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A sensitive method to determine the bonding strength between two flexible non-compatible polymers

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The influence of different surface morphologies and compatibilizers on the bonding strength between high-density polyethylene (HDPE) and isotactic polypropylene (iPP) is determined by T-peel tests. To optimize the relative comparison, the polymer foils are prepared to have distinct areas with different bond strengths. The advantage of this comparative method is the observation of significant differences of the bonding strength within a single sample, eliminating such sensitive parameters as the dimensions and peel angle. © *1998 Kluwer Academic Publishers*

1. Introduction

Laminated polymer films became of technical importance because of their properties, such as mechanical strength, ability to act as a gas or moisture repellent, heat resistance, and dyeability. The adhesion and bonding strength between two non-compatible polymers are key factors in the laminates and have been the subject of many investigations in recent years. For two flexible polymer films the T-peel test is the most convenient method to compare the natures of the interfaces on the bonding strength and has been used in the recent years as a standard method [1–6]. Peel tests do not supply absolute material data, however, but only a relative comparison, and even the peel tests have to be treated with care because of the complex stress conditions during the peeling. The relation between peel strength and interfacial strength depends on the mechanical properties, such as Young's modulus and shear modulus or yield strength, and on the dimensions of the specimen. Therefore, many authors have tried to calculate (separate) the energy of adhesion from the measured peel force with the precise knowledge of the sample dimensions and their mechanical properties [7–11].

In our examinations, T-peel tests (90°) have been performed in order to correlate different compatibilizers to increase the bonding strength between iPP and HDPE laminates. Knowing the problems of this test method it is important to keep the testing conditions such as peel angle and thickness of the polymer foils strictly constant.

Additionally, in order to compare the influence of the surface treatments, single samples with differently treated surface areas are used in our experiments.

The final goal of this research work will be the optimization of polymer surfaces with respect to adhesion. Examples presented in this paper are the use of ethylene propylene (EP) block copolymers for the bonding of HDPE on iPP surfaces. A–B diblock copolymers are known to act as compatibilizers for interfaces of two incompatible polymers A and B [12–14]. The effect of EP block copolymers on the interfacial interactions between polyethylene and polypropylene was examined in recent years [15–18], and some additional results will be presented here.

2. Experimental

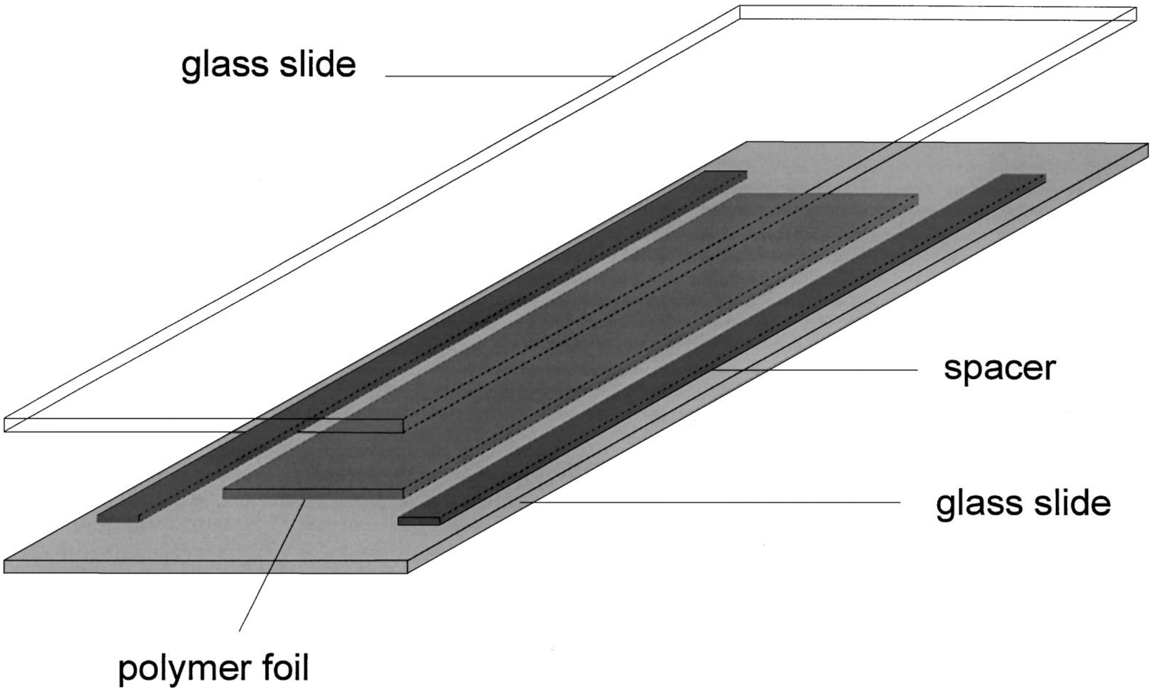
The materials used in our examinations are Vestolen A6013 (HDPE), Vestolen P6000 (iPP), Vestolen P7700 and P9500 (EP block copolymers), all from Vestolen GmbH, and EOD 93-06 (syndiotactic polypropylene [sPP]), which was kindly supplied by the FINA Corporation.

2.1. Sample preparation

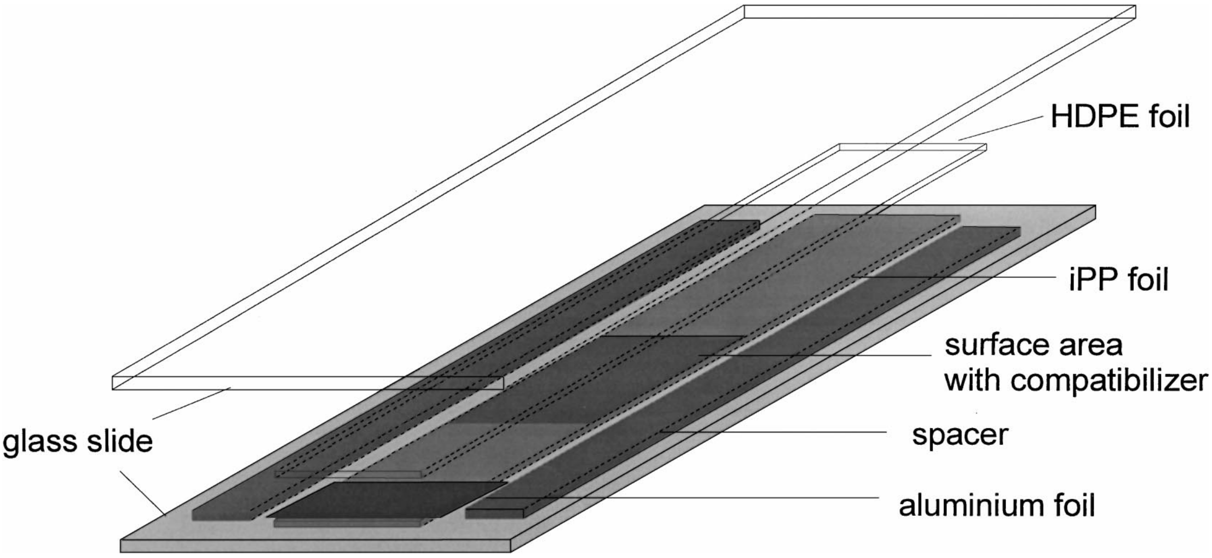
Examinations of the influence of the laminate thickness and the surface roughness were carried out with sPP and HDPE. The separated polymer foils were prepared in a laboratory heating press from granulated polymers. To prepare different surface roughnesses, the HDPE substrate foils were pressed between rough metal sheets and glass slides, respectively. The use of spacers keeps their thickness exactly constant (Fig. 1a). The sandwiches were laminated at $T_p = 128\,^\circ C$, below the melting point of HDPE and above the melting point of sPP.

For the investigations of the bonding with the EP copolymer as compatibilizer, the system iPP and HDPE was used. To ensure polymer laminates of strictly the same low roughness ($< 5\ \mu m$) and constant thickness, the polymer foils were melted between two microscope glass slides with spacers of 150 μm.

The EP copolymer was put onto the iPP surface from a solution. A small amount of a 1 wt% solution of EP in xylene was dropped on a distinct area of the



(a)



(b)

Figure 1 (a) Sketch of the polymer foil preparation, (b) sketch of the sandwich preparation.

iPP surface. After the solvent was evaporated, a HDPE foil was placed on the iPP foil, and the sandwich was subjected to a pressure of 2 MPa. To enhance interdiffusion, the sealing temperature was $T_p = 190\,^\circ C$, well above the melting point of the two polymers. With spacers of 250 μm the sandwiches obtained a well-defined thickness. A small aluminium foil separated the sandwich at the edge and ensured the fixing in the tensile test machine (Fig. 1b).

Test samples of a width of 15 mm and a length of 25 mm were cut from the laminated foils.

2.2. Mechanical testing

The bonding strength of the laminates was measured with the T-peel test (90 °) in a tensile test machine (Zwick 1445) at room temperature. The strain rate used was 10 mm min^{-1} for all measurements. The gauge length of each sample was at least 20 mm. The average peeling load was determined from the peel curve in newtons per millimeter (sample width = 15 mm). The values presented in the figures are averages of at least five measurements.

2.3. Scanning electron microscopy investigations

Scanning electron microscopy (SEM) micrographs were obtained from fractured (peeled) HDPE surfaces by a Hitachi S 4500 field emission low-voltage SEM (LVSEM). The advantage of the low voltage technology is to obtain a topographic image of the polymers without surface coating.

2.4. Calorimetry

The device used was a DSC 2920-system from TA-Instruments coupled with a TA-200 control system.

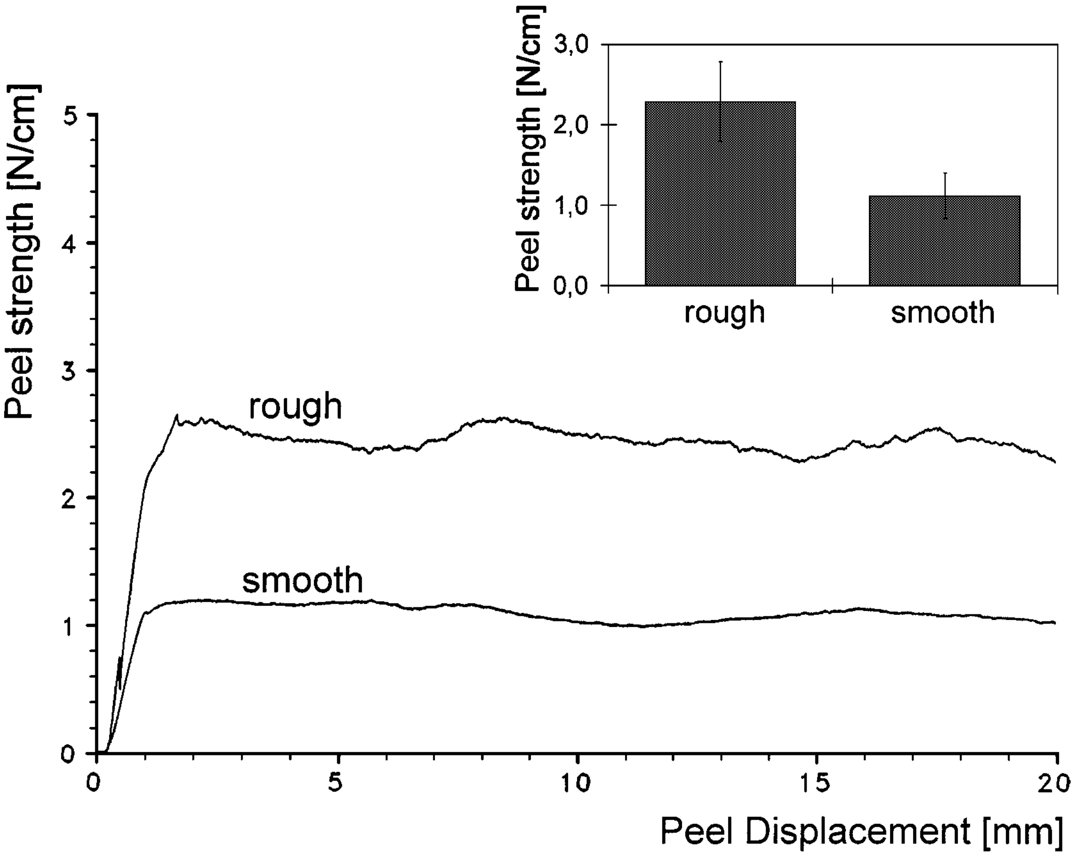


Figure 2 Peel test diagrams of rough ($R_A = 60\ \mu$m) and smooth ($R_A = 4\ \mu$m) surfaces (substrate: HDPE, deposit: sPP, $T_p = 128\,^\circ$C).

Measurements were carried out with heating or cooling rates of 10 K/min. The weight of all samples was 10 mg.

2.5. Atomic force microscopy

The roughness of the examined substrate surfaces was determined by atomic force microscopy (AFM) (Rastercope TM 3000) in the contact mode.

3. Results and discussion

Several factors influence the adhesion between two polymer surfaces. One is the mechanical interlocking [19, 20]. The influence of the surface roughness, which is the main parameter for the mechanical interlocking, is shown in Fig. 2 for the peel strength of sPP on HDPE. Rough surfaces with the possibility of interlocking increase the surface strength and worsen the significance of the measurements. Fig. 3 shows the LVSEM

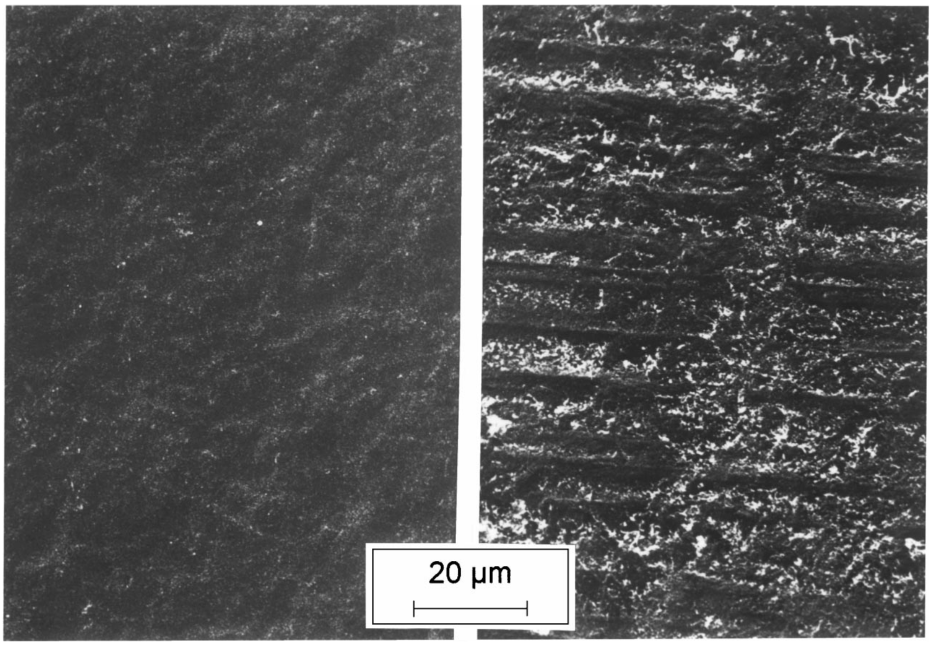


Figure 3 LVSEM photograph of smooth (left, $R_A = 4\ \mu$m) and rough (right, $R_A = 60\ \mu$m) HDPE surfaces (1 kV).

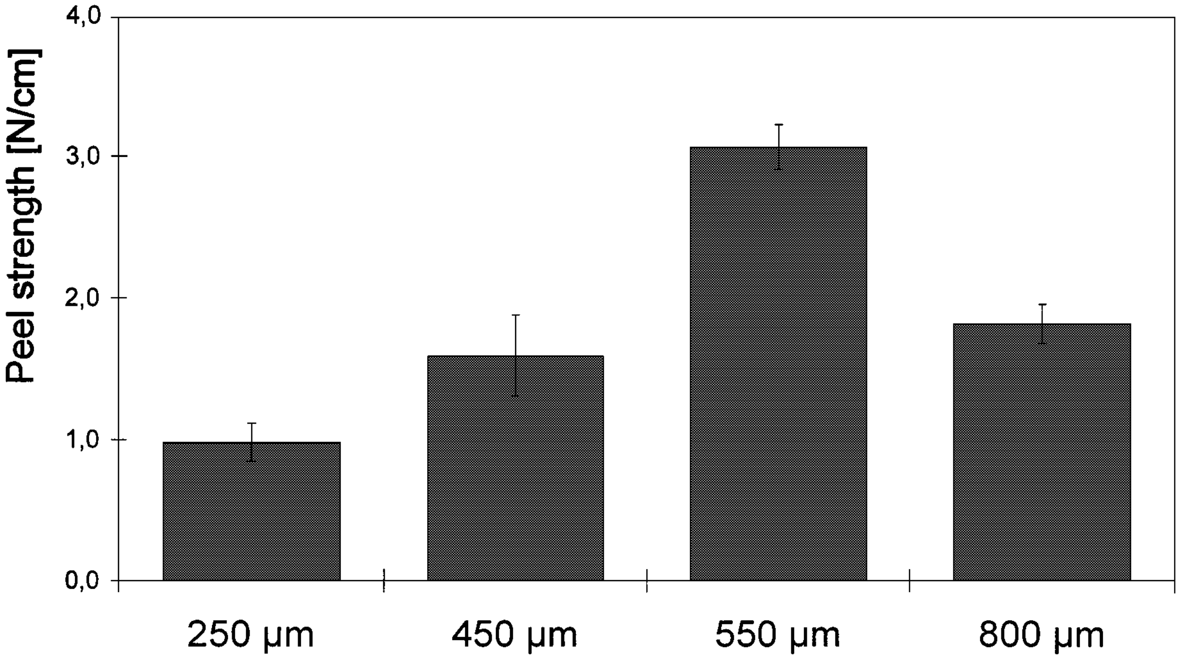


Figure 4 Peel strength versus sandwich thickness (substrate: HDPE, deposit: sPP, $R_A = 4\ \mu$m, $T_p = 128\,^\circ$C).

pictures of the surfaces employed. The roughness R_A was 60 μm and 4 μm for the rough and smooth polymer surfaces, respectively. The peel test results agree with Matting and Ulmer [19] and Kaliske [20]. The bonding strength can be doubled with rough surfaces, but the high adhesion forces due to mechanical interlocking may cover other influences.

Another parameter that may overshadow the intended effects is the dependence of the peeling force on the thickness of the polymer foils. Fig. 4 shows the influence of increasing thickness. After a nearly proportional increase with the thickness, the peel load reaches a maximum. The results are in good agreement with Gent and Hamed [9]. As the thickness increases, more

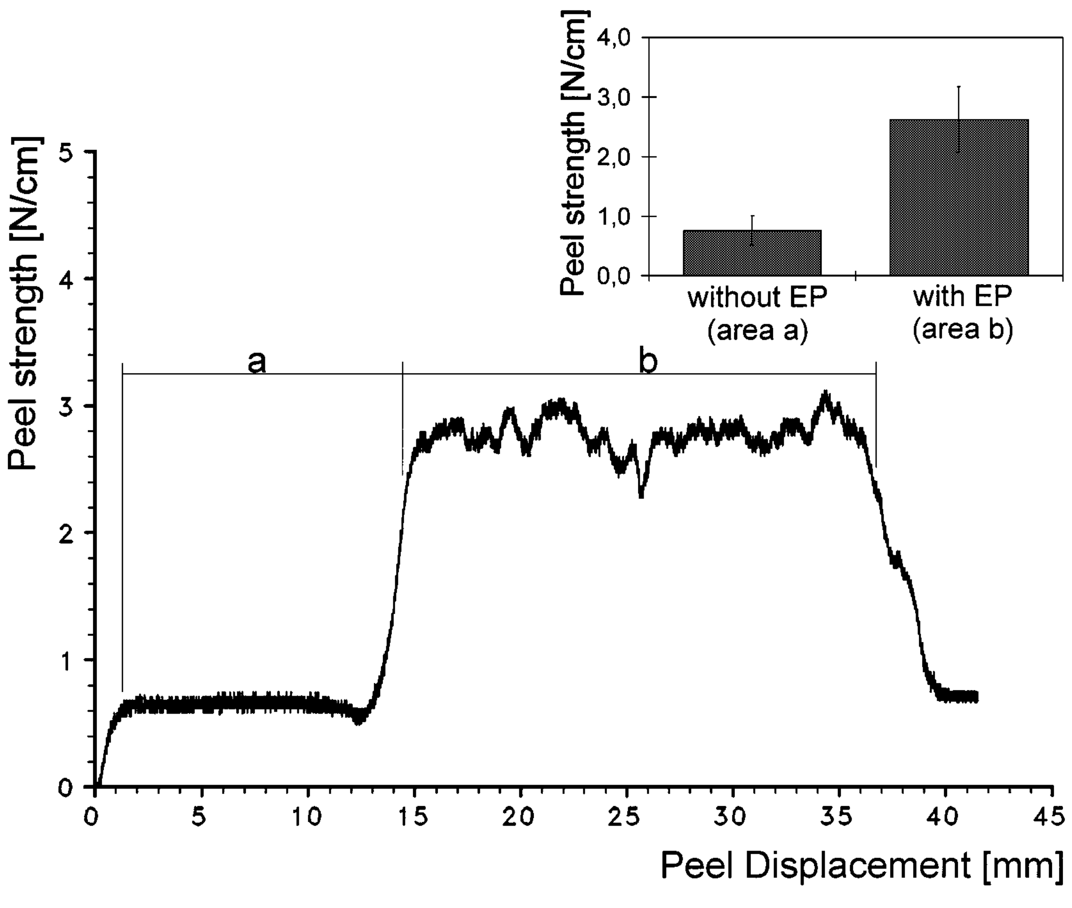


Figure 5 Peel test diagram of a partially EP solution–treated sample (iPP/HDPE, T_p = 190 °C, (a), untreated, (b), with EP7700).

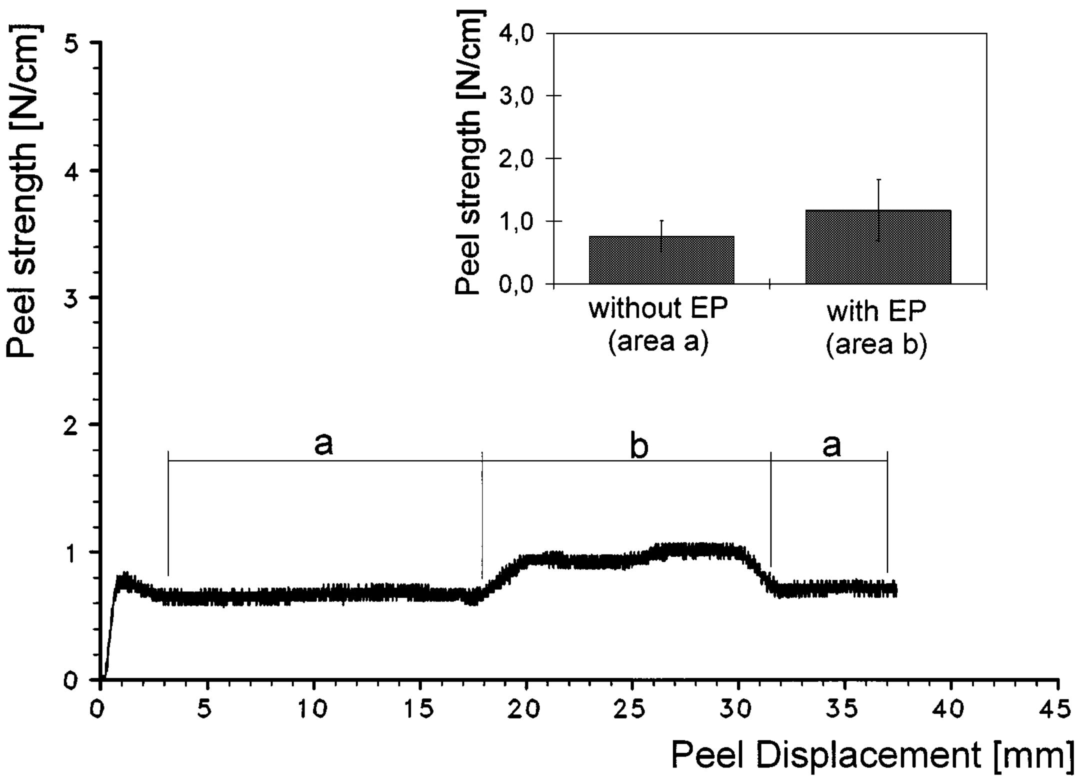


Figure 6 Peel test diagram of a partially EP solution–treated sample (iPP/HDPE, $T_p = 190\,^\circ C$, (a), untreated, (b), with EP9500).

energy by plastic deformation of the whole laminate is dissipated and the measured peel force therefore increases. When the thickness exceeds t_c, no plastic yielding of the laminate will occur. The critical thickness t_c is given by Gent and Hamed [9]:

$$t_C = 6EP/\sigma_y^2, \qquad (1)$$

where P denotes the peel force per unit width in the absence of plastic yielding, E the Young modulus, and σ_y the yielding stress. Above this critical thickness the peel force represents a direct measure of the adhesive fracture energy.

With the present improved preparation methods, the influence of the block copolymers as compatibilizer was examined. All sandwiches were thoroughly prepared with constant thickness of 250 μm (which was found to be an optimum for the experimental procedure) and smooth surfaces ($R_A < 5\,\mu$m).

The EP block copolymer adheres to the incompatible polymers polyethylene and polypropylene. The influence of the copolymer EP 7700 is shown in Fig. 5. The

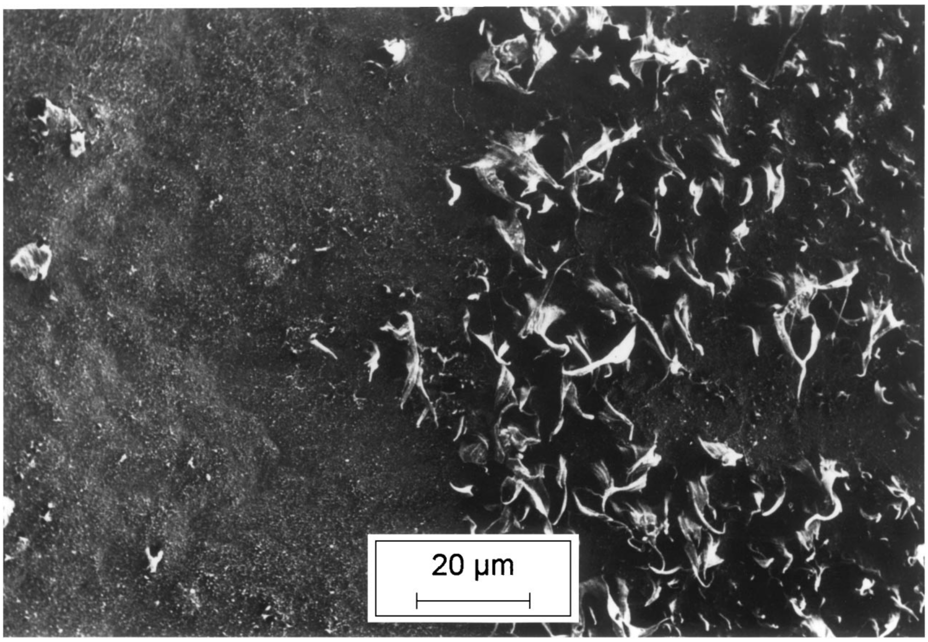
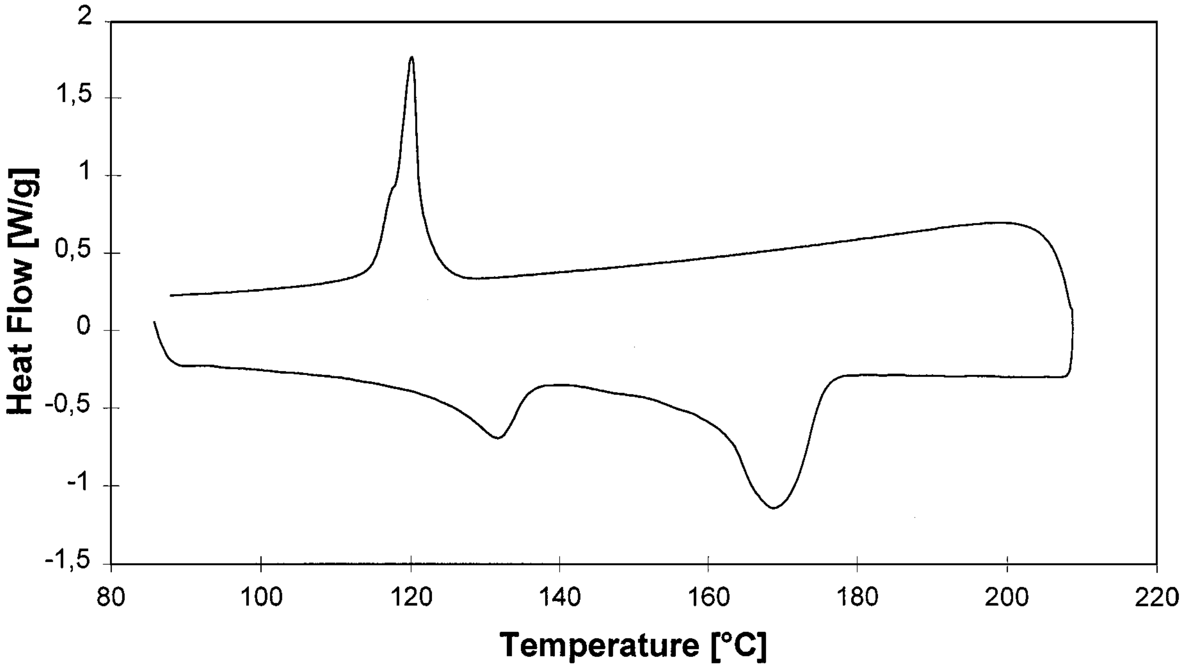
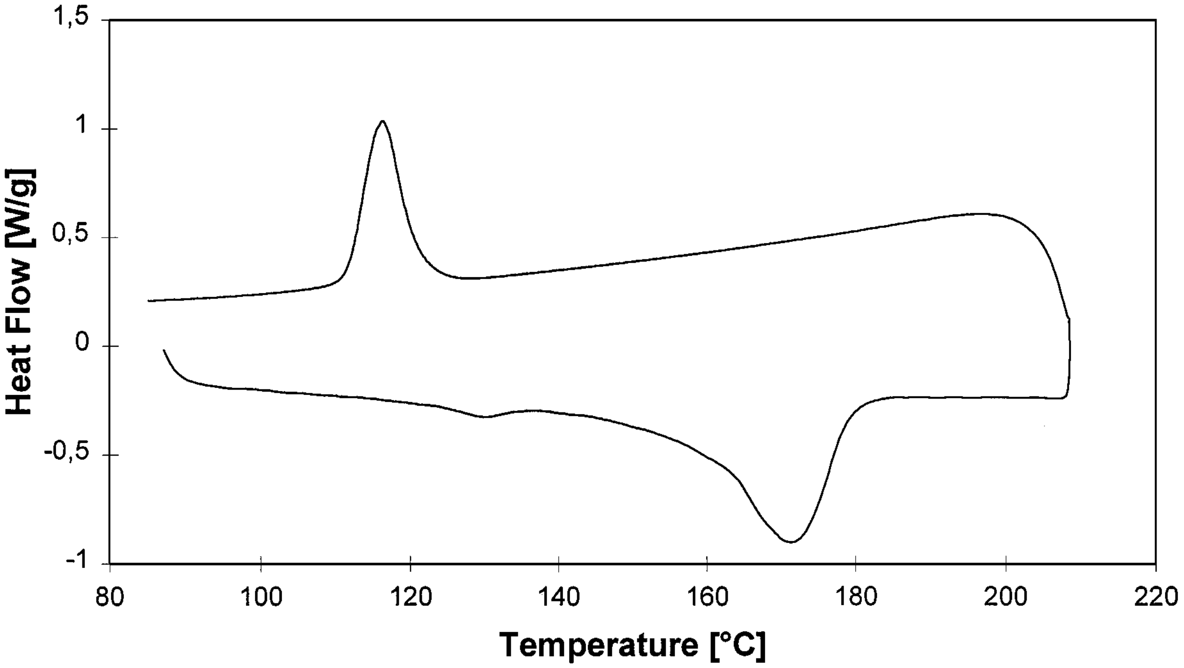


Figure 7 LVSEM photograph of a peeled HDPE surface at a boundary: left area: with EP7700, right area: without EP7700 (1 kV).



(a)



(b)

Figure 8 DSC scan for the EP copolymers (heating/cooling rate: 10 K/min) (a) EP 9500, (b) EP 7700.

peel strength is constant in the untreated area (a) and increases in the treated area (b). The improved bonding strength can also be estimated from the bar chart inserted into Fig. 5.

The advantage of the relative comparison is apparent in Fig. 6. The peel test diagram exhibits an improved adhesion in the areas covered with the copolymer EP 9500 (area b). Because of the large error range, the results are not significant in the bar chart diagram. This peel test diagram illustrates the advantage of this selective preparation method. The improvement of the adhesion would be difficult to establish without the relative comparison method.

The deformation on the peeled surfaces is shown in Fig. 7. The SEM photograph presents the boundary between the treated and the untreated part of the HDPE surface. The left area is a smooth and undeformed surface; the right area shows destroyed lamellae from plastic deformation of the bulk due to the high adhesive strength.

The higher peel force measured for EP 7700 compared with EP 9500 can be explained by their different structures, deduced from differential scanning calorimetry (DSC) measurements (Fig. 8).

Although the EP 7700 exhibits melting endotherms of the PE (132 °C) and the iPP (168 °C) components, the EP 9500 only has the melting endotherm of the iPP. This indicates that the PE blocks in the copolymer EP 7700 are long and perfect enough to form PE crystallites (no data were available from the company). This is not the case for the copolymer EP 9500. Long, perfect blocks may be the better molecular configurations for the block copolymer compatibilizers because it can cocrystallize with the homopolymers in the laminates.

Further investigations are in progress, especially concerning the influence of the morphologies close to the interfaces in the laminates (transcrystallization, epitaxy) [21].

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