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Optimizing the heat sealing parameters of multilayers polymeric films

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Abstract Polymer–metal multilayers have been widely used for decades in packaging industry, and more recently for vacuum insulation panels for building application. In both cases, the seal zone could represent a weak area for mechanical and barrier properties. The aim of this report is to investigate the heat sealing properties of multilayers composed of one polyethylene layer and one or three polyethylene terephthalate layers coated with aluminum. The quality of seal was quantified by peeling test and the failure mechanisms. In order to optimize the set of heat sealing parameters, a series of mechanical and morphological relevant parameters were measured and compared to the failure modes. A comparison between the sole sealant film and multilayers was performed in terms of range of optimal heat sealing parameters and mechanical behavior of seals. Although they present a much narrower range of optimal properties, the multilayers films show a strong advantage over the single films.

Introduction

Polymer–metal multilayers have been widely used for decades in packaging industry. More recently similar materials were found useful in buildings, as the envelope of vacuum insulation panels (VIP). Both applications are very demanding because the envelope should withstand a high barrier level to gas and water vapor for long periods of time. As a result multilayers structures may be advantageously employed to fulfill the numerous requirements. For example [1–4, 13], a polyethylene terephthalate (PET) layer may take care of the mechanical properties while the barrier to gas and water vapor could be assured by an aluminum layer. A third polyethylene (PE) layer may additionally be used for its stability and sealability. For both mechanical and barrier properties, the seal could represent a weak zone, since it is the place where no tough material or barrier layer is present, inducing mechanical weakness together with moisture and gas transmission [13].

Heat sealing technique is widely used in the packaging industry [5, 7, 12]. In principle, the two elements to be joined are heated and soften to allow interpenetration of the two surfaces, and thereby promote cohesion [5, 12]. Reproducible and high quality assembly requires the adaptation of the sealing temperature, dwell time, and pressure [5, 12]. Even if the optimization of the heat sealing parameters of single-layer films of semi-crystalline polymers and the understanding of the molecular mechanisms involved in this process have been the subject of many investigations [5, 7, 10–12], the heat sealing of multilayers has been less studied and mainly related to the sealing parameters optimization [8, 9, 13, 14].

In contrast, the sealing of monolith polymer films have been studied and well understood. On the experimental stand point, the testing of heat-seals is commonly performed using the so-called T-peel test. In the simplest case of single-layer films, some failure mechanisms have been identified: “peeling failure,” “tearing failure,” and “peeling and tearing failure” [5, 12]. The seal strength measured on single layer can then be related to the failure mode in an understandable manner [5, 12]. Among the studied parameter, the quality of the sealed zones seems to be

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primarily governed by the sealing temperature. In the case of polyolefin, the seal strength exhibits a maximum on a relatively narrow temperature range typically 20 °C starting few degrees above the melting temperature [5, 12].

The aim of this article is to investigate the heat sealing properties of multilayers and to compare them to that of a single layer. The influence of the layers will be estimated both qualitatively through the failure mechanisms of the sealed zones, and quantitatively with the values of the optimal seal strength.

Materials and methods

Materials

The samples used in this study were provided by Rexor Company (38, France). The first multilayers, named complex 1-1, is composed of two layers: a layer of PET coated with aluminum and a layer of low density polyethylene (LDPE) assembled with an adhesive layer, Fig. 1. The structure of the second one, named complex 3-1, is composed of four layers: three layers of PET coated with aluminum and a layer of low density polyethylene LDPE regularly assembled with three adhesive layers [2–4]. The thicknesses of the films employed in this study are reported in details in Fig. 1. The LDPE layer presents the lowest softening temperature and was thus naturally employed as the sealing material. The LDPE control film will also be sealed and characterized for comparison.

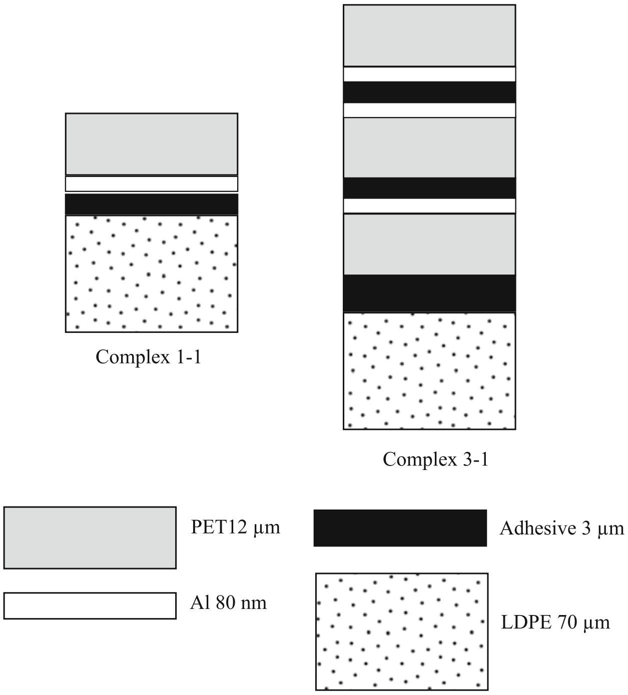


Fig. 1 Structure of the complexes 1-1 and 3-1 used in this study

Table 1 Results of DSC analysis

	1st melting peak		2nd melting peak		$\chi_{PE(\%)}$
	T_m(°C)	Onset(°C)	T_m(°C)	Onset(°C)	
LDPE	108±1	98±1	–	–	39±1
PET	–	–	239±1	248±1	–
Complex 1-1	108±1	98±1	249±1	246±1	39±5
Complex 3-1	108±1	98±1	249±1	246±1	39±5

Methods

Differential scanning calorimetry (DSC)

Polymers may only be sealed above their softening temperature: glass temperature and melting temperature for amorphous and semi-crystalline polymers, respectively [5, 12]. DSC analyses are thus a requirement to reduce the sealing temperature range worth studying. The measurements were carried out on DSC-7 instrument (Perkin Elmer). The instrument was calibrated with indium standard (T_m = 156.6 °C; ΔH_m = 28.4 J/g). The reference was an empty aluminum pan and the average mass of samples was close to 5 mg measured with precision. Measurements were performed in two consecutive heating scans at 10 °C/min in the 25–300 °C range under nitrogen gas. The results of second scans are presented in Table 1.

As shown in this Table, the multilayers (complexes 1-1 & 3-1) exhibit two melting peaks: one at 108 °C and another close to 250 °C, which correspond to melting of LDPE layer and PET, respectively. The position of the melting peak for the polyethylene was the same for the three kinds of samples (LDPE, complexes 1-1, and 3-1). The values of sealing temperatures were thus chosen above 108 °C.

In addition, the crystallinity ratio of polyethylene χ_{PE} can be deduced from these experiments. It was obtained from the total area under the first melting endotherm $\Delta H_{m(PE)}$ and calculated following:

$$\chi_{PE(\%)} = \frac{1}{W_{PE}} \frac{\Delta H_{m(PE)}}{\Delta H^{\infty}_{m(PE)}} \quad (1)$$

with w_{PE} the weight fraction of PE in multilayers (obtained from the thicknesses of different layers composing the film). $\Delta H^{\infty}_{m(PE)}$ is taken equal to 290 J/g [6], and corresponds to the crystalline PE. Given the uncertainties of this measurement, the crystallinity ratio in PE is considered the same for all materials studied in this article and is taken equal to 39±1 wt% for LDPE and 39±5 wt% for multilayers, Table 1.

Sealing

In order to characterize the sealed zones, two squared samples were first cut and superposed to be sealed, Fig. 2. Heat-seals were performed in the laboratory using Medsealer 460 MSID(K) heat sealer (Francopack). This device clamps two pieces of films between flat, 10 mm wide, heated bars. The temperature, pressure, and dwell time of the sealing bars are adjustable. Both bars were operated at the same temperature. The pressure was fixed at 240 kPa, whereas platen temperature and dwell times were varied. After the heat-seal was performed, it was rapidly cooled down to room temperature. Different experimental conditions were tested, varying dwell times: from 1 to 90 s and sealing temperatures from 110 to 180 °C.

T-peel test

The mechanical behavior was characterized with strips of 25 mm wide. These samples were used for T-peel test either at 90° or at 180°. The total length of the samples was fixed at 112.5 mm, Fig. 2.

The samples were peeled at room temperature in the T-peel configuration in an Instron mechanical testing machine, using a 2 kN load cell. For the 90° configuration, one leg was clamped at the grip, the other holding the seal zone, Fig. 2. The specimens were aligned so that the seal line was perpendicular to the tensile direction, Fig. 2. For the 180° configuration, both legs were clamped into a grip, Fig. 2.

The constant rate of loading 100 mm/min was chosen and the force F (N) and displacement (mm) were recorded during the test. For each material and sealing condition, three specimens were tested. In this report, the raw data shows the force divided by the width (F/W) as a function of the displacement. Among the different test, only one representative data was reported.

The peeling test measures the forces required to separate a heat-seal apart. The maximum force ($F_{\max}$) divided by the nominal width (W) is commonly employed to quantify the quality of the seals and defined as apparent seal strength, Fig. 3. In this article, the influence of the number of layer was studied and thus the thickness of the sample was varied. To account for this, a normalized seal strength was defined and utilized:

$$SS = \frac{F_{\max}}{W \times e} \qquad (2)$$

with SS in MPa, $F_{\max}$ in N, W in mm, and e the film thickness in mm.

As shown in Fig. 3, a constant initial slope on the peel curve crosses the x-axis in a well defined but non zero position which can be used to define a d_{onset}. This parameter was found very reproducible and studied as a possible structural indicator of the seal quality. This parameter indeed appeared strongly correlated to seal width measured by microscopy 2.2.5. All values presented here represent the average of the three tests with the corresponding error bars.

In addition, the failures mode of each test was carefully examined and will be further described in the text.

Tensile tests

In addition to T-peel test, more common tensile tests were performed using a universal testing machine, Adamel Lhomargy, equipped with a 100 N load cell. In order to compare with the peeling tests at 180°, the tensile tests were performed on samples in strip form (25 mm in width and 100 mm in length L_0) at a fixed crosshead speed at 100 mm/min. For each material, three specimens were tested. For the comparison between the tensile test and the peeling test at 180°, the graph in this report presents the nominal stress $F/(L_0 e)$ in both cases as a function of the engineering strain : (L/L_0) and $(L/2L_0)$, respectively for tensile and peeling test. For the accurate determination of yield stress σ_y of the studied films, dumb-bells specimens (5 mm in width and 30 mm in length) were tested with the

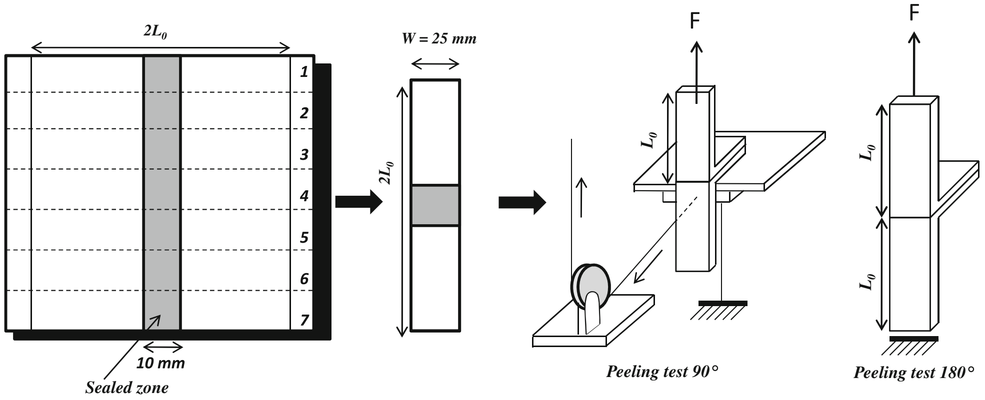
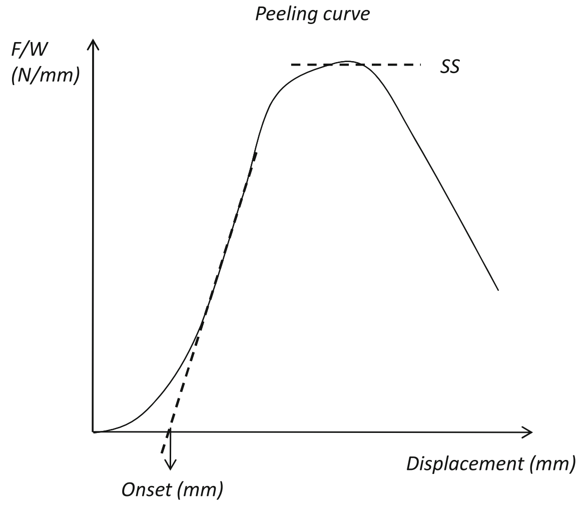


Fig. 2 Preparation of samples for sealed zones characterization



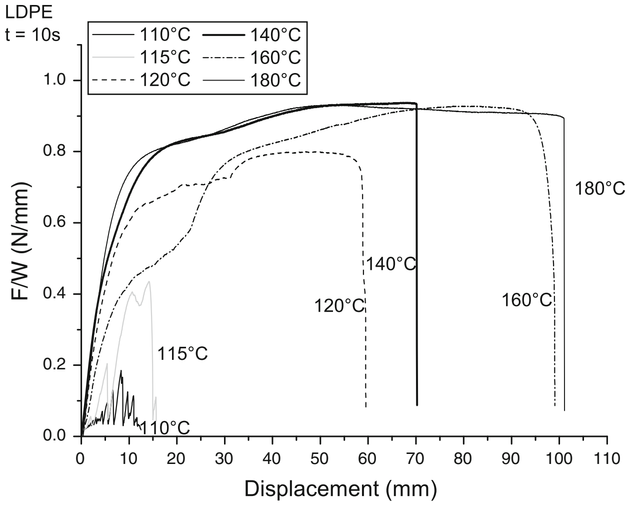


Fig. 3 Peel curves of LDPE films for all sealing temperatures tested for a sealing time of 10 s

same equipment and with again three samples for each material. The tests were conducted at a constant crosshead rate of 5 mm/min, which give a similar initial $\dot{\varepsilon}$.

Optical microscopy

Seal morphologies were studied through images obtained with a precision measurement system: Micro Vu Vertex 410. All observations were performed with at high magnification (×136), to improve resolution. The extracted parameter is the apparent seal width. It was thought that the seal was relatively good if the seal width was close to this nominal value, i.e., the width of the seal bar (10 mm).

Another type of structural observations was also performed to analyze the edges of sealed zones, in which the seals were embedded in epoxy resin and polished. In the latter case, the surfaces were analyzed with a light optical microscope DMLM from Leica in reflection mode.

Results and discussion

Effects of sealing temperature and dwell time for LDPE film

The effect of the sealing temperature was first studied. The peel curves obtained after various sealing temperatures for a dwell time of 10 s are presented in Fig. 3. For the low sealing temperatures (110 and 115 °C), large instabilities in the mechanical behavior can be observed. The corresponding seal strength (*SS*) was relatively low: 0.2 N/mm for 110 °C and 0.4 N/mm for 115 °C. The mechanical behavior became more stable at and above 120 °C, where an important increase in stiffness and seal strength was evidenced together with an appreciable mechanical stability. The seal strength at 140 and 160 °C was for example five times that obtained at 110 °C. The same evolution with the sealing temperature was observed for the others dwell times: 1, 5, 60, and 90 s, Fig. 4. This confirms the observations of Meka and Stehling [5, 12] that the dwell time has less influence on the seals as compared to the sealing temperature. These authors performed accurate experiments and simulations and concluded that dwell time must be sufficient to bring the interfacial temperature to a desired level. A slight pressure is also helpful in bringing two microscopically uneven film surfaces into intimate contact, but higher pressure has no beneficial influence on seal properties [5, 12]. Although not very relevant in the application point of view, this study also shows that long sealing time (90 s) induces an acceptable seal strength regardless of the sealing temperatures, Fig. 4. In other words, a lack of temperature may be counterbalanced by a larger sealing time.

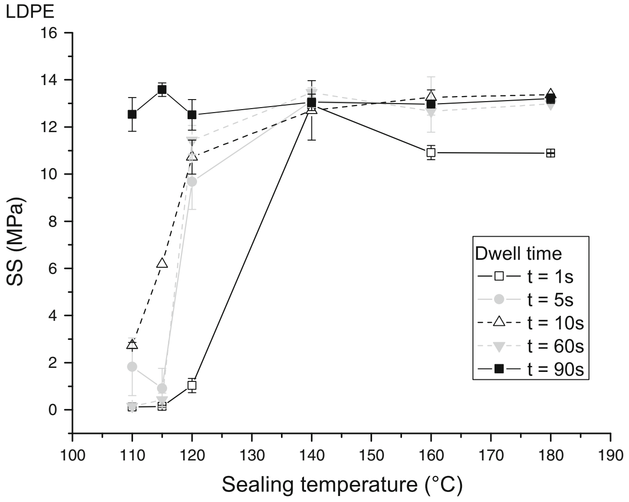


Fig. 4 Seal strength for LDPE films as a function of sealing temperature for different sealing deviations

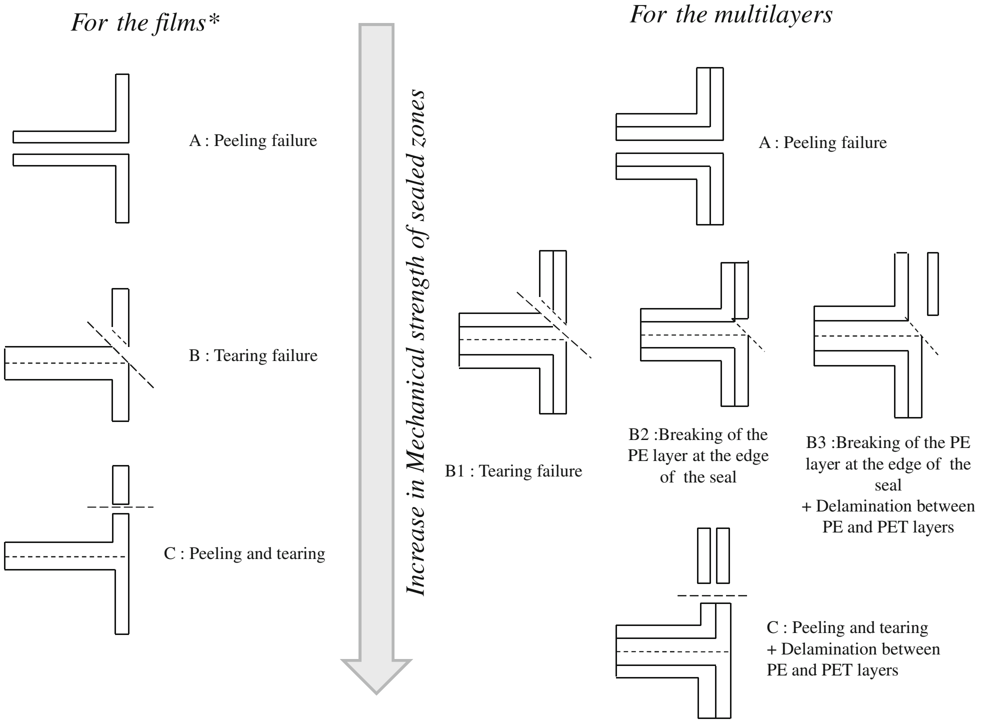


Fig. 5 Schematic illustration of the type of seal failures that can be observed for film and multilayers (*For the one-layer films, this graph is adapted from [5, 12])

The failure modes observed during peel tests can give further information on reason for the seal quality. Figure 5 schematically illustrates the three types of failure described in the literature namely "peeling failure" along the initial contact surface (A), "tearing failure" (B), and a combination of "peeling-tearing failure" (C)[5, 12]. In the latter case the sealed zone is not altered by the mechanical test, and the (C) mode usually corresponds to the maximal strength of the seal. The failure modes observed for pure LDPE (control films) are presented in the Fig. 6a. Three failures modes were observed: the peeling failure (A) for the lowest sealing temperatures (110 and 120 °C) and reasonable sealing times (from 1 to 60 s), the peeling and tearing failure (C) for the higher sealing temperatures: 140

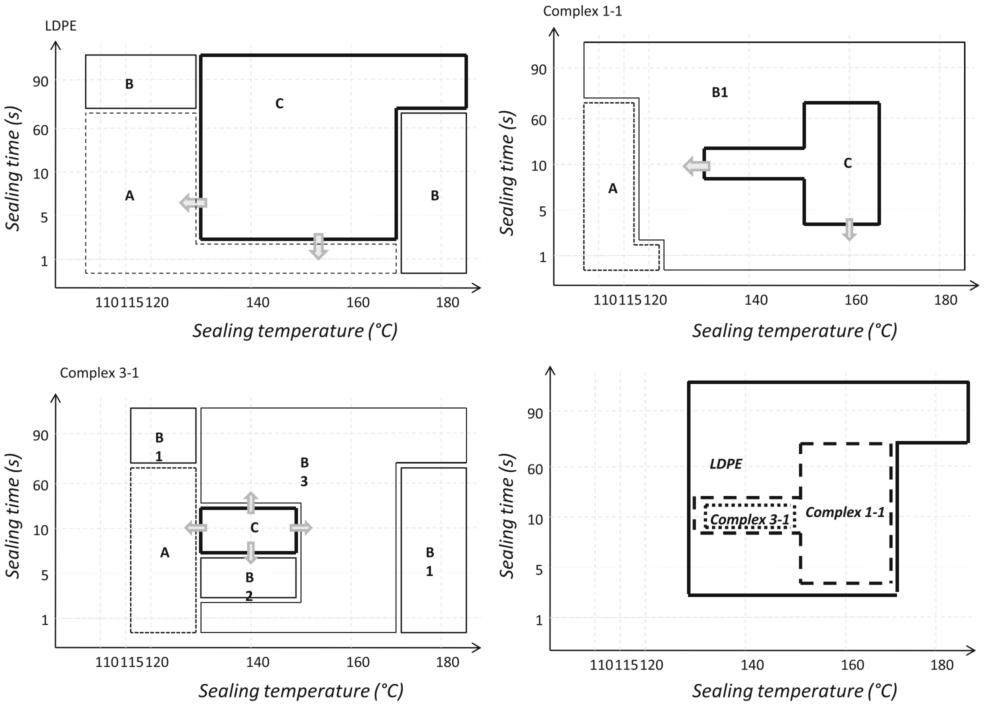


Fig. 6 Failures modes as a function of sealing temperature and time **a** for LDPE film, **b** for complex 1-1, **c** for complex 3-1, and **d** comparison of optimal zones

and 160 °C above 1 s and tearing failure (B) for the highest sealing temperature tested (180 °C) and for the highest sealing time (90 s). For the sole LDPE in this study, it thus appeared that a good sealing was obtained with a temperature from 140 to 160 °C, in agreement with published data [5, 12].

Besides the sealing modes, the *SS* may furnish quantitative information on the sealed zones. This parameter is presented as a function of sealing temperature in Fig. 4 for all dwell times studied. Regardless of the dwell time, *SS* is low at 110 and 115 °C and for sealing temperature too close to the melting point of the polymer (i.e., 108 °C), (A) mode occurs and accordingly at low SS. At high temperatures, SS reaches a plateau level corresponding to (C) mode up to 160 °C and then to (B) mode at 180 °C. The curve for the dwell time of 1 s is shifted to higher temperatures and for higher temperature (160 and 180 °C) the *SS* is slightly lower than obtained for others dwell times, indicating this dwell time of 1 s is not sufficient to bring the interfacial temperature to the desired level. The same phenomenon has been already observed for polyethylene and well explained in references [5, 12]. A maximal seal strength is observed for a dwell time of 90 s for all sealing temperatures, but the (B) mode occurs for the lower sealing temperatures (110, 115, and 120 °C), whereas the (C) mode is observed for others sealing temperatures.

Meka and Stehling [12] showed for monolith materials that the plateau seal strength SS_m is correlated with both amount of crystallinity χ_{PE} and yield stress σ_y, when the nature of polyethylene is varied, Fig. 7. This study permits to quantitatively confirm the data of Meka and Stehling. The experimental results gained with LDPE monolith were indeed placed on the linear data extracted from the literature, Fig. 7. This result essentially reveals that the yield stress σ_y of polyethylene is closely correlated with the crystallinity ratio χ_{PE}. In other words, with monolith films, the optimal seal strength SS_m (140 °C, 10 s) increases linearly with the amount of crystallinity χ_{PE} or equivalently the yield stress σ_y.

Concerning the others parameters extracted from the peel curves, the d_{onset} values are scattered at low sealing temperature, confirming a poor and variable quality of the seal. The instabilities observed in the peel curves with the same sealing conditions may also be considered as a good indicator for poor adhesion, besides the absolute seal strength. The d_{onset} reduces to zero at higher sealing temperature, suggesting a more homogeneous seals along the seal width.

In summary, the sole observation of the failure modes as a function of sealing temperature, and to a lower extent the seal time enables to estimate the set of parameters to optimize the mechanical strength of the seal.

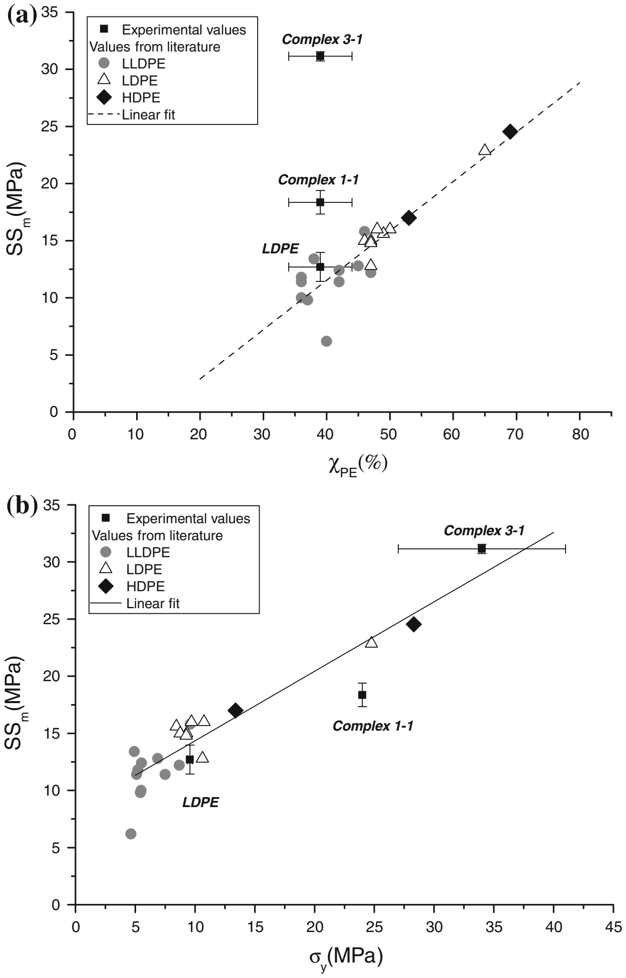


Fig. 7 **a** Seal strength SS_m as a function of crystallinity ratio χ_{PE}, comparison with data of literature [12]. **b** Seal strength SS_m as a function of yield stress σ_y, comparison with data of literature [12]

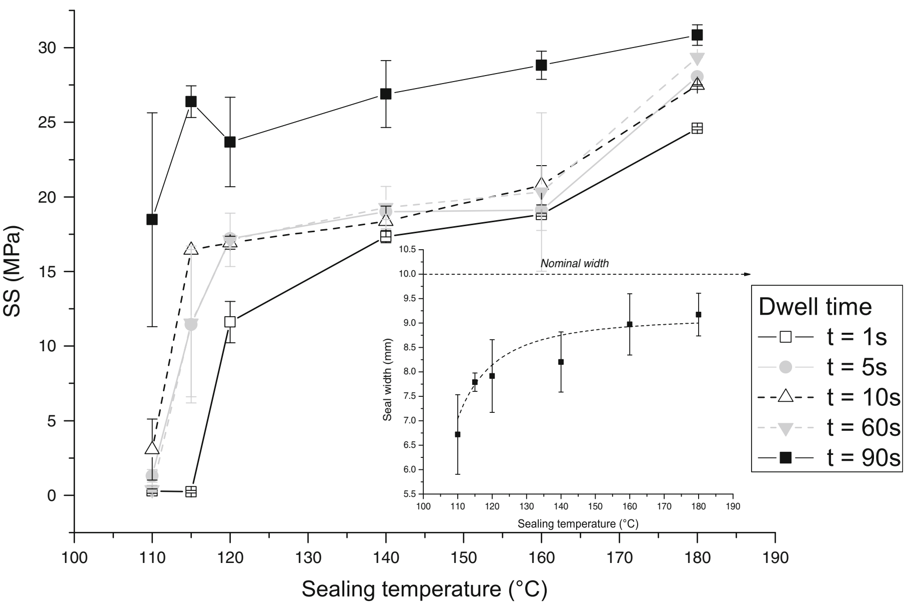
Case of multilayers

As for LDPE film, the effects of sealing temperature and dwell time have been studied for multilayers. Thus, the seal mechanical properties have been characterized by T-peel test at 90° and for each test. Failure modes were carefully examined. The different failure modes observed for multilayers are gathered in Fig. 5. For the two multilayers, the failure modes for each sealing conditions tested are presented in Fig. 6b and c, whereas the *SS* as a function of the sealing temperature for all dwell times is shown in Fig. 8. The observed behaviors are similar in nature to that of pure LDPE. The failure modes seem here again to be primarily influenced by sealing temperature.

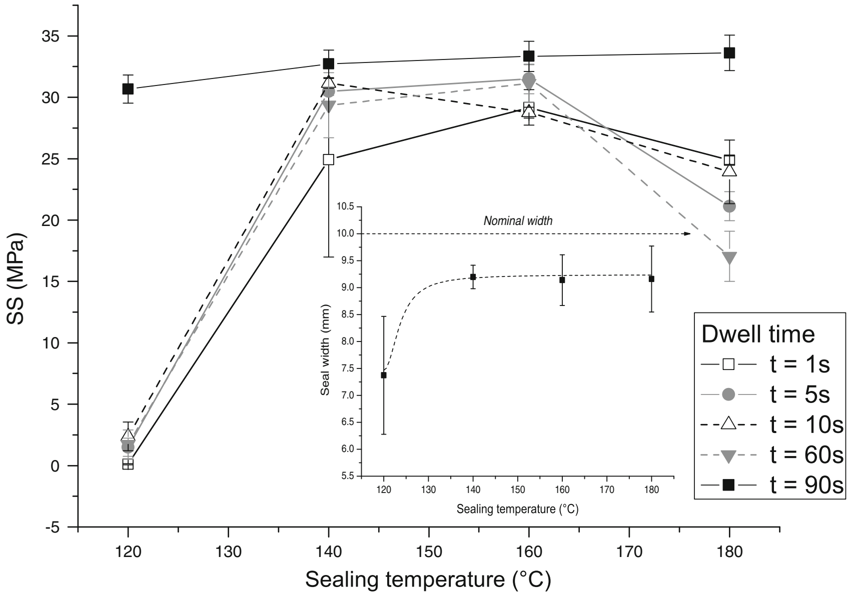
At low sealing temperatures (110 and 115 °C for complex 1-1 and 120 °C for complex 3-1) and for a sealing time between 1 and 60 s, seals were performed at temperature above but close to the melting point of the sealant material, and were consequently found to fail in peeling

Fig. 8 **a** Seal strength *SS* for complex 1-1 with an increasing sealing temperature and time, in *inset*. Evolution of the averaged seal width (for all dwell times except 90 s) with an increasing sealing temperature;
b Evolution of the seal strength *SS* for complex 3-1 with an increasing sealing temperature and time, in *inset*. Evolution of the averaged seal width (for all dwell times except 90 s) with an increasing sealing temperature

(a) Complex 1-1



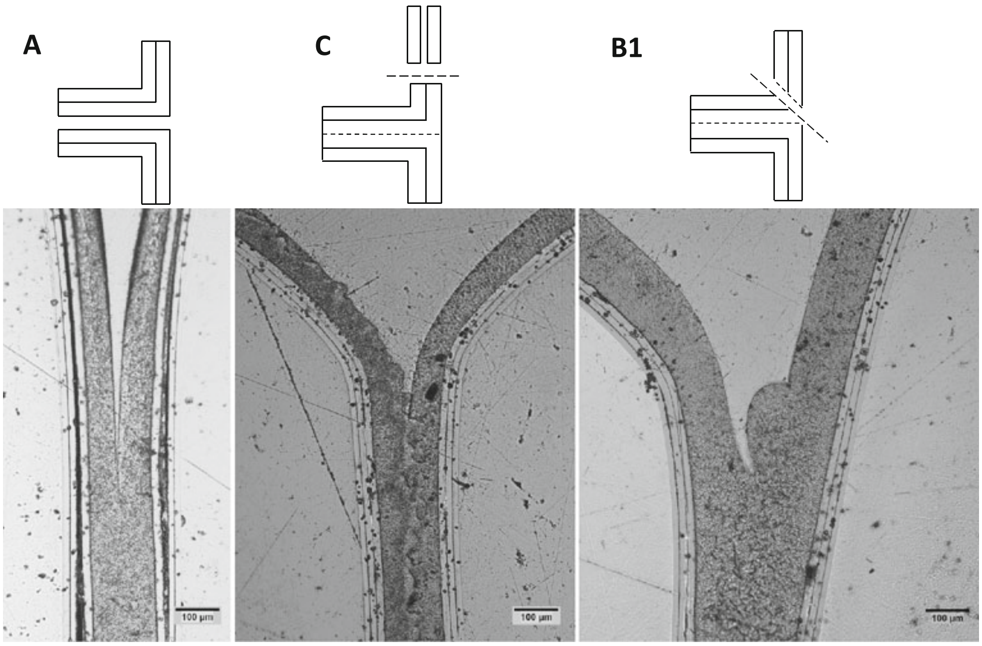
(b) Complex 3-1



mode failure (A), Fig. 6b and c. These sealing conditions also correspond for multilayers to low seal strength, Fig. 8. In contrast with the pure LDPE, no seal can be performed at 110 and 115 °C for complex 3-1. This is likely to be related to the larger thickness of these multilayers (complex 3-1): for the sealing temperature below 120 °C, the temperature reached at the interface between the two LDPE layers is not sufficient and certainly lower than the melting point of the LDPE, not allowing interdiffusion of polymer chains at the interface.

Above 120 °C, the seal strength increases as shown in the Fig. 8 and the failure mode changes to tearing failure modes ((B1), (B2), (B3)) and peeling and tearing mode (C), Fig. 6b and c. As for LDPE film, the evolution of failure modes for a dwell time equal to 1 s is the same as that observed for longer dwell times but shifted to higher sealing temperature. Moreover, for the longest sealing time tested (90 s), a maximal seal strength was observed for the two multilayers regardless of the sealing temperature, Fig. 8. Nevertheless, for these conditions the tearing modes ((B1), (B3)) are observed, which do not correspond to the maximal strength of the seal, Fig. 6b and c.

Even if the study of the multilayers sealing presents some similarities with that of the simple film, a noticeable



120°C – 10 s *140°C – 10 s* *180°C – 10 s*

Fig. 9 Microscopic observations of sealed zones for complex 3-1 for different sealing conditions (the failure modes are indicated on top of the micrographs)

difference was evidenced in the complexity of failure modes, and in the reduction of the optimal time–temperature range, Fig. 6. The variety of failure modes is larger for multilayers and also reveals that the more complex nature of multilayers as compared to the monolith counterpart. The (C) mode still corresponds to the maximal seal strength of the seal, but only occurs for 140 °C-10 s and 140 °C-5, 10, 60 s for complex 1-1 and for 140 °C-10 s for complex 3-1, Fig. 6b and c. In the latter case, the seal strength is greater than the multilayers strength. This range of optimal parameters becomes much smaller with increasing the number of PET layers, Fig. 6d.

As a result, tearing modes ((B) versus (B1), (B2), (B3)) were much more frequent for multilayers, Fig. 6b and c. For these modes, the quality of sealing is better than that observed with (A) mode but theses modes do not prevent a breaking at the edge of the sealed zone. This generated a fracture initiation that propagated quickly in the internal layers. The interface between LDPE and PET breaks first and induces in some cases a delamination process that propagates along this interface. The (B2) and (B3) modes corresponding to a degradation of this interface are only observed for complex 3-1 composed of three PET layers and one PEbd layer. In comparison, only (B1) mode was observed for complex 1-1, indicating the influence of number of PET layers on failure modes. Moreover, these failure modes seem to be in correlation with the morphology of the seal. The microscopic observations (as shown in the Fig. 9) show different seal morphologies depending on failure modes. For example, for complex 3-1 with following sealing conditions : 120 °C-10 s, which corresponds to a (A) mode, a clear separation between the two LDPE layers can be observed, in contrast with (C) mode obtained for 140 °C-10 s, Fig. 9. For the highest sealing temperature (above 160 °C), which corresponds to tearing modes ((B1), (B2), (B3)), the formation of a roll at the edges of the seal was noticed. This can be the reason for breaking of the film of LDPE layer at the edges of seal.

The maximal seal strength obtained on the plateau region for different films, increased with the number of PET layers: 12 MPa for LDPE film, 17–18 MPa for complex 1-1, and 30 MPa for complex 3-1, Figs. 4 and 8. As for LDPE film, SS_m was chosen on the plateau (obtained at 140 °C-10 s) and compared to both the yield stress (σ_y) and crystallinity ratio in polyethylene (χ_{PE}), Fig. 7.

The top of Fig. 7 shows that although the crystallinity of PE is the key parameter for monolith seals, it does not bring information when applied to multilayers. On the contrary, the relationship between SS_m and σ_y is kept even on the quantitative point of view. Complexes 1-1 and 3-1 both lay on the linear relation obtained by varying the crystallinity and established by Meka and Stheling [5, 12]. The latter relation (σ_y–SS_m) seems thus robust and true regardless of the most influencing parameters (crystallinity, number, and nature of layers).

Concerning the other parameters, a decrease in d_{onset} is noticed with an increasing sealing temperature, for dwell times comprised between 1 and 60 s. For a sealing time of

Table 2 Differentiation of failure modes by the study of mechanical and morphological parameters

Failure modes	A	C	B, B1, B2, and B3
Mechanical parameters			
SS (MPa)	Low	Elevated	Elevated
d_{onset} (mm)	Variable, $\neq 0$	0	0
Instabilities	Yes	No	No
Morphological parameters			
Seal width (mm)	$\ll W_{\text{nominal}}$	$\approx W_{\text{nominal}}$	$\approx W_{\text{nominal}}$
Observations	Clear separation between two LDPE layers	Diffusion of two LDPE layers	Formation of rolls at the edges of seal

90 s, the d_{onset} is close to 0. Thus, a homogeneous seal along its width is obtained for sealing temperature above 140 °C regardless of the sealing time, and dwell time above 60 s regardless of the sealing temperature.

The variations of the latter parameter with the sealing conditions can be compared with that of the seal width. If one considers the variation of the seal width with the sealing temperature, Fig. 8, an increase is observed and the seal width is close to the width of the seal bars for complex 1-1 and complex 3-1, respectively after 140 and 120 °C.

Finally, for monolith films and multilayers, it is possible to differentiate the failure modes by studying various mechanical and morphological parameters, as shown in the Table 2. Low seal strengths and seal widths are found for (A) mode, whereas (C) mode induces maximal seal strength (SS) and seal width close to 10 mm. Nevertheless, it is difficult to differentiate the (B1), (B2), (B3), and (C) modes by only studying these mechanical parameters. The values obtained for these parameters for these modes are very similar. A study of seal morphology is necessary to better understand the differences between these failure modes.

Advantages of multilayers for sealing

This study reveals that the case of multilayers has some great advantages as compared to simple layer. The large increase in mechanical strength of the material is one of them. The range of optimal parameters to obtain seals with good quality becomes, however, much smaller in the case of multilayers. An overlap of the optimal parameters enables to determine a globally optimal set of heat-seal condition: 140 °C/10 s, common to all samples. This indicates that the use of multilayers does not alter in depth the best conditions for heat sealing, but reduces the tolerance range. Thus, the heat sealing parameters shall be better controlled in the case of multilayers.

As said previously, the maximal seal strength increases with the number of PET layer, i.e., 12 MPa for LDPE film, 17–18 MPa for complex 1-1 film with one PET layer, 30 MPa for complex 3-1 film with three PET layers, Figs. 4 and 8. The heat sealing conditions, for which the seal strength reaches a maximal value, corresponds to a (C) mode. In this mode, the failure occurs on the leg of specimen, without affecting the seal. Thus, the seal strength is higher than that of the film. But the stiffness of the film is influenced by the number of PET layers. Thus, a higher seal strength is obtained with multilayers because of the presence of stiffer layers than the sealant material in multilayers.

In order to better understand the mechanical behavior of the seal, T-peel test at 180° obtained for a seal with a (C) failure mode (i.e., for the condition 140 °C/10 s for all films) was compared to the response of a tensile test performed on samples in strip form: the sample dimensions were kept constant, equal to that of the leg of the specimen used for T-peel test. This comparison for the LDPE film is presented in Fig. 10. The two curves are very similar, as if the seal was negligible in the T-peel test. Thus, the (C) mode not only correspond to the best within the measurements performed but also is likely to be the best that could be obtained with any way of assembling the layers.

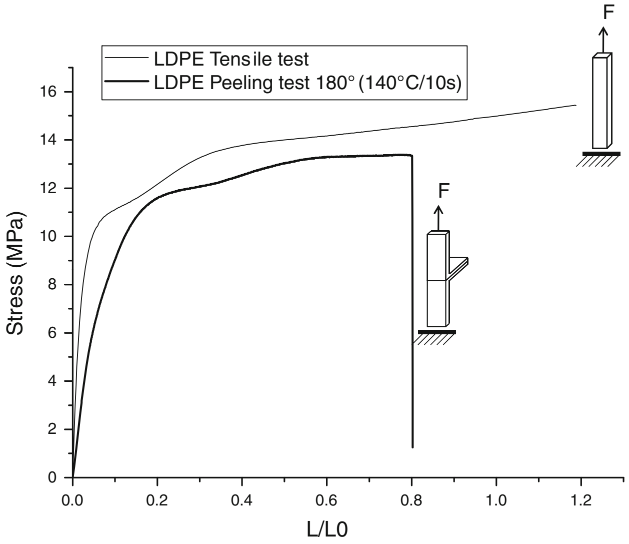


Fig. 10 Comparison between the peel test at 180° and the tensile test for LDPE film

This comparison was performed for the two other kind of sample, namely multilayers, Fig. 11.

The comparison of the three kind of samples in the the T-peel test reveals that the stress level at a given strain for the seal obtained with multilayers is higher than that observed for LDPE film, Fig. 11a. This level can be clearly related to amount of PET within the multilayers.

The tensile behavior of multilayers is improved with the number of PET layers, Fig. 11b. In addition, the complex 1-1 film presents two breaks: the first one corresponds to the PE layer, whereas the second one was attributed to PET layer. For the complex 3-1, a single break was observed that corresponded given the stress level, to the PET layer. In fact, the breakage of the three PET layers induces a large stress that can not be withstood by the ductile LDPE. To further confirm this statement, the strain at break of the complex 3-1 film is higher than the one of the PET layer in the complex 1-1. This can be explained by the presence of the PE layer in multilayers, which decreases the surface defects of PET layer(s) responsible for rupture initiation.

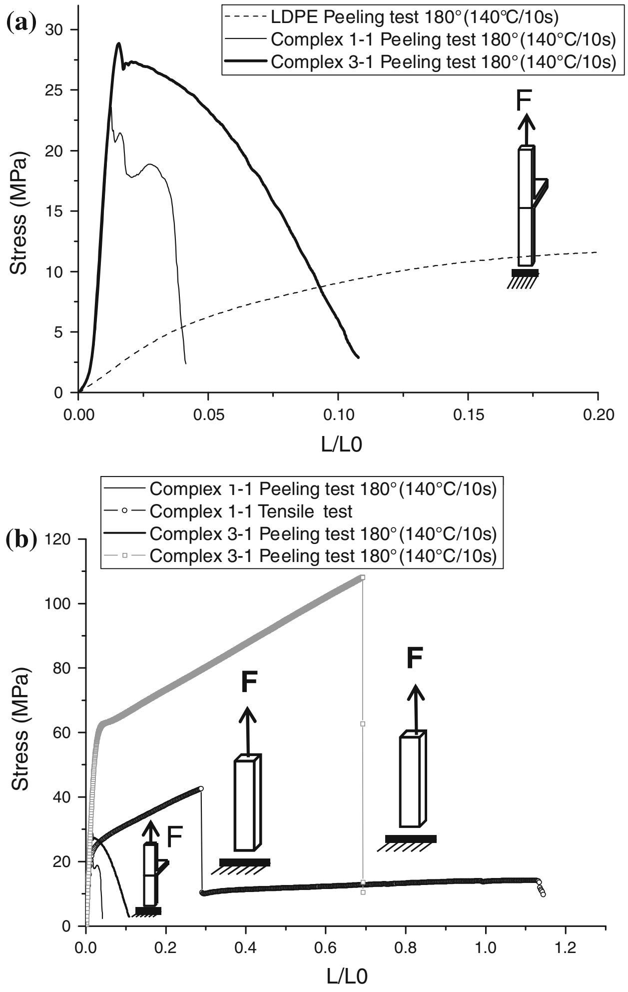


Fig. 11 **a** Peeling curves for the test at 180° of the studied materials. **b** Comparison between the peel test at 180° and the tensile test for complexes 1-1 and 3-1

The Fig. 11b also presents the comparison between the T-peel test at 180° and the tensile test for multilayers. In contrast with the results gained with the LDPE control, the multilayers never quite reached in T-peel test its mechanical properties in a tensile test. In other words, the multilayers, even if the (C) mode, maintain a level of weakness induced by the seal. This likely means that further optimization could be performed, may be by varying the width of the seal or by further optimization of the sealing conditions that would better preserve the structure of PET without inducing localized stress.

Conclusions

In this article, the heat sealing properties of multilayers are studied and compared to the sealant material, i.e., LDPE film. The sole observation of failure modes enables to estimate the set of parameters to optimize the mechanical strength of the seal. In fact, the "peeling and tearing mode" (named (C) in this article) corresponds to the maximal strength of the seal. This is confirmed on the one hand by the evolution of the seal strength versus sealing temperature or dwell time. On the other hand, for this failure mode, the curves of the tensile test and the peeling test at 180° were similar as if the seal did not exist for the peeling test.

Even if the study of multilayers sealing presents some similarities with the one of simple film: the range of optimal heat sealing parameters for example, which was very close to the one of LDPE film, some differences were evidenced. A reduction of the optimal time–temperature range was observed; this latter becomes much smaller with an increasing number of PET layers. Moreover the variety of failure modes also reveals the more complex nature of multilayers and this study does not enable a complete understanding of the relationship between failure modes and the maximal seal strength of the seal. For example, it is difficult to differentiate the tearing modes ((B) for monolith film, (B1), (B2), and (B3) for multilayers) and the peeling and tearing mode (C) by only studying the maximal seal strength SS_m.

A very interesting advantage of multilayers is presented in this study; it concerns the influence of stiffer layers (i.e., PET), compared to the flexible sealant layer (i.e., LDPE), on the mechanical behavior of the seal, since its maximal strength increases with the number of PET layers. Thus, the seals obtained from multilayers composed of stiffer layers may induce an improvement of the mechanical properties as compared to the sole flexible sealant material.

Finally the relation between the maximal seal strength SS_m and the yield stress σ_y showed by Meka and Stehling [5, 12] for simple film in PE seems also to be valid for multilayers, even if the crystallinity ratio is not the most influencing parameters.

This study has allowed highlighting the interest of multilayers for sealing. It would now be interesting to study other sealant materials in multilayers and more particularly the influence of the number, the nature (in chemical, mechanical, etc terms) of the different layers of the multilayers. The relationships between the failure modes and the mechanical behavior of seals must be better understood to not only improve the choice of materials composing the multilayers but also the strategy of layers stack necessary following the intended application. Other characterization methods shall be developed to better fulfill these requirements.

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References

1. Brunner S, Gasser Ph, Simmler H, Wakili KGhazi (2006) Surf Coat Technol 200(20–21):5908
2. Garnier Geraldine, Brechet Yves, Flandin Lionel (2009) J Mater Sci 44(17):4692. doi:10.1007/s10853-009-3724-0
3. Garnier G, Chehab B, Yrieix B, Brechet Y, Flandin L (2009) J Mater Sci 44(20):5537. doi:10.1007/s10853-009-3775-2
4. Garnier G, Yrieix B, Brechet Y, Flandin L (2010) J Appl Polym Sci 115(5):3110
5. Meka P, Stehling FC (1994) J Appl Polym Sci 51(1):89
6. Minick J, Moet A, Hiltner A, Baer E, Chum SP (1995) J Appl Polym Sci 58(8):1371
7. Mueller C, Capaccio G, Hiltner A, Baer E (1998) J Appl Polym Sci 70(10):2021
8. Poisson C, Hervais V, Lacrampe MF, Krawczak P (2006) J Appl Polym Sci 101(1):118
9. Poisson C, Hervais V, Lacrampe MF, Krawczak P (2006) J Appl Polym Sci 99(3):974
10. Qureshi NZ, Rogunova M, Stepanov EV, Capaccio G, Hiltner A, Baer E (2001) Macromolecules 34(9):3007
11. Qureshi NZ, Stepanov EV, Capaccio G, Hiltner A, Baer E (2001) Macromolecules 34(5):1358
12. Stehling FC, Meka P (1994) J Appl Polym Sci 51(1):105
13. van Malsen J, Tenpierik MJ, Looman RHJ, Cauberg JJM (2008) J Plast Film Sheet 24(1):35
14. Yuan CS, Hassan A, Ghazali MIH, Ismail AF (2007) J Appl Polym Sci 104(6):3736