressure, and amplitude of the sonotrode (at a constant frequency of 35 kHz), were investigated. Unless otherwise specified, hereafter the sealing parameters refer to a sealing time of 200 ms with a surface pressure of the sonotrode at 150 kPa and an amplitude of 17 µm. Similarly, unless otherwise specified, the type of the film refers to the *standard film.*

Two-dimensional Analysis on the Peel Strength

Sealing Time. The effect of the sealing time, in the range of 140–400 ms, on the peel strength indicates three different peel formation regimes (Figure 2): the *interlaminar peel* (I) with

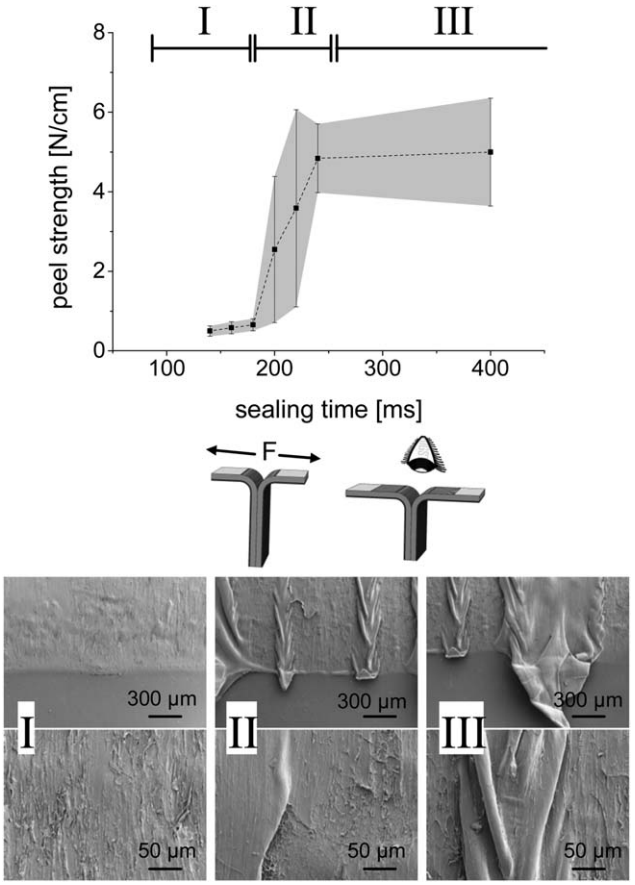

Figure 2. Peel behavior of the *standard film.* **Top**: The effect of sealing time on the peel strength. **Bottom**: Electron micrographs of various seal surfaces, that is, (I) *interlaminar peel* (sealing time 140 ms), (II) *transition tear* (sealing time 220 ms), and (III) entirely *translaminar tear* (sealing time 400 ms).

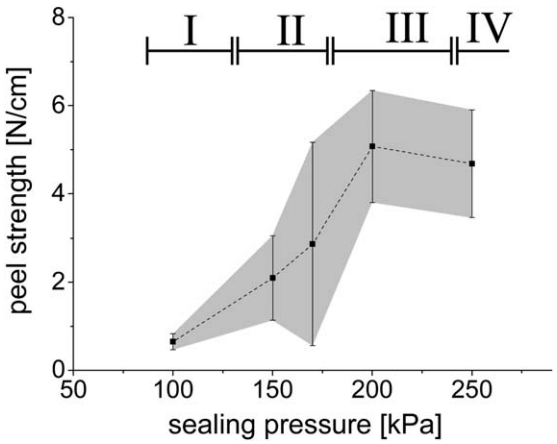

Figure 3. The effect of the sealing pressure on the peel strength. At 250 kPa the molten PE is displaced from the seal area and therefore this range is indicated as an *undefined tear (IV)* since the thickness of the PE layer has changed.

sealing times at <200 ms corresponds to an entirely interlaminar delamination—the delamination along the contact surface. These bonds show a defined binding strength (nearly independent from the sealing time) with only slight deviations. The *transition tear* (II) after a threshold of approximately 200 ms describes a dramatically increase of the peel strength—a time frame of approximately 50 ms determines whether the seal peels very easily or not. The very high deviations of the peel strength in this range illustrate the transition from the interlaminar to translaminar tear formation. Some areas peel easily, while others are strongly bonded and cannot be separated until the PE itself tears in a translaminar way. At the *translaminar tear* (III) if a certain threshold is reached (above 250 ms), the peel strength remains constant even when the sealing time increases. In this regime the entire area of the seal is strongly bonded which causes the tearing surface to be translaminar. The deviations of the peel strength decrease compared with the transition range as there are no weakly bonded areas along the entire tear surface.

The electron micrographs of the delaminated surfaces (Figure 2, below) illustrates the inter- and translaminar spread of the tear. Note that the difference between an interlaminar peel and a translaminar tear can also be clearly distinguished by the naked eye (see Figure A1). The possible effects on the molecular scale and the mechanics that lead to this tear formation are already described in detail in other publications.[3,16,21,22] One concept describes the interdiffusion and entanglement of PE, where the molecular ends on the contact surface diffuse across the contact surface and form stable bonds.

Pressure. The effect of contact pressure of the sonotrode in the range of 100–250 kPa on the peel strength indicates similar peel formation regimes (Figure 3). The visual analysis of the tear surface of the first regime (I) is consistent with the typical formation of *interlaminar peel* in Figure 2. Also, the low deviation of the peel strength at 100 kPa indicates a purely *interlaminar peel* formation. The high deviations in the peel strength between 150 and 180 kPa, and the visual analysis of the tear surface are comparable to the *transition tear* (II) in Figure 2. The plateau of the *translaminar tear* (III) at 200 kPa pressure (Figure 3) corresponds to the maximum strength of PE. Once the value of approximately 5 N cm^{-1} is reached, increased pressure does not increase the peel strength. The slight decrease of peel strength from 200 to 250 kPa is insignificant and within the standard deviation. However, the visual analysis of the peel seam indicates that at 250 kPa, the molten PE was displaced from the seal area. Since the thickness of the PE layer varies, this range is described as *undefined* (IV). Nevertheless, the tear formation corresponds to one that is entirely translaminar. On a test strip this regime can clearly be identified because some of the PE is displaced from the seal area onto the anvil (see Figure A1).

Figure 3 shows that the peel strength can be adjusted to a certain degree by changing the pressure alone. This differs from heat sealing of similar thin films, where it is assumed that the contact pressure has no influence on the seal strength as long as the melt is not displaced, since diffusion occurs independent of the pressure.[7,9] Additionally Figure 3 shows that the energy transfer in ultrasonic welding, as opposed to heat sealing, is influenced substantially by the pressure. Furthermore, the pressure is required to prevent the two films from shifting apart of each other during welding.

Amplitude. The effect of the amplitude of the sonotrode on the peel strength is investigated in the range of 12–24 µm (Figure 4). The visual analysis of the tear surface and the low deviations of the peel strength in the first regime (I) are consistent with the typical formation of the *interlaminar peel* (Figures 2 and 3). At increasing amplitude, the *transition tear* (II) from *inter-* to *translaminar tear* formation does not occur so dramatically as in the previous experiments (cf. Figures 2 and 3). However, the optical analysis of the tear formation is consistent with the previously conducted experiments as certain zones tear in a translaminar way, while others only peel

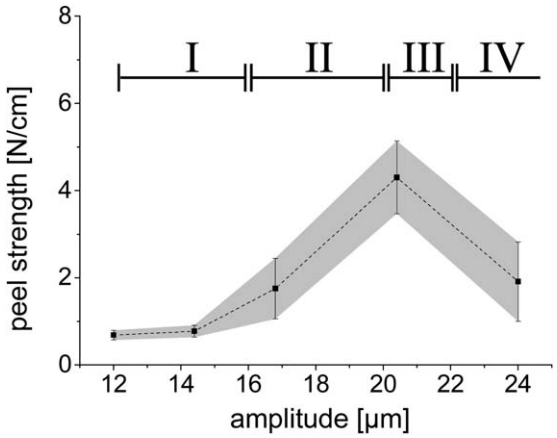

Figure 4. The effect of the amplitude of the sonotrode on the peel strength. At an amplitude of 24 µm, vibration effects prevent the sonotrode from maintaining good contact with the film, thereby leading to a significant drop in the peel strength.

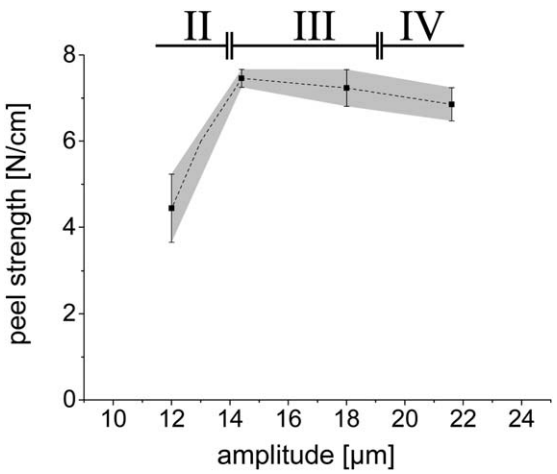

Figure 5. The effect of the amplitude of the sonotrode on the peel strength of the *LDPE film*. The decrease of the peel strength in the *undefined tear range* (IV) is not as pronounced as in the *standard film* but a shifting of the films during welding is clearly recognizable.

weakly. The maximum tear strength of PE is reached at the translaminar tear (III) regime at an amplitude of 20 µm. Unlike in the previously conducted experiments, the peel strength decreases significantly above a certain amplitude (>20 µm). Under high relative amplitudes (50 µm PE layer and 20 µm amplitude), the experimental setup of the ultrasonic welding system (especially the brass block on which the films are welded) can vibrate with the sonotrode. As a result, the sonotrode hits against the surface of the film intermittently, which prevents sufficient friction between the two films and leads to a shifting of the films during welding. Despite the higher energy input from the higher amplitude, the two PE layers do not seal well with each other. This range is described as *undefined* (IV). This unwanted induced vibration can clearly be heard during welding. Operating in this regime should be avoided due to the increased mechanical stress that can cause damage of the welder.

To further investigate this effect of insufficient contact of the sonotrode, the experiment was repeated with another, thicker type of PE film (70 µm *LDPE film*). This PE layer presents the *translaminar tear* (III)—the maximum peel strength already at an amplitude of 14 µm (Figure 5). Higher amplitudes also decrease the peel strength, albeit without such a sharp decline as observed for the *standard film* (Figure 4). This is indicated as the *undefined tear* (IV). Due to the thicker PE layer of the *LDPE film*, the induced vibrations in the experimental setup are lesser. Thus, the intermittent loss of contact of the sonotrode with the surface is significantly reduced, although a shifting of the two films during welding at 22 µm is still clearly visually recognizable. The induced vibrations can also clearly be heard during welding. To avoid damage of the welder no data points higher than 22 µm are provided.

Three-dimensional (3D) Analysis of the Welding Parameters

In this study, the amplitude and the sealing time were varied for the investigated films (Figures 6 and 7) at a constant pressure of 150 kPa. In general, a certain minimal pressure is required to ensure good fixation of the films. An excessively high pressure can lead to a displacement of the PE. However, additional tests at variable pressures show that the qualitative progression of the level curves stay the same within these pressure boundaries (see Appendix for details).

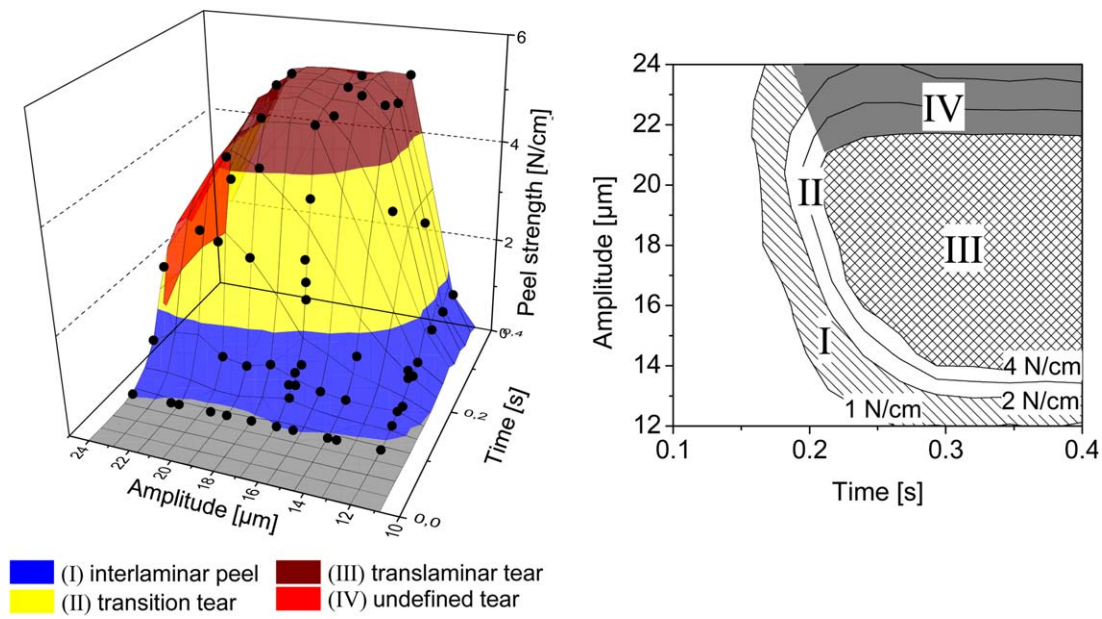

Figure 6. The effect of the amplitude and the sealing time on the peel strength of the *standard film*. **Left**: Three-dimensional analysis indicating the four different tear ranges. **Right**: Level curves from 1 to 4 N cm^{-1} peel strength. The *undefined tear* range describes a decrease of the peel strength due to vibration effects and displacement of the melted PE. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

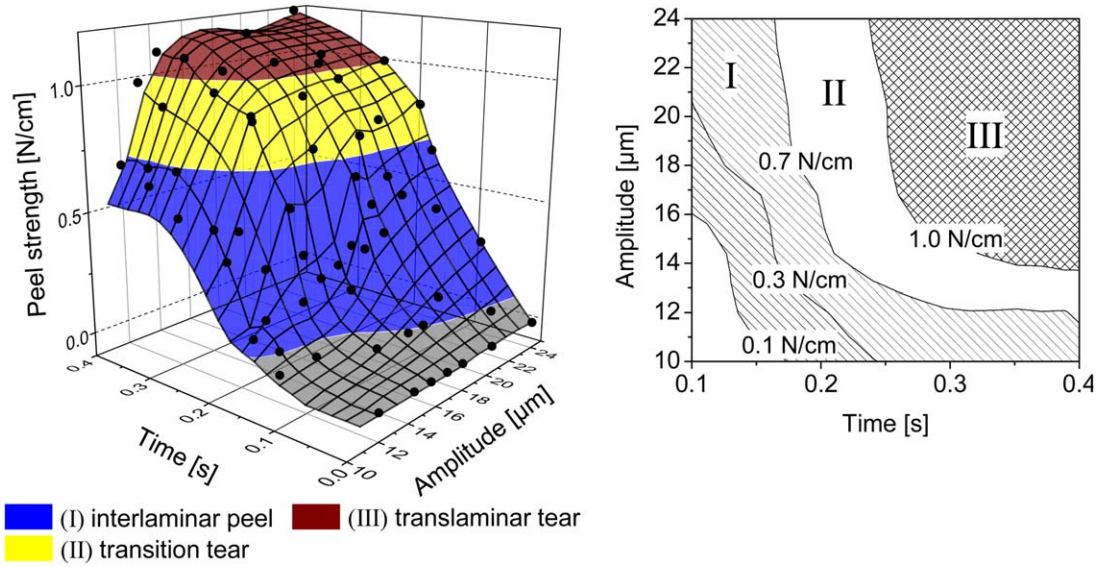

Figure 7. The effect of the amplitude and the sealing time on the peel strength of the *peel-seal film*. **Left**: Three-dimensional analysis indicating three different tear ranges. **Right**: Level curves from 0.1 to 1.0 N cm^{-1} peel strength. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Figure 6 (left) shows the dependency of peel strength on the amplitude and the sealing time of the *standard film*. For a better understanding, Figure 6 can be compared with the data points in Figures 2 and 4, which are sections of the 3D plot. The level curves of the plot are shown in Figure 6 (right). The range from 1 to 2 N cm^{-1} with only slight deviations in the peel strength corresponds to the purely *interlaminar peel* formation (I). The area from 2 to 4 N cm^{-1} peel strength corresponds to the *transition range* with high deviations in the peel strength (II) and the strongest possible bond of >4 N cm^{-1} is in the area of the *translaminar tear* formation (III). The area of amplitudes >20 µm and sealing times >0.2 s corresponds to the *undefined tear* (IV) formation, where vibration effects occur or the thickness of the layer is undefined due to displacement of the melted PE. The level curves indicate that various combinations of multiple parameters can lead to the same peel strength. Usually, when polymer films are sealed, the aim is to maximize the seal strength. However, in certain applications, a peelable seal is desired. As an example of a peelable PE film, the *peel-seal film* was investigated with similar tests as performed with the *standard film* (Figure 7). The level curve (Figure 7, right) shows that there is no undefined tear range, which is in contrast to the investigated *standard film*. Between 14 and 20 µm of amplitude, the entire range of peel strengths can be set by adjusting only the time. However, the maximum bond strength cannot be reached even at the highest amplitudes with <0.25 s of sealing time.

This study shows that the relative influence of the welding parameters on the peel behavior of different PE films is similar. Based on the tear formation, different regimes can be identified and the ultrasonic welding parameters for either strong bonds or peelable seals can be selected accordingly. Higher amplitudes cannot always be compensated by shorter sealing times and vice versa. At a certain point the displacement of melted PE and vibrational effects during welding lead to a decrease of the peel strength.

Practical Guideline for Parameter Setting

The data from Figures 6 and 7 are obtained by extensive and time-consuming T-peel tests on a tensile testing machine (~100 tests in 2 days for each film). As a more practicable alternative, a procedural scheme is derived for finding appropriate ultrasonic welding parameters by only visual inspection of peeled seal seams (Appendix). The suggested guideline (Table AI) allows analyzing the influence of welding parameters on peel strength with only 25 tests in <2 hr. It describes each step of the procedure and the expected result.

CONCLUSION

The influence of ultrasonic welding parameters on the peel behavior of PE films has been thoroughly investigated. The existence of characteristic tear formations, already known for heat sealed films, has been revealed for ultrasonically welded films. Increase of pressure, sealing time, and amplitude lead to increased peel strength. At a certain point, vibration effects occur or melted PE starts to be displaced leading to an abrupt decrease of the seal strength. The study will help to enable adjustment of peelable seals that open at defined burst pressures fabricated by ultrasonic welding. The practical guideline enables an uncomplicated, systematic and rapid choice of welding parameters and helps to circumvent time-consuming T-peel tests.

ACKNOWLEDGMENTS

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Table AI. Practical Guideline for the Rapid Identification of Appropriate Ultrasonic Welding parameters

(1) Minimum pressure	
The minimum pressure is set to ensure a good fixation of the films during the welding process so that the film surfaces do not shift relatively to each other, even at high amplitudes.	
Procedure	**Results**
Set amplitude to 80% of maximum value (19 μm).	Minimum pressure at which the two films do not shift (150 kPa for all PE films).
Set sealing time to a value that is long enough to visually observe a possible shifting of the two films (500 ms).	Test strips sealed across the entire area.
Seal test films with gradually increased pressures. If the two films shift apart of each other or are not sealed across the entire area, increase the pressure (increments of 25 kPa).	
(2) Minimum time and amplitude	
With the minimal pressure found in (1), test films are sealed at different amplitudes but with the lowest possible time. This corresponds to an entirely interlaminar peeling and gives line A in Figure A2.	
Procedure	**Results**
Chose approximately five different amplitudes ranging between maximum and minimum possible values (24–12 μm).	Test strips sealed homogeneously across the entire area.
For each of the amplitudes, the sealing time is adjusted until the minimum time to obtain a homogeneous seal is found (increments of 50 ms, sealing times ranging from 90 to 550 ms).	Interlaminar peel surface according to Figure A1.
(3) Maximum peel strength	
Test films are sealed until the maximum bond is reached. This corresponds to amplitude/time combinations, where the tear formation changes from inter- to entirely translaminar (Figure A1) and gives line B in Figure A2.	
Procedure	**Results**
Starting with the parameters found in (2), at each point, the amplitude is gradually increased (at constant sealing time) until the tear surface becomes entirely translaminar (increments of 2 μm).	Translaminar tear surface according to Figure A1.
If the highest amplitude is reached and still no maximum bond is obtained, the time is gradually increased at this point (increments of 50 ms).	This corresponds to the maximum peel strength.
Vibration	**Results**
If a certain point is reached, where unwanted vibrations in the ultrasonic welding setup appear or the films shift during welding, the upper amplitude and time limit is reached. This limit corresponds to line C in Figure A2. It depends on the available amplitude and time range of the ultrasonic welding machine whether this limit can be found.	The PE is displaced from the actual tear surface or a inhomogeneous seal occurs after the increase of amplitude or time.
(4) Parameter adjustment	
The area between line A and B in Figure A2 can now be used to adjust the peel strength by smaller increments of amplitude or/and sealing time. The area enclosed by lines B and C represents the maximum peel strength.	
Procedure for fine adjustment of peel strength	**Results**
Starting from a set of parameters on line A in Figure A2, the peel strength is increased by either increasing the sealing time (increments of 1 μm) or the amplitude (increments of 10 ms).	Variable peel strength within the interlaminar peel formation.
Procedure to obtain maximum bond strength	**Results**
If only the maximum strength is required, an operating point that is located deeply inside the area of the maximum bond is chosen. This ensures that even with deviations in the sealing time or amplitude, the bond strength stays the same.	Maximum bond strength.

The parameters in brackets correspond to the values used in this study.

APPENDIX

The procedural scheme (Table AI) describes the rapid identification of appropriate ultrasonic welding parameters by only visual inspection of peeled seal seams (Figure A1).

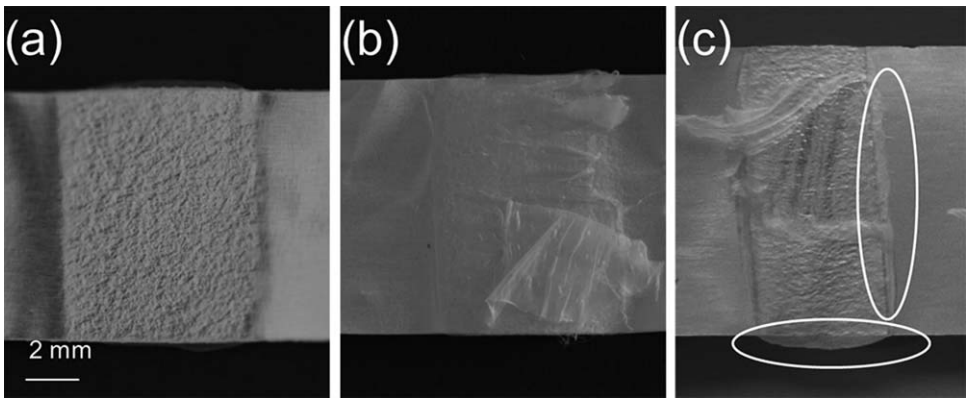

Figure A1. Visual analysis of tear formation with the naked eye. (**a**): Interlaminar peel formation; (b): translaminar tear formation (maximum peel strength). (**c**) Displaced polymer at high pressure/amplitude.

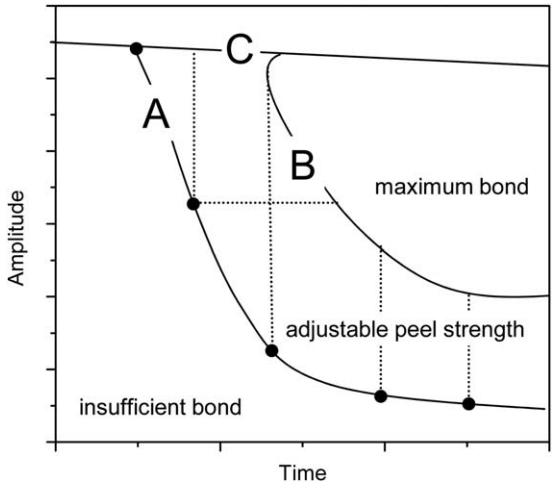

Figure A2. Simplified diagram for finding the right ultrasonic parameters according to the procedural scheme provided in Table AI. The area between line A and B corresponds to the interlaminar (I) and transition tear (II) formation. The area encircled by line B and C corresponds to the translaminar tear (III) formation and the area above line C to the undefined tear (IV) range. The slope of line A and B indicates to what degree the increased amplitude can be compensated by shorter sealing times in order to obtain the same seal strength.

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