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Effect and mechanism in compatibilization of poly(styrene-*b*-2-ethyl-2-oxazoline) diblock copolymer in poly(2,6-dimethyl-1,4-phenylene oxide)/poly(ethylene-*ran*-acrylic acid) blends

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Abstract

The compatibilization effect of poly(styrene-*b*-2-ethyl-2-oxazoline) diblock copolymer, P(S-*b*-EOx), on immiscible blends of poly(2,6-dimethyl-1,4-phenylene oxide) (PPO) and poly(ethylene-*co*-acrylic acid) (EAA) is examined in terms of phase structure and thermal, rheological and mechanical properties, and its compatibilizing mechanism is investigated by Fourier-transform infrared spectroscopy. The block copolymer, synthesized by a mechanism transformation copolymerization, is used in solution blending of PPO/EAA. Scanning electron micrographs show that the blends exhibit a more regular and finer dispersion on addition of a small amount of P(S-*b*-EOx). Thermal analysis indicates that the glass transition of PPO and the lower endothermic peak of EAA components become closer on adding P(S-*b*-EOx), and the added diblock copolymer is mainly located at the interface between the PPO and EAA phases. The interfacial tension estimated by rheological measurement is significantly reduced on addition of a small amount of P(S-*b*-EOx). The tensile strength and elongation at break increase with the addition of the diblock copolymer for PPO-rich blends, whereas the tensile strength increases but the elongation at break decreases for EAA-rich blends. This effect is interpreted in terms of interfacial activity and the reinforcing effect of the diblock copolymer, and it is concluded that the diblock copolymer plays a role as an effective compatibilizer for PPO/EAA blends. The specific interaction between EAA and polar parts of P(S-*b*-EOx) is mainly hydrogen bonding.

Keywords: Blends; Compatibilization; Poly(2,6-dimethyl-1,4-phenylene oxide)

1. Introduction

The use of polymer blends is becoming an important factor in satisfying the needs of specific sectors of the polymer industry owing to economic incentives [1–3]. Though not always the most efficient, blending is the least expensive and most versatile technique that can produce new polymeric materials from existing commodity polymers. Most of polymer blends, however, are immiscible at the molecular level because the combination entropy of mixing of two polymers is drastically smaller than that for low molecular weight compounds, whereas the enthalpy of mixing is often a positive quantity or near zero. The immiscibility between polymeric pairs is responsible for the poor phase structure and mechanical properties of polymer blends. Therefore, enhancing the compatibility of immiscible polymer pairs is a key technology to obtain polymer blends with desirable properties.

Various methods have been used for compatibilizing polymer blends, namely, the introduction of strong specific interaction (e.g. hydrogen bonding [4,5], ion–dipole interaction [6,7], ion–ion interaction [8], ion–dipole interaction [9,10], intramolecular repulsive interaction [11,12], cocrystallization [13,14], chemical reactions [15,16], etc.), the formation of an interpenetrating network and cross-linking [17], and the addition of a block or graft copolymer. The addition of block or graft copolymer in blend is very similar in a sense to the emulsifying effect of a surfactant in oil/water mixtures [18,19]. So these block copolymers are often referred to as interfacial agents or compatibilizers.

The types of block copolymers giving interfacial activity in compatibilization have been studied by many workers. Generally, when an A-*b*-B diblock copolymer is added to an A/B binary blend, a compatibilizing effect of the copolymer

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is observed [20–22]. On the other hand, the approaches using an A-*b*-C diblock copolymer to bridge the incompatibility gap between two polymers A and B have also proven to be valid [23,24]. Lately, there are a few reports on the blend of immisible polymers (A and B) and a C-*b*-D diblock copolymer, where A and B are miscible or at least partially miscible with C and D, respectively [25,26]. The type of compatibilization extends the application of a compatibilizer in many polymer pairs.

In fact, the mixing of a block copolymer with homopolymers having identical units is thermodynamically an athermal process. However, when a block copolymer is blended with two immiscible polymers, where blocks are chemically different from, but miscible with, the corresponding polymers, an exothermic interaction between miscible pairs exists. This interaction may afford an additional driving force for compatibilizing immiscible polymer blends.

PPO is a rigid polymer of comparatively high modulus and strength, high heat and chemical resistance, good dimensional stability, and low water absorption. EAA is an ethylene–acrylic acid random copolymer. Its attached functional groups may provide specific interaction with other polymers, and it is soft, tough, transparent and similar to low density polyethylene (LDPE) in large elongation and low viscosity [27], which compensate that of PPO in properties. In addition, it is lower in price than PPO for commercial use. Blending of these two polymers may offer mutually complementary, desirable physico-mechanical properties only if a proper compatibilizer is available. In this study, a styrene/2-ethyl-2-oxazoline diblock copolymer is used as a potential compatibilizer for PPO/EAA blends, where the polystyrene block is known to be miscible with PPO and the poly(2-ethyl-2-oxazoline) block is expected to be partially miscible with EAA through mutual interaction of polar groups. Its compatibilizing effect is examined through morphology, thermal behavior, rheological and mechanical properties. The compatibilizing mechanism of the diblock copolymer in the blends is supported by infrared spectroscopy.

2. Experimental

2.1. Materials and preparation of blends

The EAA (Dow, Primacor C1090) had an acrylic acid content of 13.9 mol% estimated by elemental analysis, and contained no metal ions (atomic absorption spectroscopy). The PPO, a commercial additive-free product, was used as received.

The poly(styrene-*b*-2-ethyl-2-oxazoline) diblock copolymer, P(S-*b*-EOx), was synthesized through mechanism transformation copolymerization [28]. The anionic polymerization of styrene was initiated by *n*-butyllithium and then termination with ethylene oxide and *p*-toluenesulfonyl chloride, followed by cationic ring-opening polymerization of 2-ethyl-2-oxazoline. The diblock copolymer was extracted successively with cyclohexane and methanol. The block ratio of polystyrene (PS) to poly(2-ethyl-2-oxazoline) (PEOx) was estimated to be 49/51 (wt/wt) as measured by ^{1}H n.m.r. The number-average molecular weight (M_n) of P(S-*b*-EOx) and the polydispersity index (M_w/M_n) were estimated by gel permeation chromatography (g.p.c.) to be 2.07×10^4 g/mol and 1.32, respectively.

A sample of poly(2-ethyl-2-oxazoline) (PEOx) with M_w of 2×10^4 g/mol was purchased from Aldrich Chemical Co.

o-Dichlorobenzene (C.P.) used was a standard reagent; the antioxidant 1010, tetrakis[1-(methoxycarbonyl)-2-3′,5′-di-*tert*-butyl-4′-hydroxylphenyl)ethyl]-methane, Beijing No. 3 Chemical Plant.

All polymers were completely dried under vacuum before solution-blending. The constituents of the blend and antioxidant 1010 were dissolved in *o*-dichlorobenzene in a flask equipped with a nitrogen inlet at 135°C and stirred for 6 h. Then the homogenous solution was poured into an excess of hexane. The precipitate was collected after 24 h, washed several times with hexane, and then dried in vacuum at 85°C for 5 days.

2.2. Instrumental methods

Thermal analysis was carried out using a Perkin-Elmer DSC-2C under nitrogen atmosphere. The heating rate was 20°C/min and data in the first heating were collected.

The compression-moulded blend specimens were fractured at liquid-nitrogen temperature, and the fractured surface etched with chloroform for EAA-rich blends and with toluene at 120°C for PPO-rich blends and coated with gold for scanning electron microscope (SEM, JEOL JAX-840) examination. To determine the average size, the IBAS image processing system was used. For each sample, five SEM photographs with more than 600 particles were analyzed, and then calculated for the number-average radius (R).

The dynamic rheological measurements were performed on Contraves Rheomat 120 using CP8 cone plate. Isothermal frequency sweeps were recorded at 290°C in an inert N_2 atmosphere. Specimens with dimensions suitable for rheological measurement were cut out from the same sheet as used in the morphological observation.

Tensile properties of the samples were tested on an Instron 1121 electronic tester at a drawing rate of 50 mm/min. The compression-moulded sheets were cut into standard dumbbell test specimens.

Infrared spectra were obtained on a BIO RAD FTS-7 *FT*i.r. spectrometer. At least 20 scans at a resolution of 1 cm^{-1} were averaged. A sample of PEOx/EAA blend was obtained from evaporation of the *o*-dichlorobenzene solution in vacuum on a NaCl plate.

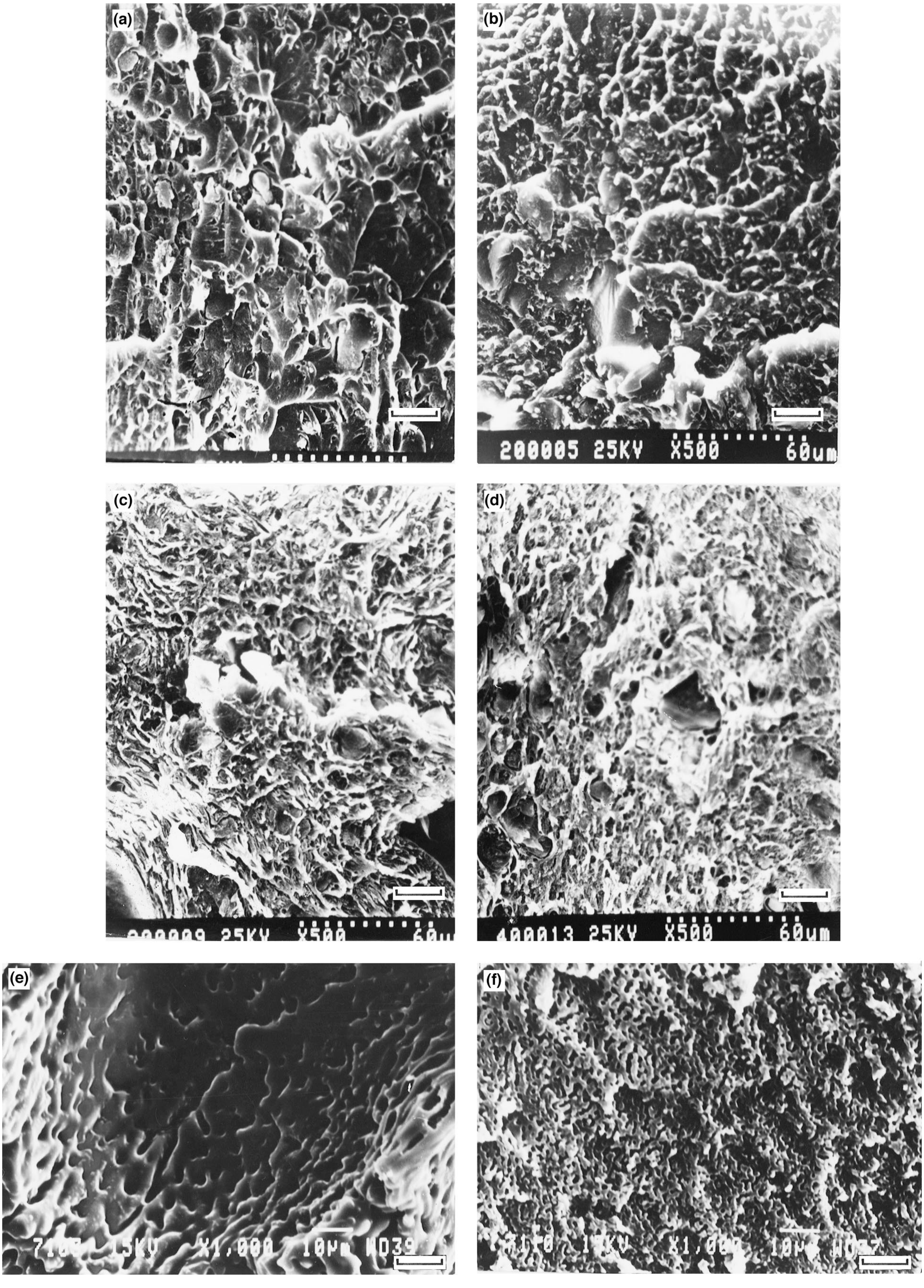

Fig. 1. Scanning electron micrographs of PPO/EAA 30/70 and 90/10 blends: (a) (30/70) 0; (b) (30/70) 2; (c) (30/70) 4.8; (d) (30/70) 10.7; (e) (90/10) 0; (f) (90/10) 4.8 wt% P(S-*b*-EOx) added. The magnitude for each marker is 20 μm.

3. Results and discussion

3.1. Morphology

Interfacial tension, adhesion between two phases, and melt viscosities of the blended polymers are key parameters governing the degree of dispersion and stability against coalescence or stratification [29].

Morphology changes in 30/70 (wt/wt) PPO/EAA blends on addition of P(S-*b*-EOx) are shown in Fig. 1, where the dispersed phase, PPO, had been etched with chloroform for clear observation. The large domain size and non-uniform size distribution of the blend (Fig. 1(a)) which are determined by, generally speaking, large interfacial tension, poor interface adhesion, and difference in melt viscosity of two phases indicate the immiscibility of PPO and EAA. When a small amount of P(S-*b*-EOx) is added (Fig. 1(b, c)), more regular and finer dispersion is observed. This results from the decrease in the interfacial tension between the two immiscible polymers and/or the enhancement of interfacial adhesion. When the amount of P(S-*b*-EOx) is further increased up to 10.7 wt% (Fig. 1(d)), the morphology of the blend does not change much as compared to that of the compatibilized blend with 4.8 wt% P(S-*b*-EOx). The similar phenomenon is also observed for 90/10 (wt/wt) PPO/EAA blends. In the uncompatibilized blend (Fig. 1(e)), the particles of the dispersed phase, EAA, are coarse, and their average radius is 2.38 μm. However, in the blend compatibilized by 4.8 wt% P(S-b-EOx) (Fig. 1(f)), the average radius of particles of the dispersed phase is reduced to 1.14 μm.

It is evident from these morphological observations that poly(styrene-*b*-2-ethyl-2-oxazoline) diblock copolymer plays a role as an effective compatibilizer for PPO/EAA blends.

3.2. Thermal analysis

It is informative to verify whether the block copolymer is located at the interface and that each block is dissolved into the corresponding blend domains. The location of the block copolymer could be elucidated by examining the information on phase composition.

Fig. 2 shows the DSC thermograms of PPO/P(S-*b*-EOx) binary blends. The T_g of PPO is 217.2°C (Fig. 2(a)), whereas the block copolymer has two T_gs at 59.3 and 105.1°C (Fig. 2(d)). For PPO/P(S-*b*-EOx) binary blends, also two T_gs are observed: the lower T_g corresponds to the PEOx block of the diblock copolymer, and a single, composition-dependent upper T_g shifts to a higher temperature at a higher concentration of PPO (Fig. 2(b–d)). The upper T_g follows the prediction by the Fox equation [30], although at too high concentration of PPO T_g is slightly lower than this prediction, suggesting that the upper T_g corresponds to a mixed phase of the PS block of P(S-*b*-EOx) and PPO [31]. Thus, the two-phase structure of the PPO/P(S-*b*-EOx) binary blends is composed of a PPO/PS phase and a PEOx phase.

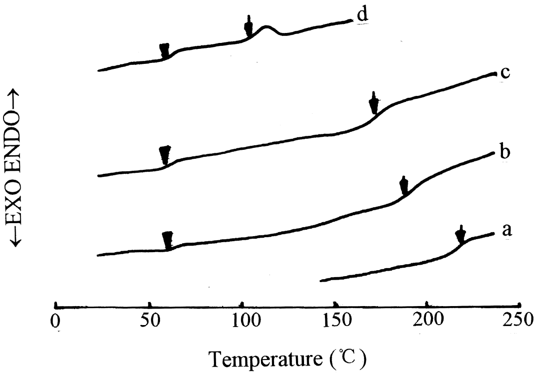

Fig. 2. DSC thermograms of PPO/P(S-*b*-EOx) binary blends. Composition (weight ratio): (a) 100/0; (b) 70/30; (c) 50/50; (d) 0/100.

Before examining a binary blend of EAA/P(S-*b*-EOx), it is necessary to discuss the melting behavior of EAA. It is known that EAA displays two endothermic peaks when annealed for more than 2 months at ambient temperature [32–34]. It is suggested that the higher endothermic peak at 97.2°C corresponds to the melting of the polyethylene crystallites in the polymer matrix and the lower endothermic peak at 31.7°C to the dissociation of acrylic acid 'aggregates' held by strong intramolecular hydrogen bonding interaction [32,34]. The former is always observed, while the latter is observed only on the first scan after a sufficient aging of EAA, shifting to higher temperature and becoming stronger on longer annealing. Thus, the lower endothermic peak of EAA provides information on intramolecular hydrogen-bonding strength.

Fig. 3 shows DSC thermograms of EAA/P(S-*b*-EOx) binary blends aged for 2 months at room temperature. As the diblock copolymer is added to EAA, the lower endothermic peak of EAA at 31.7°C shifts slightly to a higher temperature and becomes smaller until unclear (Fig. 3(b, c)). This indicates the change in bond strength in EAA on addition of the block copolymer, or intermolecular hydrogen bonding between the PEOx blocks in P(S-*b*-EOx) and the acrylic acid in EAA.

It is known that EAA is strongly self-associating (see Fig. 4) [32,35,36], and PEOx is a tertiary amide polymer

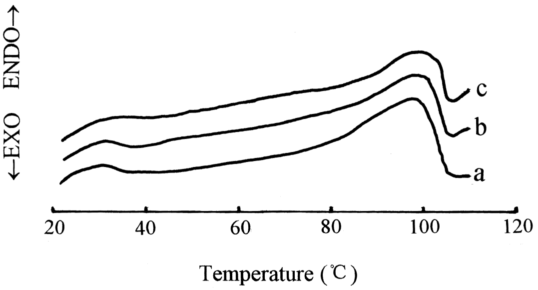

Fig. 3. DSC thermograms of EAA/P(S-*b*-EOx) binary blends. Composition (weight ratio): (a) 100/0; (b) 70/30; (c) 50/50.

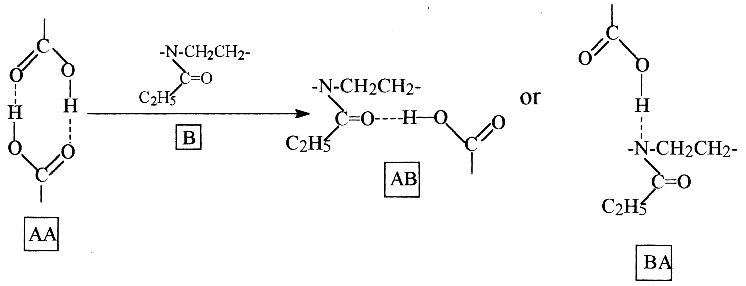

Fig. 4. Schematic diagram illustrating EAA-PEOx interaction.

[5,37]. In order to understand the interaction between EAA and the diblock copolymer, the PEOx/EAA binary blends are subjected to *FT*i.r. examination (Fig. 5). In addition to the two bands attributed to the carboxylic acid dimer (AA, 1698 cm^{-1}) and the unassociated oxazoline (B) groups (1645 cm^{-1}), two new bands are evident at 1724 and 1614 cm^{-1}. The former band is a free EAA carbonyl group (AB) formed when the EAA hydroxyl group associates with the PEOx carbonyl group (see Fig. 4), and the 1614 cm^{-1} band is assigned to PEOx carbonyl groups that are bonded to the EAA hydroxyls of the carboxylic acid groups (BA or AB) through hydrogen-bonding. Certainly, there are two possible PEOx sites for hydrogen bonding to the EAA carboxylic acid group, the carbonyl oxygen and the nitrogen atoms [35,36]. The above *FT*i.r. results point out that specific intermolecular interactions, mainly hydrogen bonding, induce partial miscibility of the PEOx block in P(S-*b*-EOx) and EAA.

DSC thermographs of PPO/EAA 30/70 blends with the addition of P(S-*b*-EOx) that had been aged at room temperature for 2 months are shown in Fig. 6. In the presence of the diblock copolymer, glass transition of PPO and the lower endothermic peak of EAA shift slightly towards each other, inferring mutual interaction, to be specific, the styrene blocks dissolving into PPO and the PEOx blocks interacting with EAA. Consequently, it is concluded that the diblock copolymer is located mainly at the interface of the two immiscible polymers PPO and EAA, and that each block of the copolymer penetrates into the corresponding component polymer, although the possibility of the existence of the diblocks themselves in either or both phases as micelles and/or as individual molecules cannot be excluded [38,39].

3.3. Rheological properties

For Newtonian fluids, Stone et al. [40] proposed a model based on the study of the rheology of a double emulsion, where the dispersed phase has a core-shell structure. The model suggested that the behavior of dispersed phase is governed by the shell viscosity when it exceeds the core viscosity. In this work, for blend components, PPO, EAA and P(S-*b*-EOx), from Fig. 7, it appears clearly that below 15.8 rad/s the viscosity of the diblock copolymer is higher than that of EAA. Therefore, considering that P(S-*b*-EOx) is located at the interface between the EAA and the PPO phases, where EAA is the minor phase for the PPO/EAA 90/10 blend, the diblock copolymer layer surrounding the EAA will govern the rheology of the dispersed phase. Let us describe the rheology of the blend modified by P(S-*b*-EOx) as a two-phase blend in which the dispersed phase behaves like the diblock copolymer. The volume fraction of the dispersed phase is then equal to the sum of the volume fractions of EAA and P(S-*b*-EOx).

In a theoretical description of the rheology of viscosity emulsion following the work of Oldroyd [41], Palierne [42] showed that the complex modulus is a function of the component moduli, the surface tension, and the viscoelastic effects of the interface. Graeling and Muller [43] found good agreement between their experimental data and the predictions of Palierne's model by neglecting the

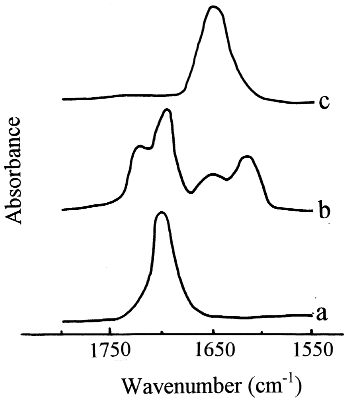

Fig. 5. FTIR spectra of films samples: (a) EAA; (b) EAA/PEOx 50/50 blend; (c) PEOx.

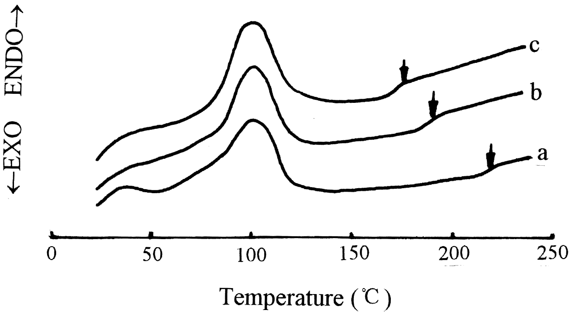

Fig. 6. DSC thermograms of PPO/EAA 30/70 blends: (a) 0; (b) 4.8; (c) 10.7 wt% P(S-*b*-EOx) added.

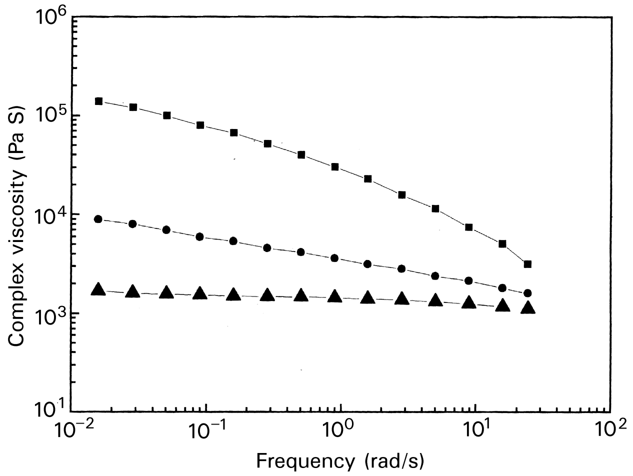

Fig. 7. Complex viscosity of P(S-*b*-EOx) at 290°C as compared to PPO and EAA. (■) PPO; (●) P(S-*b*-EOx); (▲) EAA.

viscoelastic contribution of the interface. They used the following expression for the complex modulus:

$$G^* = G_M^* \left(\frac{1 + 3\varphi H}{1 - 2\varphi H} \right) \tag{1}$$

where

$$H = \frac{4(\sigma/R)(2G_M^* + 5G_I^*)(G_I^* - G_M^*)(16G_M^* + 19G_I^*)}{40(\sigma/R)(G_M^* + G_I^*) + 2(G_I^* + 3G_M^*)(16G_M^* + 19G_I^*)} \tag{2}$$

where φ is the volume fraction of the dispersed phase, G_I^* and G_M^* are the complex moduli of the components of the dispersed phase (I) and the matrix (M), respectively, σ is the interfacial tension, and R is the average radius of particles of the dispersed phase.

In the above equations, for the uncompatibilized PPO/EAA 90/10 blend, I and M refer to the EAA and the PPO phases, respectively. Thus, σ indicates the interfacial tension between PPO and EAA. However, for the compatibilizing blend, following our previous discussion, G_I^* becomes the complex modulus of P(S-*b*-EOx) and σ is the interfacial tension between P(S-*b*-EOx) and the matrix. The coefficient σ/R has been used as an adjustable parameter in Eq. (2).

Fig. 8 shows the rheological data of the two blends without and with the addition of P(S-*b*-EOx). The comparison between our experiments and the theoretical predictions from a combined model of Stone and Palierne of these blends are also given. For the two blends, the values of σ/R, which provide the best fit for the combined model, are different (Table 1). This can be interpreted by means of the revised Taylor equation [44,45]:

$$\frac{G\eta_m A}{\sigma} = F(\eta_r)$$

where σ is the interfacial tension, η_m the viscosity of the matrix phase, G the shear rate and $F(\eta_r)$ the function of the relative viscosity.

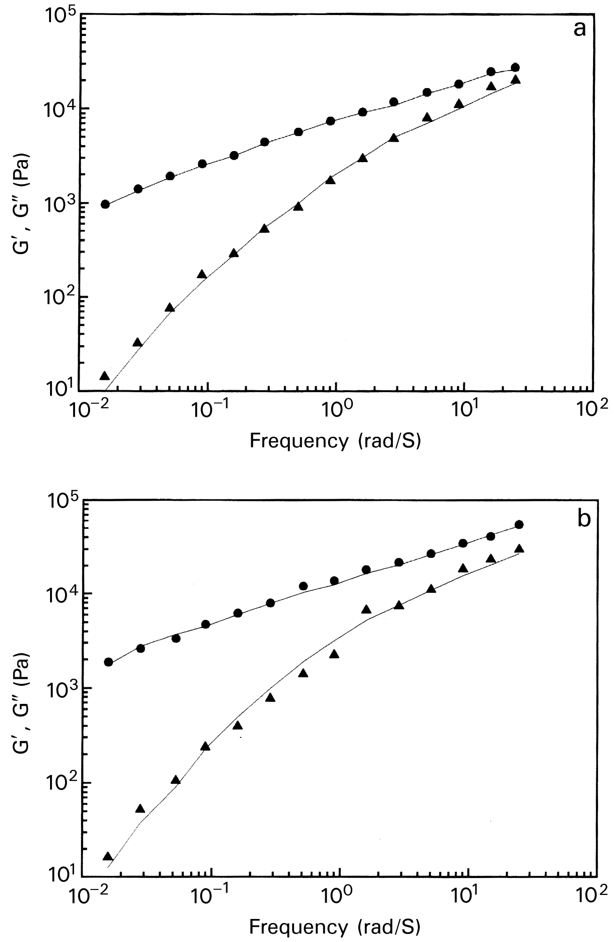

Fig. 8. Comparison of the prediction of the combined model (curves) with the experimental results for PPO/EAA 90/10 blends: (a) uncompatibilized blend; (b) with the addition of 4.8 wt% P(S-*b*-EOx). (▲) G′ experiments; (●) G″ experiments.

Taking $A = 2R$, where R is average radius of particles of the dispersed phase, we get:

$$\sigma/R = \frac{2G\eta_m}{F(\eta_r)} \tag{3}$$

It is clear from Eq. (3) that the difference in η_r, which results from the different dispersed phase in the two blends, induces the different σ/R.

It can be seen from Table 1 that the interfacial tension decreases on the addition of the diblock copolymer. This further proves that P(S-*b*-EOx) acts as an effective compatibilizer.

Table 1
Interfacial tension (σ) of PPO/EAA blends estimated from rheological measurements

Blends (weight ratio)	R[a]	φ[b]	σ (× 10^{-3} N/m)
PPO/EAA 90/10	2.38	0.1	2.31
PPO/EAA/P(S-b-EOx) 90/10/5	1.14	0.143	0.37

[a] Determined from SEM micrographs (see Figure 1).
[b] Volume fraction of the dispersed phase.

3.4. Mechanical properties

In the case of PPO/EAA blends, two types of stress–strain curves are obtained depending upon the blend composition: one is for PPO-rich blends and the other is for EAA-rich blends. As shown in Fig. 9, the PPO/EAA 70/30 blend exhibits a semibrittle behavior whereas the 30/70 blend shows considerable ductility. Fig. 10 shows the mechanical properties of the two PPO/EAA blends of different compositions, each modified by varying the amount of P(S-*b*-EOx). All blends have increasing tensile strength and modulus (Fig. 10(a, b)) with increasing amount of P(S-*b*-EOx) when it is lower than 4.8 wt%. However, the elongation at break changes in different trends: increasing for the 70/30 PPO/EAA blends and decreasing for the 30/70 PPO/EAA blends with the addition of P(S-*b*-EOx) (Fig. 10(c)). Also, Fig. 10 shows that all the mechanical properties of the blends is almost unchanged with the amount of P(S-*b*-EOx) when it is higher than 4.8 wt%. The tensile properties of immiscible and partially miscible blends depend on two interrelated factors: the adhesion between the two phases and the domain size of the dispersed component, both controlled mainly by the interfacial tension [45,46]. Based on our results, it is reasonable to conclude that all mechanical properties are changed as a result of a decrease in the interfacial tension of PPO and EAA with P(S-*b*-EOx), i.e. the interfacial activity of P(S-*b*-EOx) and its reinforcing effect in PPO/EAA blends. In the meantime, these results also indicate that the diblock

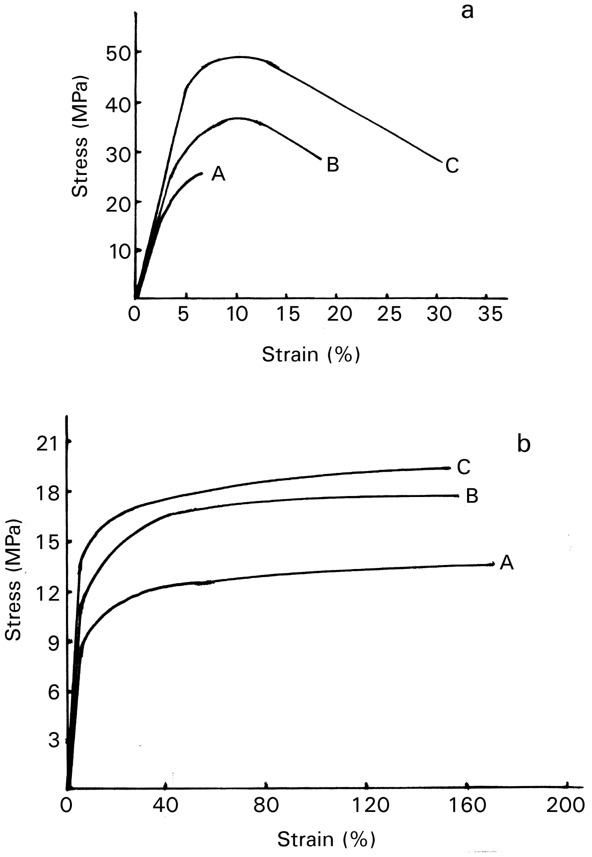

Fig. 9. Stress–strain curves of PPO/EAA 70/30 (a) and 30/70 (b) blends: (A) 0; (B) 2; (C) 4.8 wt% P(S-*b*-EOx) added.

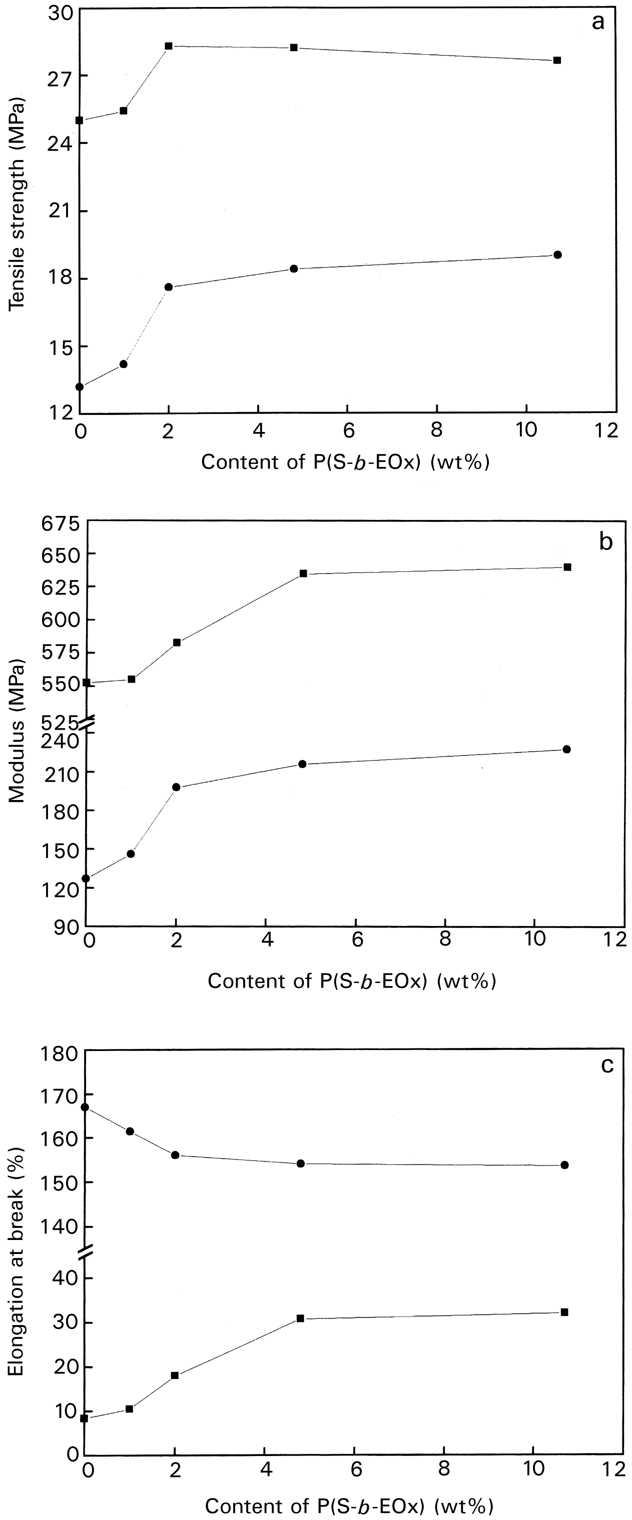

Fig. 10. Mechanical properties—(a) tensile strength, (b) modulus, and (c) elongation at break—of PPO/EAA blends of compositions of (■) 70/30; (●) 30/70 (weight ratio) compatibilized by P(S-*b*-EOx) of varying amounts.

copolymer is located at the interface between the two phases when the concentration of the compatibilizer is lower than a critical concentration, which is indicative of the point of interfacial saturation of the diblock copolymer on the dispersed phase [39,46]. For the PPO-rich blends, the added P(S-*b*-EOx) reduces the defects in the blend, improves the adhesion between the two phases, and helps stress and strain transfer through the interface. For the EAA-rich blends, the addition of the copolymer gives better adhesion between the two phases and thus the properties of the dispersed phase (PPO) (higher modulus and strength and lower elongation than the matrix phase (EAA)) play a more significant role in the blend properties.

It is well known that the blend composition has effects on the tensile properties of the blends with the two-phase structure. Fig. 11 shows the tensile properties of the PPO/EAA blends without and with the addition of 4.8 wt% of diblock copolymer. For the former, the tensile strength and modulus of the blends show large negative deviation from additivity, however, as P(S-*b*-EOx) is added the deviation diminishes towards the simple additivity rule (Fig. 11(a, b)). The elongation at break of various PPO/EAA blends is shown in Fig. 11(c). In the case of the uncompatibilzed PPO/EAA blends, the negative deviation of the EAA-rich blends is smaller than that of PPO-rich blends. This is related to the different compatibilizing effect of P(S-*b*-EOx) on the blends with different matrix phases, as shown above.

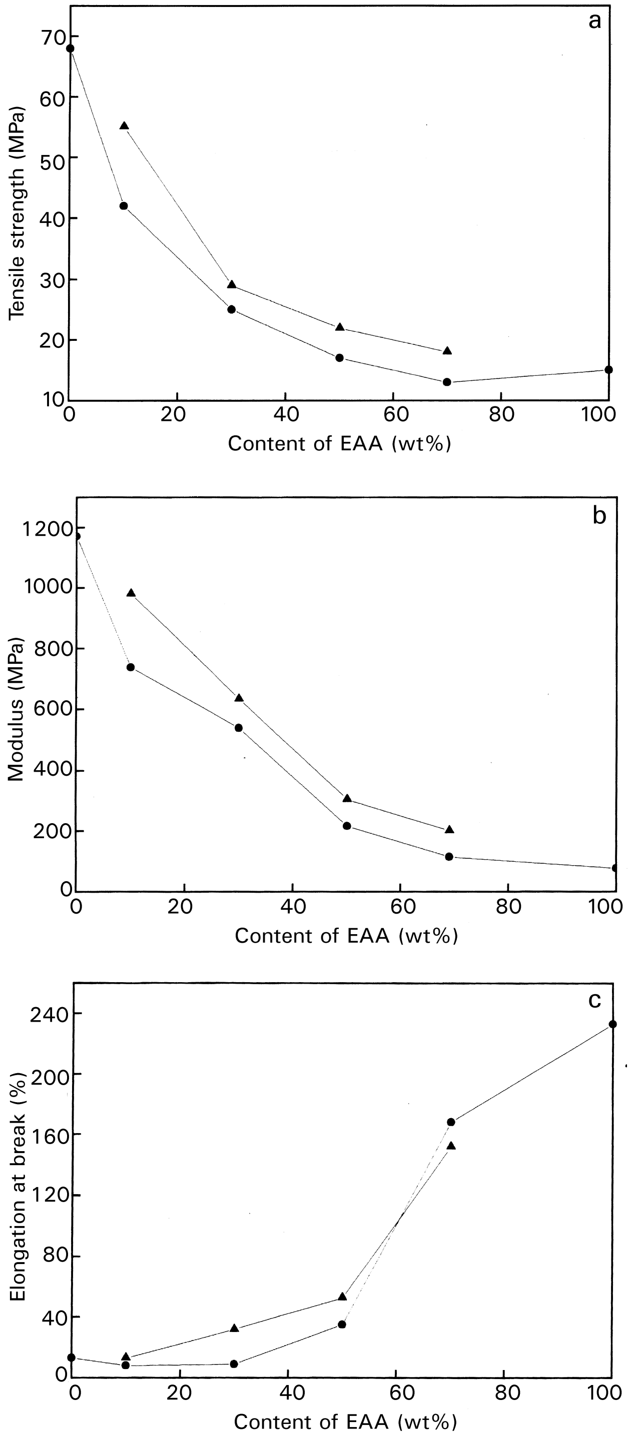

Fig. 11. Mechanical properties—(a) tensile strength, (b) modulus, and (c) elongation at break—of PPO/EAA blends of varying compositions compatibilized by P(S-*b*-EOx) amounts of (•) 0; (▲) 4.8 wt%.

4. Conclusions

This study focuses on the compatibilizing effects of C-*b*-D type block copolymer on blends of two immiscible polymers A and B, as exemplified by the use of poly(styrene-*b*-2-ethyl-2-oxazoline) diblock copolymer for the PPO/EAA system. By experimental techniques of SEM, DSC, and measurements of rheological and mechanical properties, the effects of the block copolymer on the phase structure, rheological and mechanical properties of the immiscible blend system were investigated. Also, the effect of the block copolymer on the interfacial tension of the two immiscible polymers was examined by applying a combined model of Stone and Palierne to the data obtained from rheological measurements.

More regular and finer dispersion was observed upon addition of P(S-*b*-EOx) to the PPO/EAA blends. This suggests that the interfacial tension between the two phases was reduced by the addition of the copolymer. From DSC results of binary or ternary blends of PPO, EAA and P(S-*b*-EOx), it is believed that P(S-*b*-EOx) in the ternary blends mainly locates at the interface between the two blend components, PPO and EAA, and each block is soluble or partially soluble in the corresponding components. The FTIR analysis proves that the partial miscibility of the PEOx block in P(S-*b*-EOx) and EAA is due to specific interaction between them. The rheological measurements indicates that the addition of P(S-*b*-EOx) leads to a decrease in interfacial tension, and the theoretical prediction from a combined model shows good agreement with the experimental data. By modifying with a small amount of P(S-*b*-EOx), the mechanical properties of the blends (tensile strength, modulus and elongation at break) changed for different compositions of PPO/EAA. The three properties increase with the addition of P(S-*b*-EOx) for PPO-rich blends, whereas the tensile strength and modulus increase, but the elongation at break decreases for EAA-rich blends.

Acknowledgements

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