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Improvement in the mechanical strength of asymmetric polyolefin model interfaces

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The influence of two types of a styrene–ethylene/butylene–styrene (SEBS) triblock copolymer, one as received and the other functionalized with maleic anhydride, on model interfaces of high-density polyethylene (HDPE) and isotactic polypropylene (iPP) was investigated. Using a special preparation technique, it was possible to prevent gross interdiffusion at the interface and to attribute the problem to an adhesion-dominated phenomenon. The weak mechanical strength of the unmodified interfaces, iPP/iPP and HDPE/HDPE, was greatly improved by the triblock copolymer as was shown by the results of a T-peel test. The morphologies of the peeled surfaces of the modified and unmodified interfaces were analysed with light microscopy. The morphology of both model interface systems show differences, thus revealing different processes at the interfaces and thus different interactions of SEBS with iPP and HDPE and a different influence of the functionalization. The best results were achieved using the functionalized SEBS; the influence of functionalization was greater in the HDPE system. The results are in good agreement with a model proposed by the present authors elsewhere for the concentration-dependent role of SEBS in binary iPP/SEBS and ternary iPP/PE/SEBS blends. © *1998 Chapman & Hall*

1. Introduction

In recent years, the research on multicomponent blends has increased because of both academic and industrial demand. The need for new materials has promoted fundamental research in polymer science to develop tailored materials for special applications to meet new market needs [1–5].

The properties of many polymers can be improved by adding at least a second component, thus creating a heterogeneous system with interesting new properties. Of great importance is, for example, the combination of the polyolefins polyethylene (PE) and polypropylene (PP) [4, 6–10]. In this system, PE is added to PP to improve some properties such as the impact resistance and to expand the applicability of PP. The improvements achieved are sometimes explained as a consequence of synergistic effects [6, 11]. Unfortunately, PP and PE are immiscible in a thermodynamic sense over the whole composition range. Therefore, the resulting properties of the blend often do not reach its possible optimum. Phase separation occurs, resulting in a broad inhomogeneous dispersion of the minor phase. In addition, poor adhesion between the components has a negative influence on the transfer of load between the components.

A method to overcome these problems is the use of interface-modifying polymers, so-called compatibilizers. These polymers, usually block or graft copolymers, should locate at the interphase between the immiscible components in such a way that they will traverse the interphase [11–15]. This provides better adhesion and leads to an improved transfer of load between the matrix polymer and the occlusions of the dispersed minor phase (Fig. 1). Compatibilizers should also have an emulsifying effect, e.g., by altering the surface tension in the interphase. This can lead to better dispersion of the minor phase in the matrix.

Much research has been done to develop appropriate compatibilizers for special immiscible systems to gain insight into some first-order requirements [11, 12, 16–20]. In general, a polymer which has been chosen to compatibilize a given heterogeneous system has to cause the following modifications to achieve controllable changes in properties:

(i) prevention of gross segregation to hinder phase separation on a macroscopic level;
(ii) allowing a finer dispersion of the minor component in the matrix leading to a homogeneous unimodal size distribution of the minor phase occlusions with optimal distance statistics;
(iii) improvement in the adhesion between the immiscible components to achieve a good transfer of load through the interface;
(iv) variation in the microrheological properties for improved processability and morphology stabilization.

It is clear that not all requirements can be fulfilled using just one compatibilizer, but often one is interested in improving only one or two properties of

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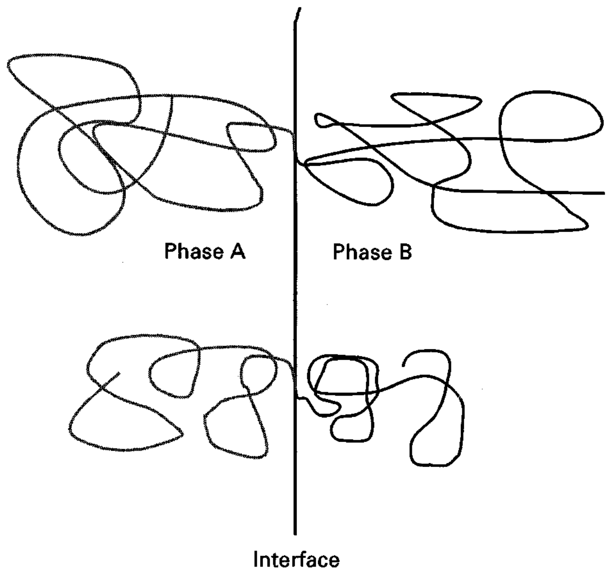

Figure 1 Location of a blockcopolymer AB at the interface between component A and component B.

a commercial polymer blend without destroying the overall or typical profile of the material. This is usually the case with commercially available block or graft copolymers which are not specially designed for a desired application.

Recently it was found that styrene–ethylene/butylene–styrene (SEBS) triblock copolymer enhances the impact resistance of PP/PE, PP/polycarbonate and PE/polystyrene (PS) blends considerably [21–25]. It was reported that SEBS acts in these systems as an interfacial agent [21, 25–27]. The concept that SEBS modifies the interface is for the system PE/PS obvious with regard to Fig. 1, but for PP/PE it is quite unusual. Owing to its architecture, SEBS should only compatibilize mixtures of styrene compounds with polyolefins or polyphenylene ethers and polyolefins. On the other hand, it is known that some properties (i.e., the impact resistance or the tear strength) of common large-volume thermoplastics such as PS, PE and PP can be modified using PS/elastomer block copolymers to form binary blends [28, 29].

Paul and co-workers [27, 30] investigated the modification of poly(ethylene terephthalate)(PET))/high-density polyethylene (HDPE) blends by SEBS. It was found that as a result of the modification the mechanical properties changed from almost friable to remarkably tough. This is interesting in so far as neither PET nor HDPE is identical with one of the copolymer segments.

Similar results have been found by the present authors for the system isotactic polypropylene (iPP)/PE [31–34]. A concept for the role of SEBS in iPP/PE blends has been proposed [31, 34] including the possibility that SEBS forms a thermoreversible network through physical cross-linking of the SEBS in the iPP matrix [35].

A schematic presentation of a thermoreversible network based on a thermoplastic elastomer is shown in Fig. 2a. The A domains of the ABA triblock copolymer (i.e., the styrene blocks in SEBS) are located in their own clusters because of microphase separation. Phase separation leads to a domain structure consisting of areas of pure A block composition, areas of pure B block composition and an area of mixed composition which constitutes the interface volume. If the polymer architecture of the block copolymer allows ties between domain areas (such as a triblock ABA copolymer) a phase-separated network structure results. The criteria for microphase separation into the domain network structure is dependent on the Flory–Huggins interaction parameter, χ_{AB}, i.e., on the squared difference in solubility parameter. In [36], a criterion for this parameter was derived for hydrocarbon block copolymers and it was shown that the interaction parameter for a SEBS block copolymer was clearly above the critical base value. The equilibrium morphology of styrene block copolymers with a PS-elastomer ratio smaller than one consists of phase-separated A domains dispersed in a matrix of the B phase. Owing to the triblock architecture of the ABA copolymer, a network structure is formed by bridging between domains of A blocks, which are located in different A domains. The bridges or links are formed by the elastomer block B of the ABA copolymer. Because of their relative high glass transition temperature compared with the desired service temperature, the A domains build up the hard phase giving the thermoplastic elastomer its strength. These hard domains represent the cross-links of the thermally reversible network. They create a barrier to shear flow just as covalent links do. The elastomer blocks B (i.e., the ethylene/butylene (EB) blocks in SEBS) give the network flexibility. A mechanical model for a thermoreversible network is given in Fig. 2b. The elastomer phase is represented by springs, and the hard phase by solid spheres.

The experimental results presented in [31–34] show a complex influence of SEBS on both the binary blend iPP/SEBS and on the ternary blend containing PE as an additional component. The role of SEBS in the ternary system is concentration dependent. The addition of small amounts of SEBS increases the adhesion between the matrix and the PE component. SEBS is found to be located at the iPP/PE interface and connects the PE phase through local physical cross-links to the PP matrix. At higher concentrations, SEBS clusters in separate domains in the matrix and mixes to some extent in an interlamellar way in the matrix. In this way the two concepts, building up a thermoreversible network and localization of SEBS at the interface, has been confirmed by experimental results.

The investigation of the binary blends revealed a concentration-dependent degree of dispersion of the SEBS domains in either separate clusters and/or an interlamellar mixing with the iPP matrix. The improvement in the mechanical properties could be explained here by increasing the matrix stiffness through local physical cross-links and through classical effects as seen in elastomer-modified thermoplastics, e.g., through the elastomeric character of the EB blocks of the SEBS clustered in the matrix (see Fig. 2).

Having revealed a concept for the role of SEBS in polyolefin blends, the question arises whether the

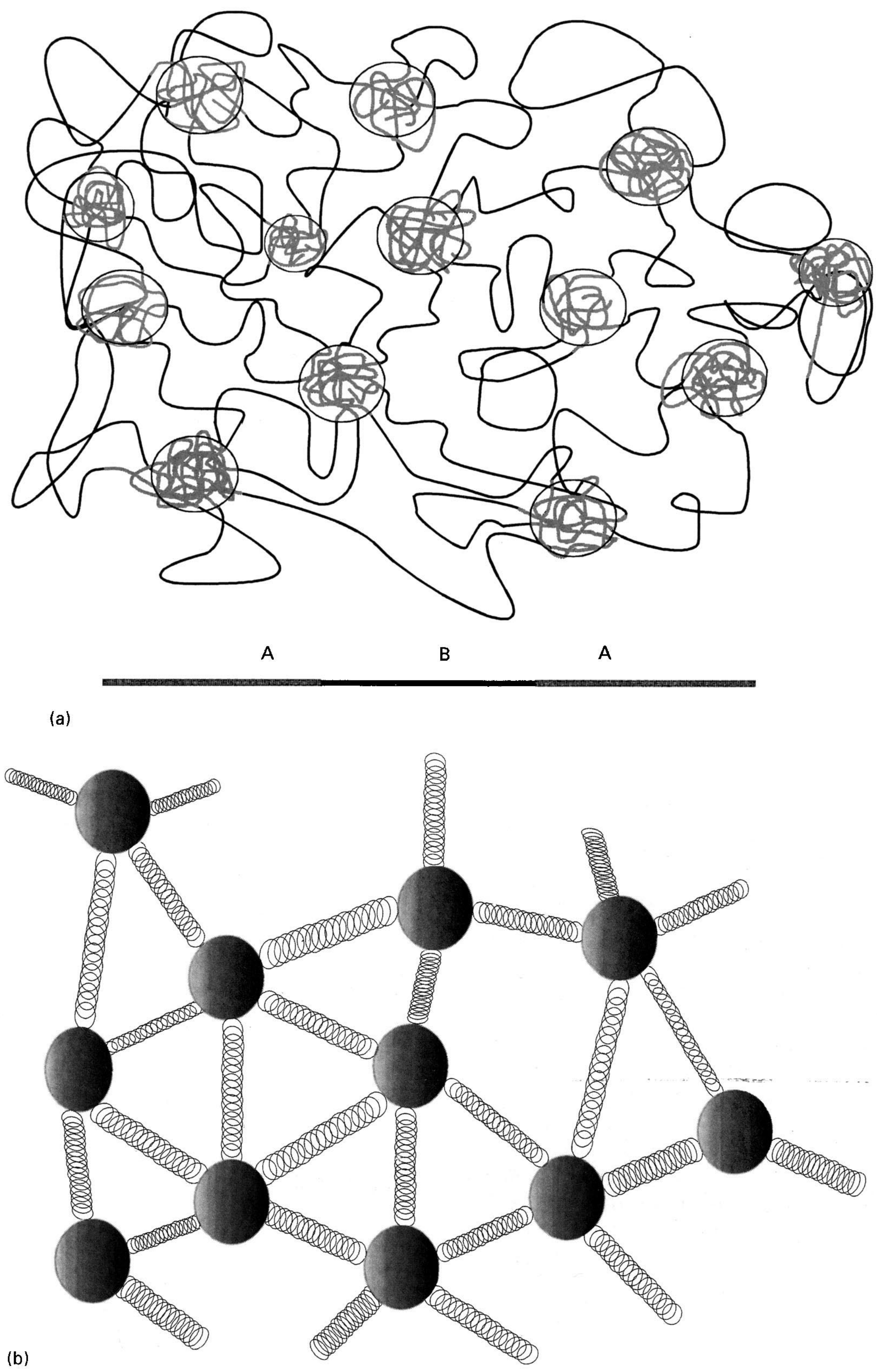

Figure 2 Model of a physical network: (a) ABA copolymer, where A represents the hard and B the soft domains; (b) mechanical concept based on springs (elastomer phase) and solid spheres (hard phase).

interactions at the interface are separately detectable and thus provable. Different interactions of SEBS with iPP or HDPE can lead to different mechanisms at the interphase, e.g., under load or during crystallization. For this reason we investigated the influence of SEBS on model interfaces in iPP/SEBS/iPP and HDPE/SEBS/HDPE blends.

2. Experimental procedure

2.1. Materials and sample preparation

HDPE, iPP and two types of SEBS co-polymer, one functionalized with maleic anhydride ($\text{SEBS}_{\text{func}}$), and the other unmodified ($\text{SEBS}_{\text{non-func}}$), were used to form binary and ternary blends. SEBS is a triblock copolymer, composed of (hard) styrene blocks with a glass temperature, $T_g \approx 370$ K, and (soft) EB blocks with $T_g \approx 220$ K. The (styrene–ethylene) to butylene ratio is about 30 to 70; the ratio of ethylene to butylene sequences on the EB block amounts to about 0.52 [23, 24]. The number-averaged molecular weight of the styrene block is $M_n = 7000$. The materials used were all commercial grade with typical additives. No extraction or cleaning was done; the polymers were used as received. Their characterization data are listed in Table I.

Two-layer samples of type A/A (A = iPP or HDPE, respectively) and three-layer samples of type A–X/A (X = $\mathrm{SEBS}_{\mathrm{func}}$ or $\mathrm{SEBS}_{\mathrm{non\text{-}func}}$, respectively) were prepared as follows: first, homopolymer sheets were prepared by compression moulding. Small amounts of the homopolymer were put between aluminium foils and were melt pressed in a computer-controlled hydraulic press at 465 K (iPP) and 435 K (HDPE) for 3 min without pressure and then another 5 min applying a pressure force of 50 kN. The sheets were cooled down under pressure with a cooling rate of 5 K min^{-1} to 390 K (iPP) and 380 K (HDPE). Then the samples were cooled rapidly to room temperature by quenching them in a water bath with a temperature of 280 K. One set of samples (sheets) were modified on one side with a thin homogeneous film of $\mathrm{SEBS}_{\mathrm{func}}$ and $\mathrm{SEBS}_{\mathrm{non\text{-}func}}$ respectively. For this purpose, a small quantity of SEBS was dissolved in xylene at $T =$ 310 K and poured onto the surface of the sheet. Care was taken that no air inclusions or any visible contamination occurred. The sheets were placed in an oven for 3 days at a temperature of 310 K to evaporate the solvent.

Model interfaces were prepared using two sheets, one modified with SEBS (A–X), and the other unmodified (A). The two sheets were laminated together in a laboratory press at a laminate temperature, T_{press} (T_{press}(iPP) = 420 K; T_{press}(HDPE) = 395 K), a laminate pressure, p_{press}(= 50 kN), and a laminate time, t_{press} (= 180 min). T_{press} was chosen so that it was below the melting temperature of iPP or HDPE (slightly below the onset temperature of the melting peak [31]). This was controlled by differential thermal analysis measurements. By doing this, we achieved a good adhesion-type interaction at the interface. It is known that, for temperatures T_{press} above the melting point, interdiffusion occurs as well as crystallization at the forming interface during cooling [37, 38] which makes the interpretation of the failing mechanisms more difficult. Menning [39] showed that choosing T_{press} too close to the melting temperature leads to an extreme behaviour of the interface; depending on the degree of self-diffusion of the components, the interface will show a behaviour between pure adhesion (no entanglement at the interface) and pure cohesion (migration of homopolymer chains and forming of an interphase). Thus, by choosing the laminating temperature, T_{press}, sufficiently below the melting temperature, we make sure that there is predominantly adhesive interaction at the interface. The other process parameters were chosen on the basis of an investigation on the preparation technique for model interfaces presented in [40].

We prepared the following model interface systems: iPP/iPP, iPP–$\mathrm{SEBS}_{\mathrm{func}}$/iPP, iPP–$\mathrm{SEBS}_{\mathrm{non\text{-}func}}$/iPP, HDPE/HDPE, HDPE-$\mathrm{SEBS}_{\mathrm{func}}$/HDPE, HDPE-$\mathrm{SEBS}_{\mathrm{non\text{-}func}}$/SEBS (the solidus (/) designates the interface due to compression moulding and A–X is the poured composite). Additionally, layer samples from unmodified sheets were prepared to investigate neat surfaces without peeling effects, revealing the morphology of iPP and HDPE as far as can be resolved by light microscopy.

2.2. Measurements

2.2.1. Mechanical investigations

90° peel tests were performed according to DIN 53282 [41]. Strips with lengths, l_{absolute}, of 150 mm and widths, b of 12 mm were cut from the samples for every composite. Three samples were measured for each test. The active peeling length, l, amounted to 100 mm. The samples were peeled in uniaxial direction in a Zwick tensile tester with a speed of 10 mm min^{-1}. The peeling resistance, p_r, was determined using the equation [41]

$$p_r = F_{\mathrm{av}} b^{-1}$$

where F_{av}, the average peel force, was determined graphically. The average peel energy per unit area relative to the peeled contact area was determined using the following equation:

$$E_{\mathrm{av}} = \frac{F_{\mathrm{av}} l}{b l_0} \; (\mathrm{J\,m^{-2}})$$

TABLE I Characterization data of the investigated polymers: ρ, density; T_m, melting temperature; T_g, glass transition temperature; M_n, number-averaged molecular weight; M_w, weight-averaged molecular weight; MFI; melt flow index

Polymer	ρ (g cm^{-3})	T_m (K)	T_g (K)	M_n	M_w/M_n	MFI[a]
iPP	0.901	439	271	57 500	5.14	6.7[b]
HDPE	0.959	411	152	—	—	7.7[c]
$\mathrm{SEBS}_{\mathrm{func}}$[d]	0.899	—	220	52 600	1.15	15[e]
$\mathrm{SEBS}_{\mathrm{non\text{-}func}}$[f]	0.91	—	220	63 200	1.03	17[e]

[a] In grams per 10 min.

[b] Measured at 503 K; a mass of 2.16 kg was used.

[c] Measured at 463 K; mass of 5 kg was used.

[d] Functionalized with maleic anhydride (less than 1%).

[e] Measured at 473 K; a mass of 5 kg was used.

[f] Not functionalized.

2.2.2. Light microscopy

Light microscopy experiments were carried out using a Leitz Metallux II optical microscope. The microscope could be used in polarization or phase contrast mode. Investigations of the peeled surfaces were carried out using the test specimens of the mechanical experiments. From each sample, small pieces were cut out such that both inner sides of the asymmetric interface A–X/A could be investigated, and care was taken that opposite areas of the peeled surfaces (labelled sides 1 and 2) were observed to investigate the asymmetry of the interfaces.

Films of neat iPP and HDPE were also investigated for reference of an undistributed topology.

3. Results and discussion

As expected, the results show that the sample is asymmetric on both sides of the SEBS film, reflecting the special preparation technique of the laminates. The results of the mechanical tests (T-peel strengths) are displayed in Fig. 3. The samples iPP/iPP and HDPE/HDPE show a rather weak peel strength of about 5×10^{-3} N m^{-1}, revealing weak adhesion at the interface. This is due the inert and unpolar surface and the weak interaction possibilities of polyolefins.

On adding an SEBS film, the peel strength increases by two orders of magnitude; while the iPP–SEBS/iPP samples reveal peel strengths of about 10^{-1} N mm^{-1}, the values for the HDPE–SEBS/HDPE system appears to be up to five times higher. Here, the influence of the functionality of SEBS can be seen; while the peel strength for HDPE–SEBS$_{\text{non-func}}$/HDPE is about 2.6×10^{-1} N mm^{-1}, the HDPE–SEBS$_{\text{func}}$/HDPE sample shows a value of 5×10^{-1} N mm^{-1}.

Presenting the results of the mechanical investigation as the average peel energy per unit area, relative to the peeled contact, the values were in close agreement with tests performed by Keitz *et al.* [42]. They measured the average shear stress at failure for three layer sandwich laminates A/B/A in the simple lap shear method. The average shear stress for the HDPE/SEBS composite was 1.6 that for the iPP/SEBS composite. Our results show the same tendency; the average peel energy per unit area for the model interface HDPE–SEBS/HDPE is at least twice the values for the iPP–SEBS/iPP systems. The difference is greater for the model interfaces, modified with functionalized SEBS.

Light microscopy investigations of the unpeeled iPP and HDPE sheets reveal a typical spherulite structure. No other morphologies could be detected. The same holds for the peeled inner surfaces of the iPP/iPP and HDPE/HDPE laminates (see Fig. 4a and b) for HDPE and iPP, respectively. No differences due to peeling were found. No differences of the surface topology compared with the unpeeled samples were found.

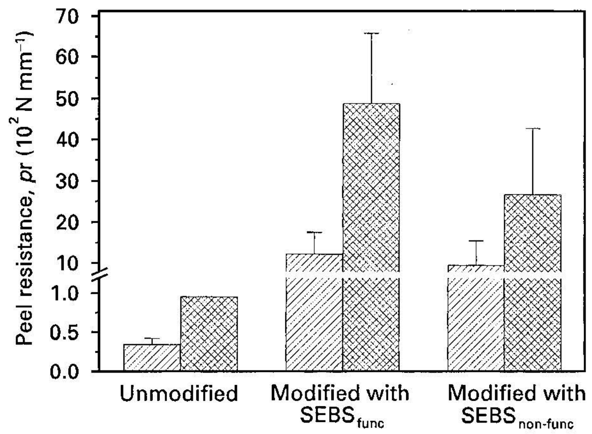

Figure 3 Peel resistance of the model interfaces (after DIN 53282 [37]). (▨), iPP; (▩), HDPE.

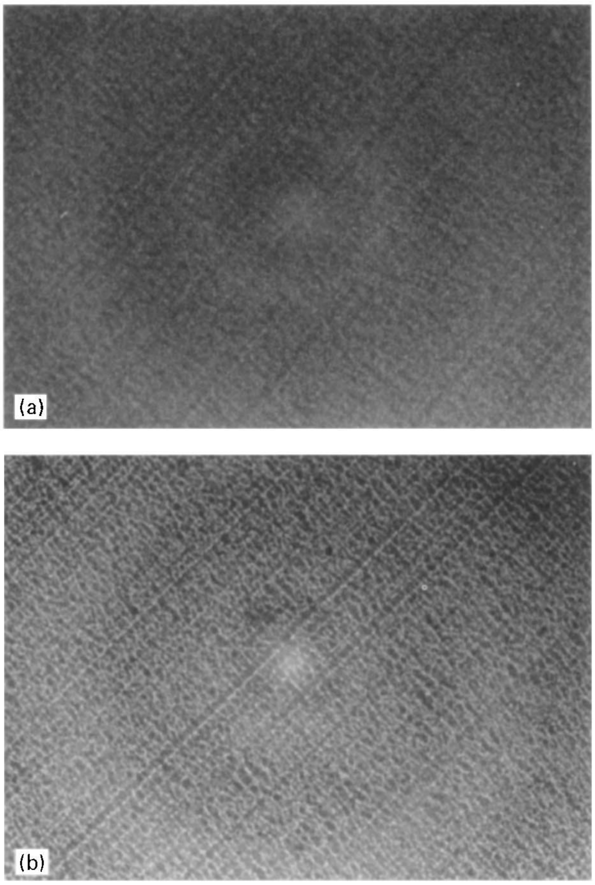

Figure 4 Morphology of the reference interfaces (phase contrast): (a) HDPE; (b) iPP. (Magnification, 652 ×.)

Figs 5 and 6 show the peeled surfaces of the HDPE–SEBS/HDPE and iPP–SEBS/iPP samples. Here, a clear difference of the surface morphologies are found. Fig. 5 displays the surface morphology of HDPE laminated with SEBS$_{\text{func}}$. Both peeled surfaces show, despite the asymmetric preparation technique, the same topology. A ruptured SEBS film is seen, and no deterioration of the underlying HDPE structure is noticed. No differences between the two sides of the SEBS film (poured side and pressed side) is detectable, revealing a uniform failure in the interface. However, the samples containing SEBS$_{\text{non-func}}$ reveal a different texture on both sides of the film (see Fig. 6). The pressed side reveals a morphology similar to that of the neat HDPE sample (Fig. 6a), except for some sporadic appearance of SEBS spots. The peeled surface of the other side (poured side) shows a nearly homogeneous SEBS film with small ruptures in the surface. Both sides are thus clearly distinguishable.

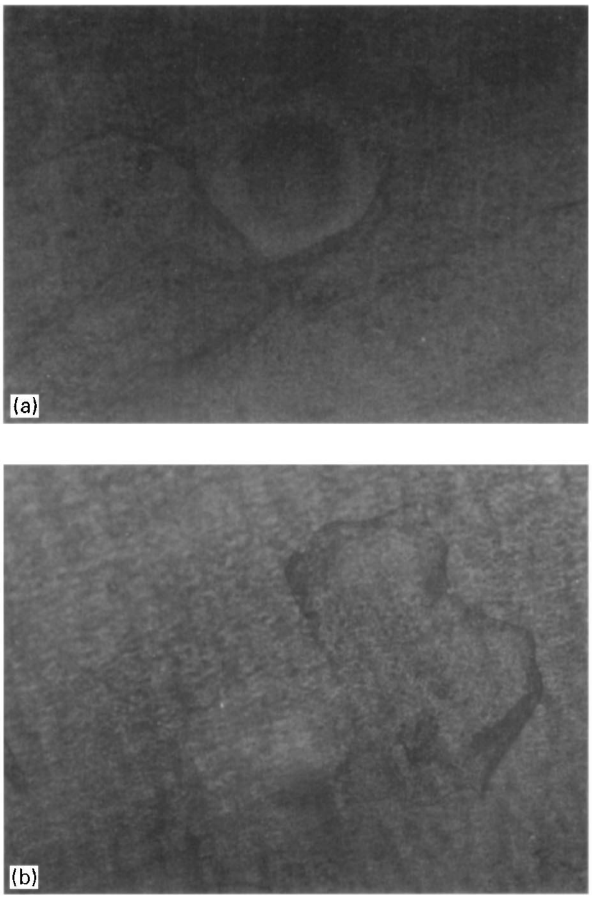

Figure 5 Morphology of the system HDPE–SEBS$_{\mathrm{func}}$/HDPE (phase contrast): (a) side 1; (b) side 2. (Magnification, 129 ×.)

The morphology of iPP–SEBS/iPP differs from that of the HDPE samples. Figs 7 and 8 show the surfaces of the peeled samples. Fig. 7 reveals microcracks in the iPP sheets perpendicular to the peeling direction. These cracks or breaks have the same size and concentration on both sides of the SEBS film. An asymmetry of the surface morphology, which could be due to the preparation technique, is not visible. Only some detachment can occasionally be seen. For the sample containing SEBS$_{\mathrm{non\text{-}func}}$ (Fig. 8), one can distinguish between the two sides. The surfaces also show microcracks. However, on the poured side, the cracks appear in the SEBS film while, on the other surface (the pressed side), they are located in the iPP sheet. The SEBS film is nearly undamaged on the poured side. Some (negligible) detachment occurs; only film buckling is seen.

The mechanical results on the HDPE–SEBS/HDPE system indicate good interaction between PE and SEBS. This interaction is increased by the functionality of the SEBS. The laminates containing a SEBS$_{\mathrm{func}}$ film show "homogeneous" failure in the interface (see Fig. 5); the crack runs entirely in the SEBS film. This is a well-known phenomenon and indicates that the adhesion between PE and SEBS can be described, at least on the poured side of the SEBS film, by a model of mechanical interlocking. In this case, the preparation of the samples seems to have an influence; the SEBS solution, having a lower viscosity than the melt, can flow into the grooves imposed on the HDPE surface by the aluminium foils during compression of the film.

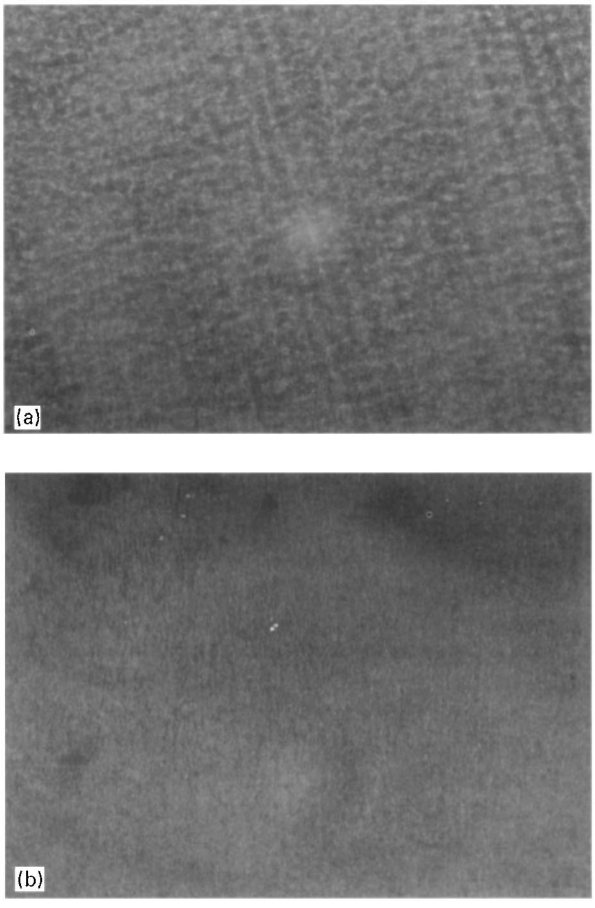

Figure 6 Morphology of the system HDPE–SEBS$_{\mathrm{non\text{-}func}}$/HDPE (phase contrast): (a) side 1; (b) side 2. (Magnification, 129 ×.)

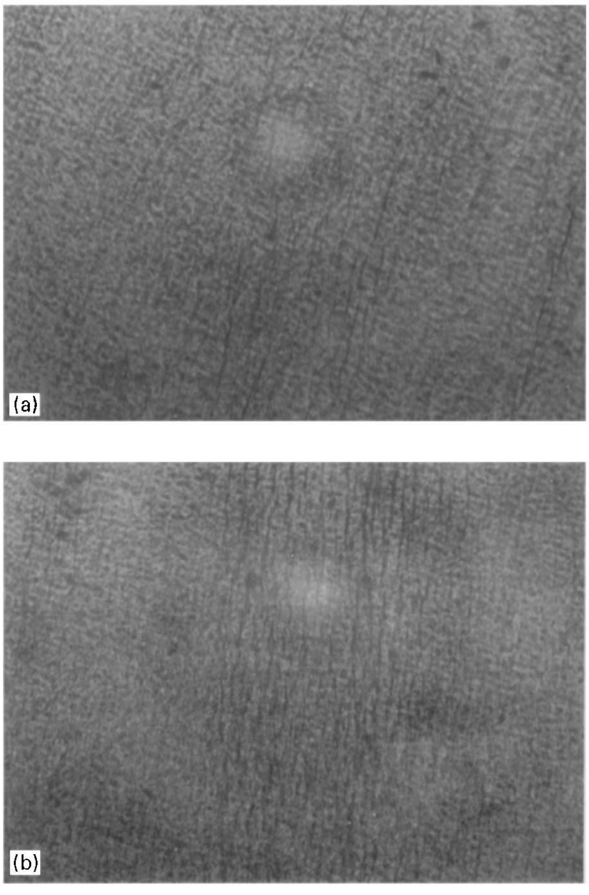

Figure 7 Morphology of the system iPP–SEBS$_{\mathrm{func}}$/iPP (phase contrast): (a) side 1; (b) side 2. (Magnification, 652 ×.)

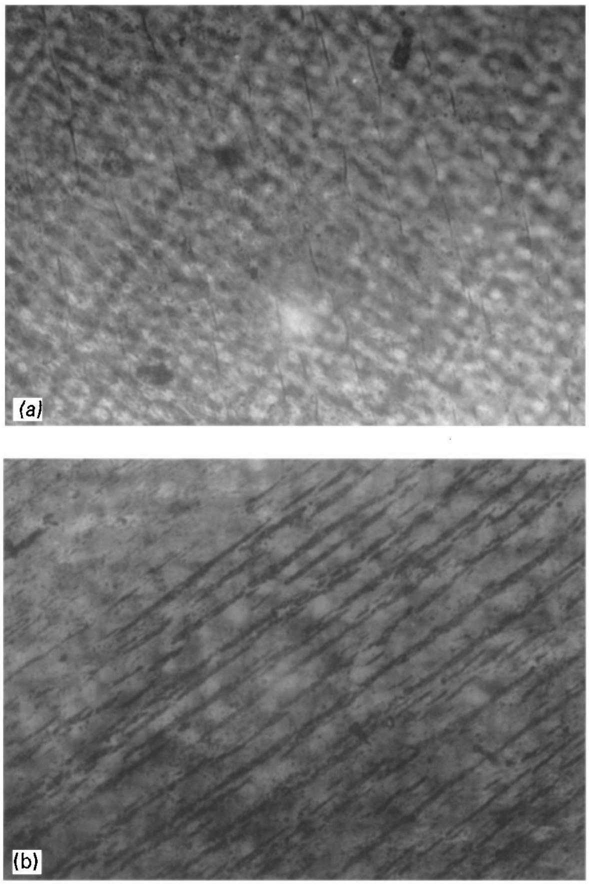

Figure 8 Morphology of the system iPP–SEBS$_{\text{non-func}}$/iPP (phase contrast): (a) side 1; (b) side 2. (Magnification, 129 × .)

In the case of the SEBS$_{\text{non-func}}$, although the peel strength increases also, no homogeneous failure is found, and one can distinguish between the two sides (poured side and pressed side) of the peeled sample. This indicates that the adhesion between PE and SEBS is increased, but not to values which would be typical for mechanical interlocking. The choice of the laminate temperature, T_{press}, below the melting point of PE prevents gross interdiffusion of SEBS and PE at the interface, thus preventing macroscopic entanglement. Therefore, also for this reason the increasing interfacial strength should be due to adhesion. We believe that interaction between the EB blocks of the SEBS and PE sequences is responsible for the improvement in the peel strength.

The interfacial strength at the model interface iPP/iPP is also greatly increased by modifying the interface with SEBS. The increase in peeling resistance is nearly the same for both SEBS types (see Fig. 4). This shows that the interaction between SEBS and iPP is nearly not affected by functionalization of the SEBS as it is in the HDPE systems. In SEBS there is no sequence that would match iPP, except that the butylene monomer unit in the EB block of SEBS shows some local compatibility with the $-(CH_2-CHCH_3)-$ sequence of iPP. Hagenbeck [43] and Petermann and Gohil [44] reported good interaction between the components iPP and poly(1-butene) because the configurations of the molecules of both polymers are rather similar. Gergen [45] reported that the butylene monomer unit is incorporated into the ethylene sequences of the EB block to prevent crystallization of the ethylene sequences without shifting the T_g of SEBS to higher temperatures. We assume that there are fewer butylene sequences for the interaction with iPP than ethylene sequences for the interaction with HDPE in the EB blocks of SEBS. This would readily explain the different dependences of the interfacial strength on the functionalization of SEBS.

HDPE and iPP reveal different morphologies under the light microscope. On HDPE peeled surfaces, no deterioration of the underlying HDPE was detectable. In contrast, the iPP contacting the SEBS interface reveals microcracks perpendicular to the peeling direction. As we have seen above, the failure mechanism of the SEBS film shows buckling and rippling, but no homogeneous rupture in the interface region as it does in the HDPE system. The appearance of the microcracks in the iPP may lead to the conclusion that the SEBS transfers the stress away from the interface in the bulk iPP until the stress due to peeling is greater than the adhesive forces between SEBS and iPP. This is in agreement with the possibility that SEBS builds up a thermally reversible network and with different failure mechanisms of the SEBS film at the interfaces. These mechanisms are not influenced by the functionality of the SEBS, except that there is more homogeneity in the results in the case of SEBS$_{\text{func}}$. Also the intrinsic asymmetry of the interfaces due to the preparation techniques is of no influence.

From these results, conclusions concerning bulk blends of iPP, PE and SEBS can be drawn.

4. Conclusions

1. SEBS shows a different interaction with HDPE and iPP. The EB sequences in the copolymer can interact better with HDPE than with iPP. Butylene sequences interact with iPP. The effect, however, is lower since the number of butylene sequences is lower than the number of ethylene sequences.

2. The compatibility of the butylene sequences of the EB block with iPP explains the good dispersion found in bulk binary blends of iPP with SEBS. The homogeneous and fine dispersion of SEBS clusters in the iPP matrix and the building of a physical network explains the good mechanical behaviour of this system.

3. The existence of a thermally reversible network is proved by a number of morphological characteristics of the peeled surfaces of both systems, although different stress transfer processes for both systems are observed. While in the system HDPE–SEBS/HDPE the stress builds up in the SEBS film, causing the crack to run homogeneously through the film, in iPP–SEBS/iPP samples the stress is transferred away from the interphase into the iPP matrix causing cracking perpendicular to the peeling direction.

The high peel strength of the system HDPE–SEBS/HDPE containing functionalized SEBS reveals an excellent interaction between PE and the functionalized EB blocks of the SEBS. The breaking of the samples reveals a pure adhesive mechanism at the interface. The peel strength is controlled entirely by the mechanical strength of the SEBS film.

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References

1. D. R. PAUL and S. NEWMAN, "Polymer blends", Vol. 1 (Academic Press, New York, 1978).
2. *Idem.*, "Polymer blends", Vol. 2 (Academic Press, New York 1978).
3. L. A. UTRACKI, "Polymer alloys and blends: thermodynamics and rheology" (Hanser, New York, 1990).
4. L. A. UTRACKI and R. A. WEISS, "Multiphase polymers: blends and ionomer", ACS Symposium Series 395 (American Chemical Society, Washington, DC, 1989).
5. J. KARGER-KOCSIS, "Polypropylene: structure, blends and composites", Vol. 2 (Chapman & Hall, London, 1995).
6. A. LOVINGER and M. WILIAMS, *J. Appl. Polym. Sci.* **25** (1980) 1703.
7. D. W. BARTLETT, J. W. BARLOW and D. R. PAUL, *ibid.* **27** (1982) 351.
8. O. F. NOEL and J. F. CARLEY, *Polym. Engng Sci.* **15** (1975) 117.
9. J. W. TEH, *J. Appl. Polym. Sci.* **28** (1983) 605.
10. L. A. UTRACKI, in "Multiphase polymers: blends and ionomers", ACS Symposium Series 395, edited by A. Utracki and R. A. Weiss (American Chemical Society, Washington, DC, 1989) Ch. 7.
11. R. FAYT, R. JEROME and P. TEYSSIE, *J. Polym. Sci., Polym. Phys. Edn* **27** (1989) 775.
12. *Idem., J. Polym. Sci., Polym. Lett. Edn* **19** (1981) 79.
13. A. P. PLOCHOCKI, S. S. DAGLI and R. D. ANDREWS, *Polym. Engng Sci.* **30** (1990) 741.
14. D. R. PAUL, in "Polymer blends", Vol. 2, edited by D. R. Paul and S. Newman (Academic Press, New York, 1978) Ch. 12.
15. M. MAGLIO and R. PALUMBO, in Proceedings of the Second Polish–Italian Joint Seminar on Multicomponent Polymeric Systems, Lodz, September, 1982, edited by M. Kryszewski, A. Galeski and E. Martuscelli (Plenum, New York, 1984).
16. R. FAYT, R. JEROME and P. TEYSSIE, *J. Polym. Sci., Polym. Phys. Edn* **19** (1981) 1269.
17. *Idem., ibid.* **20** (1982) 2209.
18. *Idem., Makromol. Chem.* **187** (1986) 837.
19. G. H. MICHLER, "Kunststoff-Mikromechanik: Morphologie, Deformations- und Bruchmechanismen" (Carl Hanser, München, 1992).
20. J. J. ELMENDORP, PhD thesis, TH Delft (1986).
21. V. FLARIS, PhD thesis, Melbourne University, Melbourne (1993).
22. A. K. GUPTA and S. N. PURWAR, *J. Appl. Polym. Sci.* **30** (1985) 1799.
23. K. R. SRINIVASAN and A. K. GUPTA, *ibid.* **53** (1994) 1.
24. M. C. SCHWARZ, J. W. BARLOW and D. R. PAUL, *ibid.* **37** (1989) 403.
25. D. W. BARTLETT, D. R. PAUL and J. W. BARLOW, *Mod. Plastics* **58** (1981) 60.
26. V. FLARIS, M. D. ZIPPER, G. P. SIMON and A. J. HILL, *Polym. Engng Sci.* **39** (1993) 125.
27. D. R. PAUL, in "Thermoplastic elastomers: a comprehensive review", edited by N. R. Legge, G. Holden and H. E. Schroeder (Hanser, New York, 1987) Ch. 12/6.
28. ANON., Shell Chemical Company, Houston, TX, Technical Bulletin SC: 165–177 (1977).
29. A. L. BULL and G. HOLDEN, *J. Elastomers Plastics* **9** (1977) 281.
30. T. D. TRAUGOTT, J. W. BARLOW and D. R. PAUL, *J. Appl. Polym. Sci.* **28** (1983) 2947.
31. U. PLAWKY, M. SCHLABS and W. WENIG, *ibid.* **59** (1996) 1891.
32. U. PLAWKY and W. WENIG, in Proceedings of the 13th IUPAC Conference on Chemical Thermodynamics, Clermont-Ferrand, 17–22 July (Soc. Chim. Français, 1994) p. 266.
33. *Idem.*, in Proceedings of the EPS Conference on Macromolecular Physics, Vol. 18C, Eindhoven, 4–7 July (1994).
34. *Idem., Macromol. Symp.* **102** (1990) 183.
35. ANON., Shell Company, London, Technical Report (1986).
36. W. P. GERGEN, R. G. LUTZ and S. DAVISON, in "Thermoplastic elastomers: a comprehensive review, edited by N. R. Legge, G. Holden and H. E. Schroeder (Hanser, New York, 1987) Ch. 14.
37. R. P. WOOL, in "Polypropylene", Vol. 1, "Structure and morphology", edited by J. Karger-Kocsis (Chapman & Hall, London, 1995) Ch. 8.
38. B.-L. YUAN and R. P. WOOL, *Polym. Engng Sci.* **30** (1990) 1454.
39. G. MENNING, in "Polypropylene", Vol. 1, "Structure and morphology", edited by J. Karger-Kocsis (Chapman & Hall, London, 1995) Ch. 7
40. G. LIN, W. WENIG and J. PETERMANN, *J. Mater Sci.* (1996) submitted.
41. ANON., "Winkelschälversuch" DIN 53282 (Beuth, Berlin, 1979).
42. J. D. KEITZ, J. W. BARLOW and D. R. PAUL, *J. Appl. Polym. Sci.* **29** (1984) 3131.
43. G. HAGENBECK, Diploma thesis, University of Duisburg, Duisburg (1982).
44. J. PETERMANN and R. M. GOHIL, *J. Mater. Sci.* **14** (1979) 2260.
45. W. P. GERGEN, "Uniqueness of hydrogenated block copolymers for elastomer application" (Shell Development Company, Houston, TX, 1989).

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