Influence of Thermoplastic Elastomers on Adhesion in Polyethylene–Wood Flour Composites

KRISTIINA OKSMAN, HENRIK LINDBERG

Luleå University of Technology, Department of Wood Technology, Skeria 3, S-93187 Skellefteå, Sweden

ABSTRACT: The mechanical properties of recycled low-density polyethylene/wood flour (LDPE/WF) composites are improved when a maleated triblock copolymer styrene–ethylene/butylene–styrene (SEBS–MA) is added as a compatibilizer. The composites' tensile strength reached a maximum level with 4 wt % SEBS–MA content. The compatibilizer had a positive effect on the impact strength and elongation at break but decreased the composites' stiffness. Dynamic mechanical thermal analysis (DMTA), a lap shear adhesion test, and a scanning electron microscope (SEM) were used to investigate the nature of the interfacial adhesion between the WF/SEBS and between the WF/SEBS–MA. Tan δ peak temperatures for the various combinations showed interaction between the ethylene/butylene (EB) part of the copolymer and the wood flour in the maleated system. The shear lap test showed that adhesion between the wood and SEBS–MA is better than between the wood and SEBS. The electron microscopy study of the fracture surfaces confirmed good adhesion between the wood particles and the LDPE/SEBS–MA matrix. © 1998 John Wiley & Sons, Inc. J Appl Polym Sci 68: 1845–1855, 1998

Key words: wood flour/thermoplastics composites; copolymer compatibilizer; molecular interaction; dynamic mechanical thermal properties; morphology

INTRODUCTION

The interfacial adhesion between cellulose-based reinforcements or fillers and thermoplastics has been the focus of a large amount of research during the last 10 years. The nominal adhesion between wood and olefin polymers is poor due to large differences in surface energies. To bridge this energy gap, different types of coupling agents or compatibilizers have been tested.[1–8]

Several investigations have shown that polypropylene modified with maleic anhydride (MAPP) acts as a compatibilizer in cellulose fiber/polyolefin composites, improving the composites' mechanical properties.[1–4] Composites using MAPP show interfacial interaction between the cellulose fibers and the olefin matrix as revealed by electron microscopy.[1–3] Oksman and Clemons[7,8] showed that a styrene–ethylene/butylene–styrene–MA (SEBS–MA) triblock copolymer has a compatibilizing effect when used with wood flour (WF) and polyethylene (PE) and polypropylene (PP) systems. The composites' mechanical properties such as tensile strength and impact strength were improved with small amounts of SEBS–MA. An electron microscopy study confirmed the improved adhesion in the filler/matrix interfaces.

SEBS–MA triblock copolymers have a bridging effect between different fillers and the olefin matrix. Several authors have shown that different elastomers are capable of forming an interphase around a filler particle and therefore improve the interfacial adhesion between phases.[8–13]

SEBS is a hydrogenated form of a styrene/butadiene/styrene (SBS) block copolymer. The na-

ture of the EB part of the block copolymer is equivalent to a random copolymer of ethylene and butylene. Maleic anhydride (MA) can be grafted to the unsaturated part of the EB chain to improve the physical and chemical properties of the polymer, by providing polarity to promote hydrophilicity and improve adhesion and compatibility with other polymers and fillers.[14,15]

Earlier work has shown that the MA part of the MAPP compatibilizer reacts with the hydroxyl groups on the cellulose fiber surface, resulting in covalent bonds (esterification)[16,17] and hydrogen bonds.[17] Other studies have shown that styrene may interact with the wood.[18-20] This positive interaction is believed to come from a possible acid–base interaction between wood and styrene. Styrene is known to have a basic character[21,22] and it is possible that wood surfaces are slightly acidic. However, cellulose fibers have been found to results in an acid–base interaction with styrene.[21] Two different mechanisms can therefore be expected to occur when using SEBS–MA in a PE/WF composite system: (a) The MA at the EB block is expected to form chemical and hydrogen bonds with the hydroxyl groups on the wood surface, resulting in improved interaction between the olefin matrix and the wood surfaces. (b) The styrene endblocks attached to the wood surface and the EB blocks blended the olefin matrix.

The objective of this work was to optimize the SEBS–MA compatibilizer content by studying its effect on the mechanical properties of recycled low-density polyethylene (LDPE)/WF composites. Model composites were made to study the molecular interaction between different components in the SEBS/WF and maleated SEBS/WF systems.

EXPERIMENTAL

Materials

The following commercially available materials were used: *LDPE/WF,* Polywood, (Convertere Recycling AB, Billingsfors, Sweden) in the form of compounded pellets with a WF content of 40% by weight and recycled LDPE from milk cans, the pellets' MFI being about 0.1 g/10 min (190°C/2.16 kg); maleated styrene–ethylene/butylene–styrene (*SEBS–MA*), Kraton FG 1901X (Shell Chemical Company, Houston, Texas, USA) MA-functionalized triblock copolymer, polystyrene (PS) content 28% by weight, density 910 kg/m³; and the corresponding styrene–ethylene/butyl-

ene–styrene (*SEBS*), Kraton G 1652 (Shell Chemicals). The molecular weights of the blocks are 7200–64,000–7200.[14]

Processing

The materials for mechanical testing and scanning electron microscopy study were injection-molded in a conventional machine (Arburg 320 M 750-210) into test bars (ASTM D 638) having a cross section of 13 × 4 mm and a length of 217 mm. The melt temperature was 180°C and the injection speed 35 cm³/s at a pressure of 230 bar. The LDPE/WF-compounded pellets and the SEBS–MA compatibilizer were premixed and then injection-molded. The compatibilizer content for mechanical testing was varied between 2 and 10 wt %.

The model composites for dynamic mechanical testing and electron microscopy were mechanically blended in a Brabender mixer at 100 rpm at about 180–190°C. The polymer was introduced into the mixing cavity and the WF then gradually added. Mixing continued until a constant torque was reached, taking about 5 min. Each batch of the blend was at most 30 g. After blending, the materials were pressed in a laboratory press to form 0.5–2-mm-thick plates. The press temperature was about 200°C and the pressure about 10 MPa. Brabender blending was used to study the molecular interaction between the compatibilizer and the wood, and the blends studied had a high compatibilizer content to explore the effect.

Mechanical Properties

The mechanical properties were determinated using an Instron tensile testing device (Instron Corp., series IX automated materials testing system 1.38, Model 4411) with a crosshead speed of 2 mm/min. The Young's modulus of elasticity, maximum strength, and elongation at break were automatically calculated from the stress–strain data. At least five test bars of each composition in both dry and wet conditions were tested. The wet conditioning of the samples was achieved by immersion in water in an oven, at a temperature of around 100°C for 24 h.

Dynamic mechanical thermal properties, polymer damping peaks (tan δ), and the storage modulus (E′) were determined using a dynamic mechanical thermal analyzer (DMTA; Rheometric Scientific Mk III). The typical specimen size was 8 × 6 × 0.5 mm and the data presented in this

work were run at 1 Hz, using the tensile testing mode over a temperature interval of −100 to 120°C and a heating rate of 2°C/min.

Lap Shear Test

Thin SEBS–MA and SEBS films were pressed (1 mm) and cooled to room temperature. Two wood plates of 60×20 mm were sliced and ground and about 400 mm^2 of the polymer films placed between the ends of the plates. This sandwich was then heated (180–190°C) and pressed together for about 5 min and then cooled to room temperature. The specimens were then mounted in the tensile testing machine. The force necessary to separate the two wood plates was determined at a crosshead speed of 1 mm/min. At least nine specimens were measured and the average force normalized by the area of the bonded surface to calculate the lap shear strength (MPa).

Electron Microscopy

Fracture surfaces for the examination of the composites' morphology were obtained from the room-temperature impact and liquid nitrogen temperature test samples. The specimens' microstructures were studied using a JSM 5200 (JEOL) and a CamScan S 4-80DV scanning electron microscope (SEM) at an acceleration voltage of 20 kV. All specimens were sputter-coated with gold.

RESULTS AND DISCUSSION

Mechanical Properties

The interface between the reinforcing/filler particles and the matrix has a great influence on the mechanical properties of a composite. The mechanical properties can therefore give indirect information about interfacial behavior. Small molecules (e.g., water and extractives) usually collect at the high-energy interfaces, which reduces the composite's long-term properties. If there is insufficient adhesion between the filler and matrix, the interface may be susceptible to attack by water, resulting in loss of strength.[23] In previous work,[7] the mechanical properties of LLDPE/WF composites with SEBS and SEBS–MA were tested in both dry and wet conditions. The results of the study showed that maleated SEBS enhanced the interaction between WF and LLDPE, resulting in improved mechanical properties. The tensile

Table I   Summary of Effects of Compatibilizers on the PE/WF Composites

Compatibilizer	σ_{max}	ε	E	Charpy Impact
SEBS	○	−	○	○
SEBS–MA	+	−	○	+

Comparisons were made by using student's t-test, 5% significance level. "+" is positive, "−" is negative, and "○" is no or little effect.

strength of different composites was not affected by the water immersion. A summary of the effects of SEBS and SEBS–MA on the WF-filled LLDPE are shown in Table I.

Table I shows that the addition of SEBS–MA has a positive effect on the composites' tensile and impact strengths while the elongation at break was reduced and the E modulus was unaffected. The addition of SEBS had no positive effects on the composites' mechanical properties.

In this study, the mechanical properties of composites with various SEBS–MA contents were studied. Figure 1 shows the tensile strength properties of the LDPE/WF composites. The SEBS–MA content was varied between 2 and 10 wt % with the strength reaching its maximum level at 4 wt %. Further addition of SEBS–MA did not improve the tensile strength, but, instead, a decrease of tensile strength was observed. It is possible that higher loading of the elastomeric compatibilizer will influence the matrix properties more and more and after, in this case, 4 wt % become a dominating factor for the tensile strength properties. It is known that if elastomers are used as impact modifiers in thermoplastics, the maximum strength is decreased.[23] Earlier work on impact-modified PP showed that the elastomeric phase increased the materials' impact strength and elongation at break but decreased their strength and stiffness.[24,25]

The effect of the SEBS–MA content on elongation at break is shown in Figure 2. The elongation at break increases with increasing compatibilizer content and the maximum level was not reached at 10 wt % addition. It can be seen from Figure 2 that the 4 wt % addition increased the elongation at break by 70%, while the further increase between 4 and 10 wt % was only about 30%. It is possible that the large increase between 0 and 4 wt % is due to interfacial effects, but with increasing elastomer content, soft rubber inclusions in the matrix are expected to dominate.
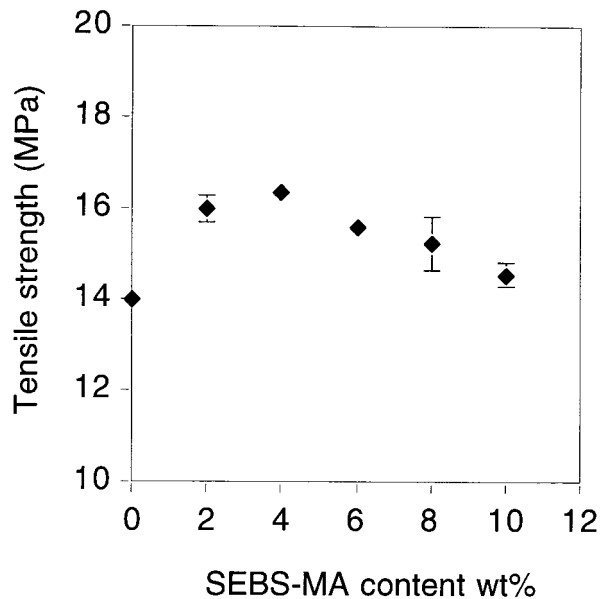
The effect of increased SEBS–MA content on

Figure 1   Maximum tensile strength of the LDPE/ WF composites as a function of the SEBS–MA compatibilizer content.

the composite's stiffness is shown in Figure 3. The tensile modulus of the composites decreased with increasing SEBS–MA content. Earlier studies have shown that a soft elastomeric interphase around the filler can reduce the E modulus of the composites even with low elastomer content.[9–12] In this work, the tensile modulus decreased from about 1.8 GPa to about 1.2 GPa with the addition
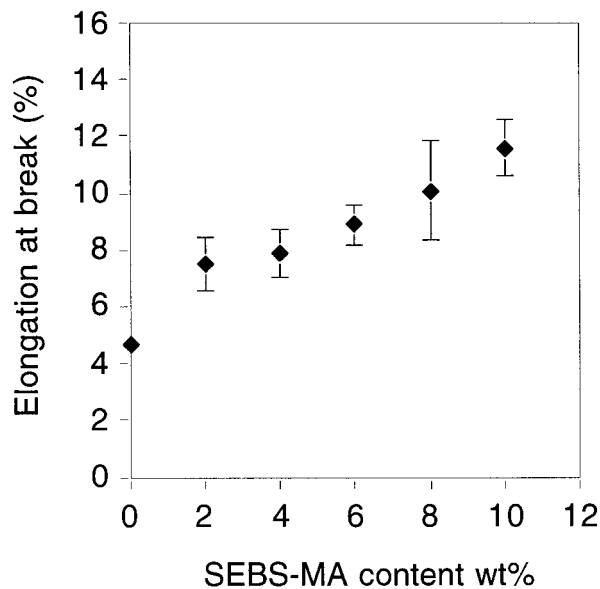
of 2 wt % SEBS–MA (about 33%). The E modulus of elastomer-filled polymers can be calculated theoretically using the inverted Lewis and Nielsen equation.[23] This indicates a reduction of the matrix E modulus of less than 5% with 2 wt % elastomer content. This suggest that SEBS–MA forms an interphase around the WF particles which results in the loss of a reinforcing effect. An elastomeric interphase around the filler particles has been shown to cause a greater reduction in the composite E modulus than in a morphology when the elastomer exists as discrete domains in the matrix.[10,12] The large decrease between 0 and 2 wt % of the compatibilizer confirms this, as the additional E modulus decrease between 2 and 10 wt % is smaller.

The charpy unnotched impact test results can been seen in Figure 4. A 2 wt % SEBS–MA content increased the impact strength from 18.1 to 33.1 kJ/m^2 (82%) and 4 wt % from 18.1 to 35.2 kJ/m^2 (93%), while the total improvement (with 10 wt % SEBS–MA) was 153%. The effect seen with as little as 2 wt % SEBS–MA content is probably due to interfacial effects because this low elastomer content should not affect the matrix properties to this extent. The improved tensile and impact strength suggests interfacial adhesion between the matrix and filler interface, whereby stresses can transfer from the matrix to the wood particle which results in more energy being needed to start crack propagation.

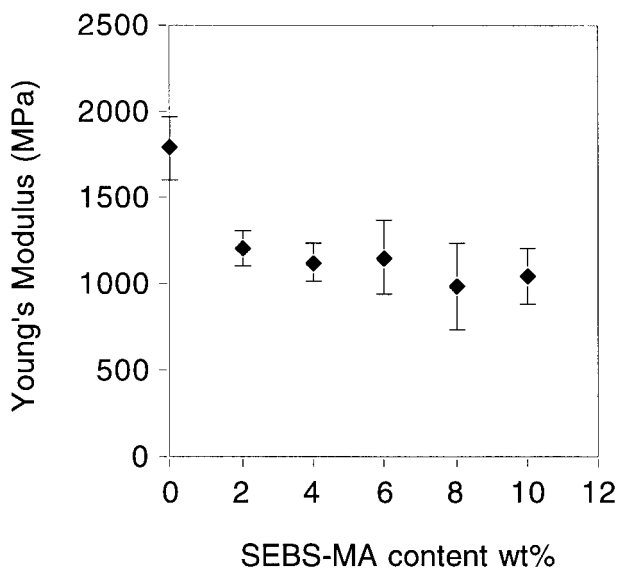The positive effects on the mechanical proper-

Figure 2   Effect of SEBS–MA content on the elongation at break for the LDPE/WF composites.

Figure 3   Effect of SEBS–MA compatibilizer content on the stiffness of the LDPE/WF composites.
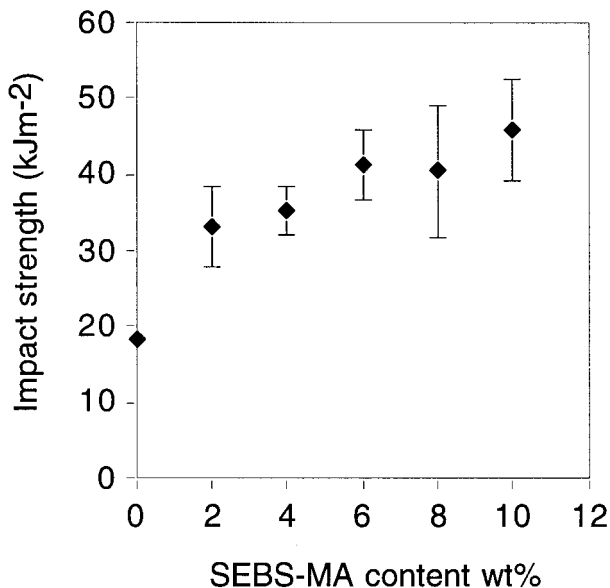
Figure 4 Effect of the SEBS–MA content on the Charpy unnotched impact strength for the LDPE/WF composites.

ties even with small additional percentages of the compatibilizer are interesting. One explanation can be that the interfacial energy gap between the hydrophilic wood and hydrophobic polymer as well as viscosity differences can lead to the thermoplastic elastomer preferring to locate at the wood particle surface instead of being dispersed in the polymer matrix during injection molding.

After immersion in water at about 100°C for 24 h, the tensile strengths of the LDPE/WF composites and composites with 4 wt % of the SEBS–MA compatibilizer were compared. The test results did not show any significant differences between the unmodified composites' maximum tensile strength, while the tensile strength for the composites with SEBS–MA was reduced from 16.3 ($\pm$0.1) to 15.7 ($\pm$0.2) MPa. The reduction in tensile strength indicates that there are few chemical bonds between the WF and the SEBS–MA. It is possible that the injection-molding process (without pre-extrusion) gives too short a time at high temperature for activation of the MA to take place. It has been reported that maleated PP needs 5 min at 180°C to become activated and the amount of ester bonds is larger when MAPP is activated compared to when it is inactivated.[17] The single process step used here is probably not enough to activate the MA in the SEBS.

A lap shear test can be used for comparing interfacial bonding between the wood and compatibilizer.[12,14] The lap shear tests were carried out to obtain information about the adhesion strength between the wood and SEBS and the wood and SEBS–MA. The results showed that the bond strength between maleated SEBS and the wood was 2.1 ($\pm$0.5) MPa compared to the 0.6 ($\pm$0.4) MPa between SEBS and the wood. The adhesion between SEBS–MA and the wood was thus almost four times greater, which suggests stronger bonding between the wood and SEBS–MA surfaces.

Dynamic Mechanical Measurements

Dynamic mechanical measurements can give knowledge about the interaction between molecules in different components in the composites. The position of the primary damping peak (tan δ) of the copolymers used can provide information about the interaction between the polymers and wood at the molecular level. In this system, there are two primary transitions: the EB block glass transition and the styrene block glass transition [see Fig. 5(a)]. SEBS tan δ peak positions are reported in the literature as about -40°C for the EB and about 110°C for PS, measured with the DMTA.[14] The small differences when compared to the SEBS used here may be due to the different material (molecular weight) and test parameters (e.g., frequency and heating rate).

In Figure 5(a), tan δ is shown as a function of temperature. MA, which is grafted on the hydrogenated EB, seems not to change the EB tan δ peak position, but the peak amplitude of both the EB and PS are changed. In fact, the SEBS seems to have lower PS content compared to SEBS–MA even if the manufacturer's data indicate the same PS content for both.

In Figure 5(b), the dynamic storage modulus (E') of the SEBS–MA and SEBS is shown as a function of temperature. The PS acts as a reinforcement in the elastomeric EB matrix for the block copolymer. It can been seen from the figure that the E' of the SEBS is much lower above the EB transition than that of the SEBS–MA. This also confirms that the styrene content is higher in the SEBS–MA copolymer than in SEBS. It is believed that the EB–MA block forms a discontinuous phase, but further experimental work is required to confirm this.

The tan δ peak temperatures for the SEBS–MA and SEBS copolymers containing 0, 20, or 40% by weight of WF are shown in Table II. The primary tan δ peak positions of the soft rubbery EB phase in the maleated SEBS were moved to
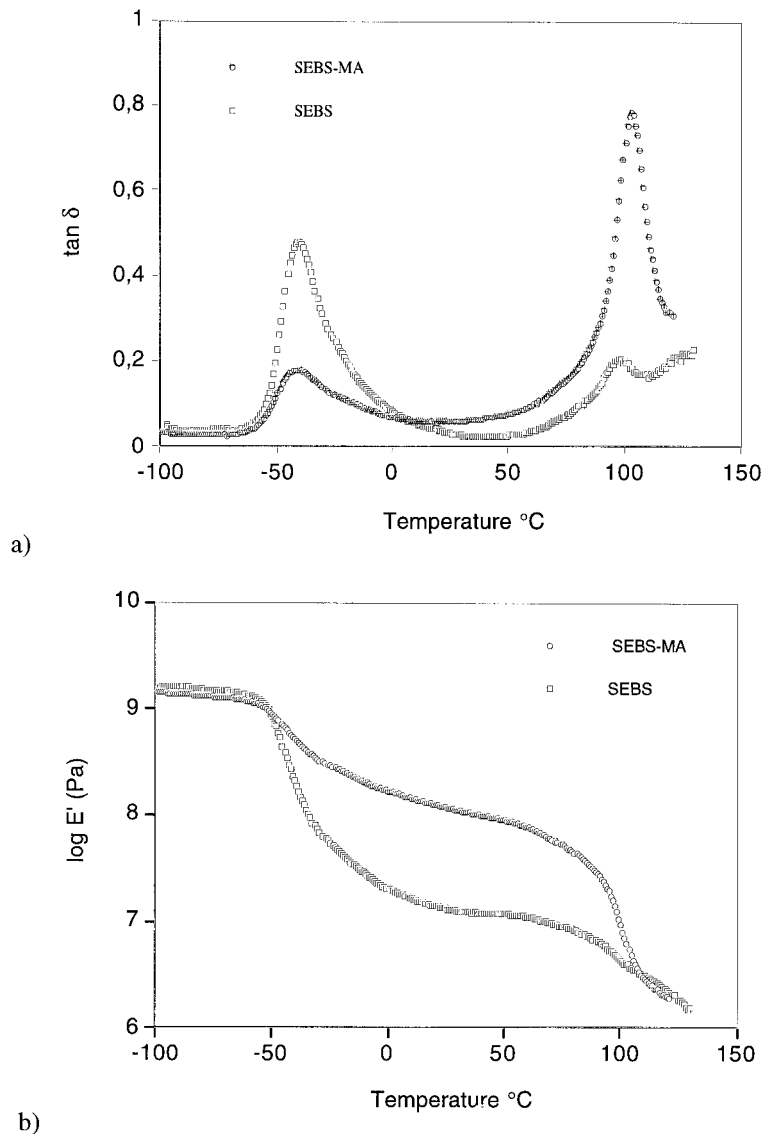
Figure 5   DMTA curves of SEBS–MA and SEBS copolymers: (a) the primary tan δ peaks of the rubbery EB phases and PS phases as a function of temperature; (b) the dynamic storage modulus as a function of temperature.

higher temperature, while the tan δ peak position for EB in SEBS was unaffected when WF was added. The tan δ peak temperatures for the PS phase was relatively unaffected in both SEBS and SEBS–MA systems when WF was added.

The tan δ curves for the pure SEBS–MA and SEBS–MA with various levels of WF content are presented in Figure 6. The tan δ peak of rubbery EB (where the MA is located) is moved to higher temperatures and the damping amplitude is increased with the addition of WF. The tan δ peak shift to higher temperatures means that the polymer probably interacts with the wood filler sur-

face and that the molecular motion of EB is restricted.[18,19,23] The shift in tan δ should be proportional to the surface area of the filler so the effect is expected to increase with increasing WF content. The position of the tan δ peaks for PS are unaffected and the damping amplitude is decreased with increasing WF content.

Figure 7 shows the tan δ curves for SEBS and SEBS with various levels of WF content. The temperature of the rubbery EB tan δ peak is not affected by the addition of WF, which supports the expectation that grafted MA on the EB block interacts with the WF (see Fig. 6).

Table II   Tan δ Peak Temperature of EB and PS with Different WF Content

WF Content (wt %)	SEBS		SEBS–MA	
	EB	PS	EB	PS
0	-40 ± 0	98 ± 1	-42 ± 2	103 ± 0
20	-40 ± 2	100 ± 2	-37 ± 0	102 ± 1
40	-38 ± 1	98 ± 1	-35 ± 2	101 ± 2

Tan δ is a mean of three measurements. ± values are standard deviations.

The peak amplitude of the EB–MA primary transition is increased with the addition of WF. According to Nielsen,[23] the possible reasons for an increasing damping amplitude are (1) particle–particle friction where particles touch each other as in weak agglomerates, (2) particle–polymer friction where there is essentially no adhesion at the interface, and (3) excess damping in the polymer near the interface because of induced thermal stresses or changes in polymer conformation or morphology.

An increase in the amplitude of the glass transition (tan δ peak) indicates that the number of similar molecule segments involved is larger. This is common when the polymer is subjected to tensile forces, which results in an increase in intermolecular distances. The coefficients of thermal expansion between the particle and the surrounding polymer in the melt-formed composites
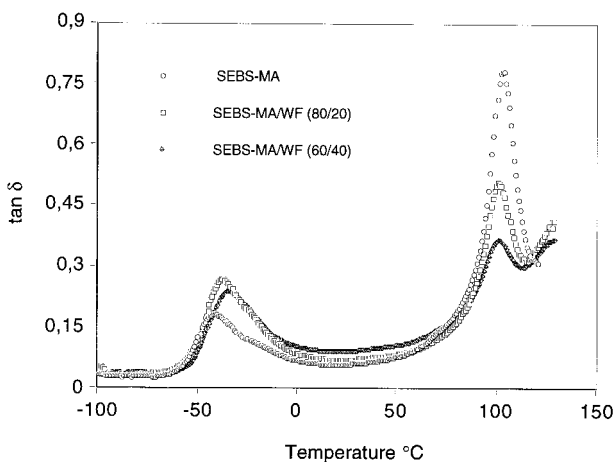
Figure 6   DMTA curves of the SEBS–MA copolymer with different WF content. The rubbery tan δ peaks (EB–MA) and the PS tan δ peaks are presented as a function of the temperature. The WF content was varied: 0, 20, and 40 wt %.
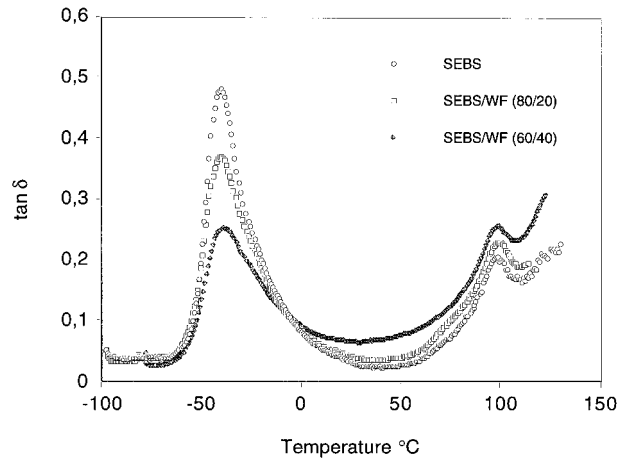
Figure 7   DMTA curves of the SEBS copolymer with different WF content. The rubbery tan δ peaks (EB) and the PS tan δ peaks are presented as a function of the temperature. The WF content was varied: 0, 20, and 40 wt %.

are not the same. Tensile stress increases in the case where the coefficient of thermal expansion is smaller for the filler particle than for the polymer matrix. This would result in an increased number of molecular segments and, therefore, an increase in the tan δ amplitude. On the other hand, if the stress results in an increased free volume, the primary transition peak would shift to lower temperature, which was not observed here.

It is possible that particle–particle friction occurs due to high WF particle content, but, then, the peak amplitude would be expected to increase with increased WF content. The results for the SEBS/WF system (Fig. 7) shows that the tan δ peak of EB does not shift with the addition of WF and that the peak amplitude decreases with increasing WF content. The decrease of the tan δ peak amplitude with WF is expected and the results do not indicate any particle/particle friction in this system.

Another possible reason for the increased tan δ peak amplitude, according to Nielsen,[23] is particle–polymer friction without adhesion. Because the EB chain does not interact with the wood, it is only the MA in the EB (the largest part of EB does not interact) and the PS in the SEBS which are expected to interact.

The increase of the damping peak amplitude can also depend on the WF moisture content. The dynamic mechanical properties of unsaturated polyester and sawdust showed that the tan δ peak amplitude increased with increased moisture content.[26] The WF used here is not expected to con-
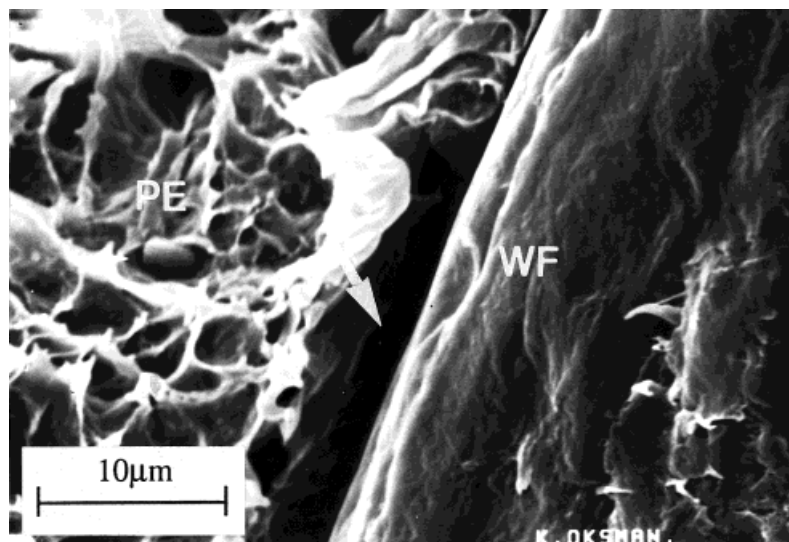
Figure 8 SEM micrograph of room-temperature fracture specimens showing the interface/interphase region between the wood filler and the LDPE matrix.

tain any moisture; the only possibility is moisture penetration into the sample surfaces at low temperature during the DMTA measurement. However, the conditions were the same for both systems and the EB tan δ peak amplitude did not increase in the SEBS/WF system.

Figure 6 shows that the styrene transition peak position is unaffected and that the amplitude decreased with increasing WF content for the SEBS–MA/WF system. A decreased number of molecules of styrene due to the addition of WF is an expected result. That the PS part did not show any interaction with the wood was not expected. Earlier studies showed that the PS tan δ peak shifts about 10°C toward higher temperatures when WF was added to the PE/PS composites.[18–20] The lack of interaction between the wood and PS can be due to the wood surface character being changed due to esterification and hydrogen bonding between OH— groups in the wood and MA. However, for the SEBS/WF system shown in Figure 7, the PS tan δ peak position is unaffected by the addition of WF as was the case with the SEBS–MA system.

Electron Microscopy

Examination of the fractured surfaces of the composites by SEM can provide information about the adhesion in the filler/matrix interface. The previous electron microscopy study of LLDPE/WF composites with SEBS and SEBS–MA as compatibi-

lizers showed improved adhesion between the matrix and wood particles when SEBS–MA was used, while no interaction was seen when SEBS was used.[7]

Figure 8 shows the microstructure of the injection-molded LDPE/WF composite, showing a wood particle embedded in the LDPE matrix. The interphase region between the wood particle and the polymer matrix shows that the interface is clean and that there is a gap between the wood particle and PE matrix which indicates poor adhesion between the surfaces. Further evidence for this is that the wood particle has a smooth surface and it can be easily seen that the fracture passes through the weak interface.

Figure 9(a) shows the microstructure of the injection-molded LDPE/WF composite with 5 wt % SEBS. The microstructure is similar to that of the LDPE/WF in Figure 8. Figure 9(b) shows the microstructure of the LDPE/WF composite with 5 wt % SEBS–MA. Good adhesion between the wood particle and polymer matrix can been seen with no gap between the WF particle and the polymer matrix. The wood surface is not as clean and smooth as in previous figures and the SEBS–MA is believed to locate on the wood surface.

The behavior seen in Figures 8 and 9 supports the composites' mechanical properties shown in Table I. The SEBS–MA copolymer compatibilizer is capable of improving the interfacial adhesion between wood and olefin polymers, resulting in increased tensile and impact strength.
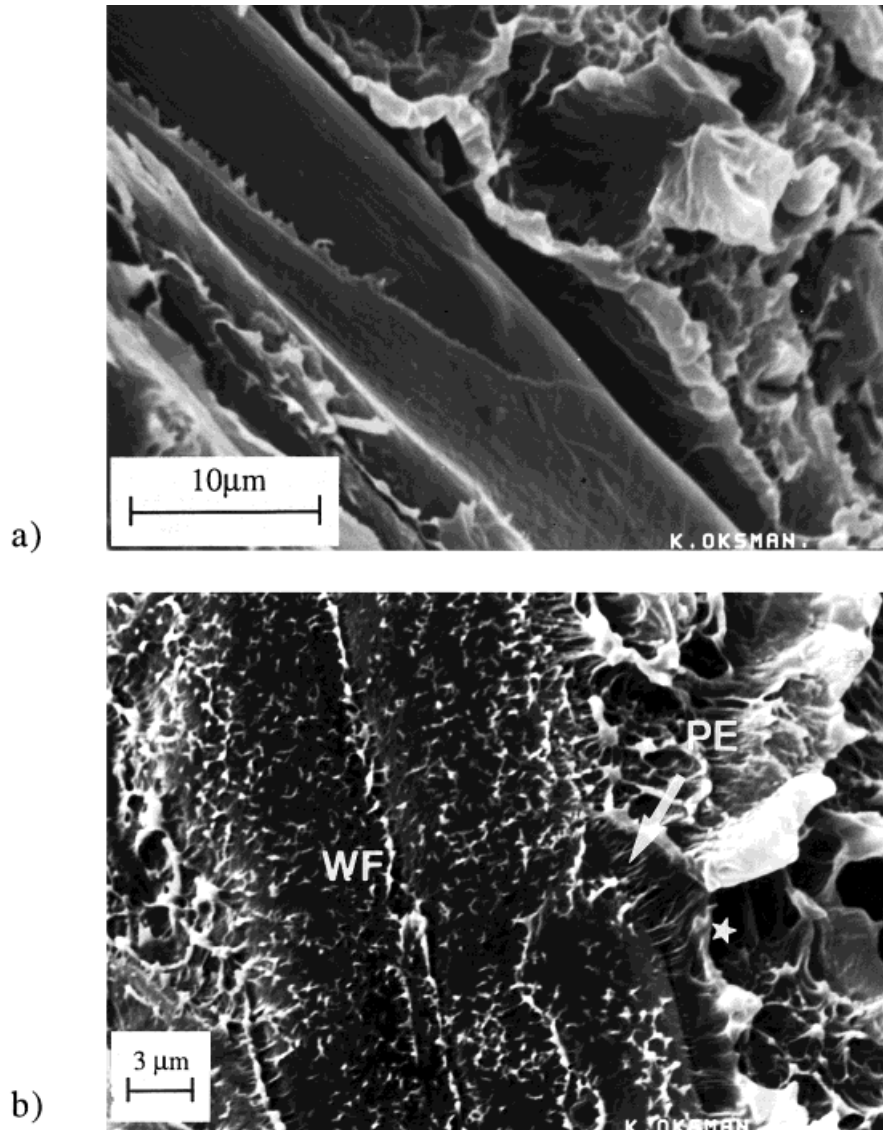
Figure 9 SEM micrograph of room-temperature fractured impact specimens: (a) LDPE/WF composite with the SEBS compatibilizer; (b) same composite with SEBS–MA as the compatibilizer.

Figure 10 shows the microstructure of Brabender-blended LDPE/SEBS–MA/WF composites where the SEBS–MA content is increased to 40 wt % to facilitate exploring the interphase region between the WF and the polymer matrix. The micrograph shows very good interfacial adhesion between the PE/SEBS–MA matrix and the wood particles. It is difficult to differentiate the wood particles from the polymer matrix when compared to the systems shown in Figures 8 and 9(a). It can been seen that the fracture paths passed either through the wood particles or through the polymer matrix and not in the interface between

these. The wood particle surfaces are covered by the matrix polymer. There are no gaps between the wood and the LDPE/SEBS–MA matrix. This confirms that SEBS–MA acts as a compatibilizer in LDPE/WF composites.

The SEBS–MA and SEBS were compounded with the LDPE/WF compound in a injection-molding machine and it is possible that compounding is not as good if compared to the Brabender mixer or extrusion. However, the results of both mechanical testing and electron microscopy have shown that the SEBS–MA has improved the interfacial adhesion between the wood particles
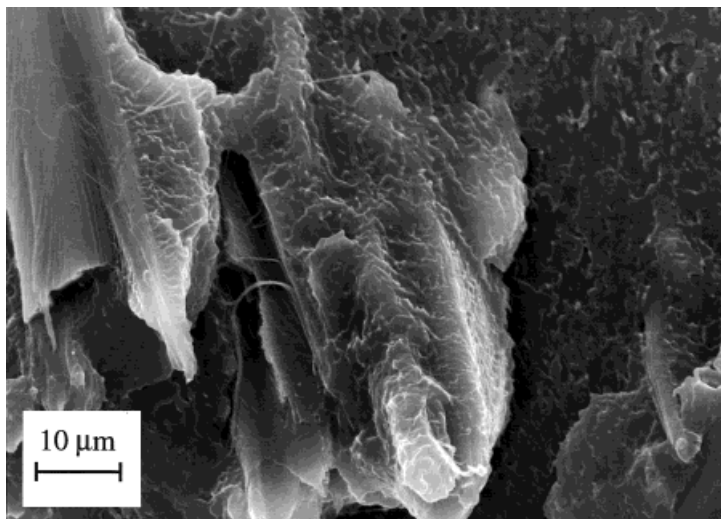
Figure 10   SEM micrograph of liquid nitrogen fracture specimen for LDPE/WF composites with high (40 wt %) SEBS–MA content.

and the LDPE matrix on the injection-molded composites and, therefore, the blends are expected to have sufficient compounding.

CONCLUSIONS

The SEBS–MA acts as a compatibilizer between wood particles and the polyolefin matrix in LDPE/WF composites. The composites' tensile strength was improved with the addition of SEBS–MA and reached its maximum level with 4 wt % SEBS–MA content. The addition of the compatibilizer had a positive effect on the impact strength and elongation at break but decreased the composites' tensile modulus.

The compatibilizer mechanism between SEBS–MA and the wood surface is believed to depend on chemical bonding (esterification) and hydrogen bonding between the MA and OH— groups on the wood particle surface. DMTA measurements confirmed interaction between the EB part (where the MA is grafted) of the block copolymer and the WF and that interaction increased with increasing WF content. The primary transition of EB–MA was shifted to higher temperatures. The styrene part of the block copolymer did not show any interaction with the WF.

The shear lap adhesion test confirmed better adhesion between the wood and SEBS–MA compared to the wood and SEBS. The electron microscope study showed good adhesion in LDPE/WF/SEBS–MA composites, while composites without the compatibilizer and with SEBS did not show any interaction between the WF and the polymer matrix.

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