Relationship Between Interfacial Tension and Dispersed-Phase Particle Size in Polymer Blends. I. PP/EPDM

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ABSTRACT: Simple blends with different viscosity ratios of the components as well as compatibilized blends varying both in type and content of compatibilizers were used to study the relation between interfacial tension and dispersed-phase particle size for PP/EPDM (80/20 wt %) blends in this work. Four compatibilizing systems, poly(ethylene-*co*-methacrylic acid) ionomers (EMA–I), dicumyl peroxide (DCP), DCP combined with EMA–I, and DCP in combination with trimethylol propane triacrylate (TMPTMA), were used. For blends prepared in an internal mixer, a power law relation was found between capillary number and torque ratio of the blends' components. This relation was used to estimate the interfacial tension for the compatibilized blends. The relation between steady-state torque of the blends as a measure of viscosity and the estimated values of interfacial tension were also investigated. © 2002 Wiley Periodicals, Inc. J Appl Polym Sci 86: 3148–3159, 2002

Key words: interfacial interaction; morphology; PP/EPDM blends; compatibilizers; viscosity ratio

INTRODUCTION

The majority of commercial polymer blends possess a multiphase morphology, and in most cases the satisfactory physical and mechanical properties of these materials are related to the presence of a stabilized finely dispersed phase.

The control of the morphology of the dispersed phase in such systems depends on varying degrees of interfacial interaction,[1–3] viscosity ratio,[1,4,5] shear rate,[5,6] elasticity of components,[8,10] and processing conditions.[3–11] The interfacial tension is one of the most effective parameters, such that a knowledge of its contribution to morphology can provide great insight into controlling the morphology and thus the final mechanical properties of these blends.[3]

A variety of techniques have been established for interfacial modification of polymer blends that have been classified into reactive and nonreactive interfacial modification.[12,13] Nonreactive modification can be attained by adding a third component as a compatibilizer capable of specific interactions, such as entrapment by mechanical interlocking, physical crosslinking, hydrogen bonding, or transition metal complexation.[12–14] The most common forms of such compatibilizers are suitable block or graft copolymers, terpolymers, and inomers. These compatibilizers are concentrated at the interface between blend components and act as emulsifiers, reducing interfacial tension and improving interfacial interaction.

In reactive compatibilization, the compatibilizer is produced during the melt-blending process. It can be achieved by blending functionalized constituents or by adding a low molecular weight compound or reactive polymers, which can lead to the *in situ* formation of copolymer as compatibilizer through crosslinking, grafting, or some other reactions.[13]

The main objective of the present work was to study the relationship between the interfacial tension and dispersed particle size in PP/EPDM (80/20 wt %) blends. To establish such a relationship, the role of the viscosity ratio of the blend components on the rubber particle size was studied.

THEORY

Polymer blending involves the mixing of a two-phase melt–melt polymeric system. The deformation applied on such systems from the mixer induces an increase in interfacial area that is counteracted by interfacial tension. With further deformation of the dispersed phase, a point is reached at which interfacial forces becomes comparable to the deformation rates applied by the blending device. In this stage interfacial area–reducing processes set in and droplets oppose against deformation. Finally, an equilibrium structure, consisting of droplets small enough to resist the disruptive forces, will be produced.[15]

It is an oversimplification to assume that these droplets retain their individuality over the course of the blending process, given that collision of droplets can lead to coalescence, and the resulting morphology is controlled by the balance of breakup and coalescence events.[15]

The first experimental and theoretical study on droplet deformation was conducted by Taylor,[15] who considered the deformation of Newtonian droplets in a Newtonian matrix in a simple shear-flow field. From his study it appeared that the behavior of droplets is governed mainly by two dimensionless groups: (1) the capillary number defined as $K = \eta_m R \dot{\gamma} / \gamma_{12}$, which is actually the ratio of disruptive viscous shearing forces ($\eta_m \dot{\gamma}$) and the interfacial forces (γ_{12}/R); and (2) the viscosity ratio $P = \eta_d / \eta_m$, where R is the droplet radius, $\dot{\gamma}$ is the mixing shear rate, γ_{12} is the interfacial tension between phases, and η_d and η_m are the viscosities of the dispersed phase and matrix, respectively.

Taylor[15] derived a relation for the maximum droplet size that would be stable, based on capillary number and viscosity ratio as

$$K = \frac{R \eta_m \dot{\gamma}}{\gamma_{12}} = \frac{2(P+1)}{(19/4)P + 4} \qquad P < 2.5 \qquad (1)$$

Equation (1) indicates that the viscosity ratio, the shear stress ($\sigma = \eta_m \dot{\gamma}$), and the interfacial tension are critical variables to consider in controlling droplet deformation and breakup in Newtonian fluids.

Taylor also predicted that no droplet breakup occurs when $P > 2.5$. Karam and Bellinger[17] and Tavgac[18] have also studied the breakup of Newtonian droplets in a Newtonian matrix over a wide range of viscosity ratios in a uniform shear flow. Their results described a U-shape dependency of critical capillary number (the capillary number above which the droplet breakup occurs) on viscosity ratio. Similar results have been reported by some other researchers.[19–21]

For molten polymer with viscoelastic behavior, deformation-resisting forces arising from elasticity can also affect the particle deformation. Van Oene[8] studied the mechanisms of two-phase formation in a mixture of two viscoelastic fluids. He pointed out that for such systems elasticity of the dispersed phase leads to an increase of the final droplet size and introduced a dynamic interfacial tension based on quiescent interfacial tension and first normal stress of dispersed phase and matrix.

On the other hand, when the concentration of the dispersed phase exceeds a critical value, collision of droplets can no longer be neglected and final droplet size can be predicted by the balance between droplet breakup and coalescence. There are several consistent reports in the literature that droplet diameter sharply increases with the concentration of the dispersed phase.[22,23] Tokita[24] has derived an expression for predicting the particle size of the dispersed phase in polymer blends as a function of composition, as follows:

$$d = (24/\pi)P\gamma_{12}\phi_d[\eta_m\dot{\gamma} - (4/\pi)P\phi_d E_{dk}] \qquad (2)$$

where d is the particle size, E_{dk} is the bulk breaking energy of the dispersed phase per unit volume, ϕ_d is the dispersed-phase volume fraction, and P is the probability that a collision will result in a coalescence. This relation indicates that the equilibrium particle size increases with the interfacial tension and volume fraction of the dispersed phase and decreases with the shear rate.

There are many other reports on nondilute mixtures of two viscoelastic fluids that support the relation between particle size and interfacial tension for the blends produced in intensive mixers such as twin-screw extruder and internal mixer. Wu,[1] for example, has obtained a correlation between the capillary number and viscosity ratio of some blends prepared in twin-screw extruders as follows:

$$\frac{R \eta_m \dot{\gamma}}{\gamma_{12}} = 4P^{\pm 0.84} \qquad (3)$$

where the (+) sign in the exponent applies for $P > 1$ and the (−) sign for $P < 1$. He pointed out that the minimum droplet size was obtained at a viscosity ratio about unity. His results also showed that lowering interfacial tension will result in a smaller particle size. Some other authors[3,25] have also shown that for compatibilized polymer blends, interfacial interaction is the most important parameter that can control morphology and final particle size.

EXPERIMENTAL

Materials

Four types of polypropylene (PP), varying in melt-flow index, and two types of ethylene–propylene–diene terpolymers (EPDM) with different Mooney viscosities but with almost the same C_2 content were used for blending. The interfacial agents used were poly(ethylene-co-methacrylic acid) ionomer (EMA–I), dicumyl peroxide (DCP), and trimethylol propane trimethacrylate (TMPTMA). The main characteristics of the materials used and their suppliers are listed in Table I.

Blend preparation

The first sets of blend samples were prepared by blending separately PP1, PP2, PP3, and PP4 each with

Material	Properties	Supplier
EPDM1	Mooney viscosity ML(1+4), 125°C = 28 C2 content = 50%, ENB content = 5%, ρ = 0.86 g/cm^3	Bayer Co., Buna AP241
EPDM2	Mooney viscosity ML(1+4), 125°C = 46 C2 content = 50%, ENB content = 5%, ρ = 0.86 g/cm^3	Bayer Co., Buna AP341
PP1	MFI[a] = 0.27, T_m = 165°C, ρ = 0.9 g/cm^3	Iran Petrochemical, MOPLEN D-60-R
PP2	MFI[a] = 0.7, T_m = 165°C, ρ = 0.9 g/cm^3	Iran Petrochemical, MOPLEN Q-30-G
PP3	MFI[a] = 1.5, T_m = 165°C, ρ = 0.9 g/cm^3	Iran Petrochemical, MOPLEN S-30-U
PP4	MFI[a] = 3, T_m = 165°C, ρ = 0.9 g/cm^3	Iran Petrochemical, MOPLEN I-30-G
EMA ionomer	Na-neutralized, % neutralization = 50% Ethylene/metharylic acid = 91/9 sp. gr. = 0.94, MFI[b] = 1	Dupont, Surlyn 8528
DCP	Granule type	
TMPTMA	Absorbed on calcium silicate sp. gr. = 1.26 Percent active = 72%	Akrochem, Akrosorb 9675

[a] 250°C/2.160 kg.
[b] 190°C/2.160 kg.

EPDM1 and EPDM2 in proportion of PP/EPDM 80/20 wt % without the use of any interfacial agent (Table II).

Second sets of the blend samples were prepared using different types of interfacial agent systems in PP1/EPDM1 (80/20) wt % blends, as described in Tables III and IV, for systems a and b and systems c and d, respectively.

Preparation of the blends was done by melt blending of the dried material in a Brabender internal mixer, equipped with roller-type (W50E) blades at 190°C and 60 rpm. Blending was carried out by first feeding PP into the mixer and after 3 min mixing, EPDM was charged into the mixer and 3 min later compatibilizing agents were added. Mixing was then continued for 10 min, after which the mixture was discharged.

The steady-state torque (SST) of the components and the blends, recorded after 10 min of mixing, was used as a measure of viscosity. Values of the steady-state torque ratio (TR) of the components for the simple blends are listed in Table II.

To consider the effect of compatibilization on the TR of the compatibilized blends, PP and EPDM were separately melt mixed with the same amount of interfacial agents as that used in the blends. The calculated steady-state TR values of the compatibilized blends are listed in Tables III and IV.

Morphology studies

Blend morphologies were studied using a scanning electron microscopy technique (SEM S360; Cambridge

TABLE II
Characterization of Simple Blends

Sample code	Matrix/composition (wt %)	Dispersed phase/composition (wt %)	TR[a]	η_d[b] (Pa · s)	η_m[b] (Pa · s)	d (μm)
1	PP1/80	EPDM1/20	1	1460	670	0.83
2	PP2/80	EPDM1/20	1.37	1460	608	0.98
3	PP3/80	EPDM1/20	1.78	1460	521	1.33
4	PP4/80	EPDM1/20	4.93	1460	320	3.5
5	PP1/80	EPDM2/20	1.63	189	670	1.1
6	PP2/80	EPDM2/20	2.25	189	608	1.4
7	PP3/80	EPDM2/20	2.9	189	521	4.8
8	PP4/80	EPDM2/20	8	189	320	5.2

[a] Ratio of steady-state torque of EPDM to PP at 10 min of mixing, 60 rpm, 190°C.
[b] Shear viscosity at 200 s^{-1}, 190°C.

TABLE III
Characterization of Compatibilized Blends (Systems a and b)

Sample code	Compatibilizing agent composition (wt %)			TR^a	d (μm)	Interfacial tension (γ_{12}) (mN/m)	Steady-state torque (SST)^b
	DCP	EMA-I	TMPTMA				
System a							
9	—	2	—	1	0.68	0.2635	15.8
10	—	4	—	1.04	0.42	0.1605	17.3
11	—	6	—	1	0.32	0.136	18
12	—	8	—	1.1	0.34	0.1254	17.1
13	—	10	—	1.2	0.36	0.1213	17.5
System b							
14	0.02	—	—	1.13	0.63	0.21	15.2
15	0.04	—	—	1.25	0.64	0.1426	15
16	0.06	—	—	1.34	0.96	0.260	14.8
17	0.08	—	—	1.53	1.06	0.2406	12.5
18	0.1	—	—	1.7	1.18	0.2268	11.8

^{a} Ratio of steady-state torque of EPDM to PP at 10 min mixing, 60 rpm, 190°C, and with the same content of compatibilizing agent as that of the blend.
^{b} Steady-state torque of blend at 10 min mixing, 60 rpm 190°C.

Instruments, Worcester, MA) in conjunction with automatic image analysis. SEM studies were performed on cryogenically fractured surfaces of blend specimens, which were etched for 24 h at room temperature by cyclohexane. The surfaces was sputtered with gold before viewing. For each blend sample, three micrographs were analyzed and the number-average diameter of particles was calculated.

TABLE IV
Characterization of Compatibilized Blends (Systems c and d)

Sample code	Compatibilizing agent composition (wt %)			TR^a	d (μm)	Interfacial tension (γ_{12}) (mN/m)	Steady-state torque (SST)^b
	DCP	EMA-I	TMPTMA				
System c							
19	0.02	2	—	1.06	0.54	0.1952	15.9
20	0.04	2	—	1.14	0.53	0.1734	15.8
21	0.06	2	—	1.15	0.67	0.2214	15.7
22	0.02	4	—	1.07	0.19	0.069	18.9
23	0.04	4	—	1.15	0.3	0.1288	17.2
24	0.06	4	—	1.17	0.49	0.1593	16
25	0.02	6	—	1.04	0.16	0.06196	19.1
26	0.04	6	—	1.1	0.29	0.1056	17.8
27	0.06	6	—	1.12	0.52	0.1831	16.1
28	0.02	8	—	1.1	0.17	0.061	18.4
29	0.04	8	—	1.23	0.28	0.0887	17.7
30	0.06	8	—	1.22	0.56	0.1764	16.8
31	0.02	10	—	1.16	0.21	0.0715	18.2
32	0.04	10	—	1.15	0.3	0.0977	18
33	0.06	10	—	1.27	0.58	0.1757	15.5
System d							
34	0.02	—	0.02	1.1	0.58	0.378	16.2
35	0.04	—	0.2	1.18	0.48	0.4103	15.7
36	0.06	—	0.2	1.24	0.53	0.1637	15.5
37	0.02	—	0.4	0.91	0.31	0.4895	18.7
38	0.04	—	0.4	1.03	0.26	0.2167	17.5
39	0.06	—	0.4	1.17	0.32	0.1637	16.6
40	0.02	—	0.6	0.92	0.46	0.1456	18.6
41	0.04	—	0.6	1.02	0.34	0.10563	17.8
42	0.06	—	0.6	1.15	0.29	0.1092	16.6

^{a} Ratio of steady-state torque of EPDM to PP at 10 min mixing, 60 rpm, 190°C, and with the same content of compatibilizing agent as that of the blend.
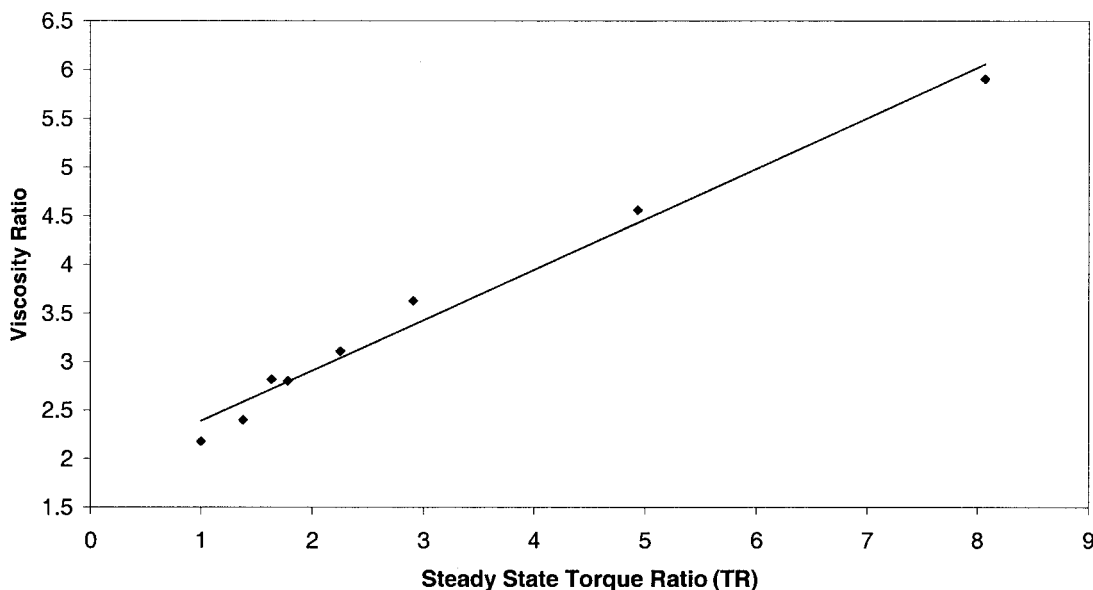^{b} Steady-state torque of blend at 10 min mixing, 60 rpm 190°C.

Figure 1 Viscosity ratio (η_d/η_m) versus torque ratio (TR).

RESULTS AND DISCUSSION

Simple blends

To analyze our experimental results, a relation similar to that suggested by Wu[1] for the twin-screw extruder was used for the samples blended in the internal mixer in the following form:

$$\frac{d\eta_m\dot{\gamma}}{\gamma_{12}} \approx (\eta_d/\eta_m)^n \qquad (4)$$

It has been reported[26,27] that the steady-state TR in an internal mixer can be correlated to the viscosity ratio. To examine this relation for the simple blends under study, η_m and η_d for PP and EPDM samples were measured using a capillary rheometer (Instron 3211) at the same temperature (190°C) and shear rate as those used for blending of the sample blends in the internal mixer. A simple relation based on rotor geometry and speed was used to calculate the mixing shear rate as[27]

$$\dot{\gamma} = \frac{2\pi N}{\ln(R_e/R_i)} \qquad (5)$$

where N is the rotor speed and R_i and R_e are rotor radius (bob radius) and external radius (cup radius), respectively. A linear relation between viscosity ratio (η_d/η_m) and torque ratio (TR), shown in Figure 1 blends, indicates that η_d/η_m in eq. (4) can be replaced by TR, leading to the following form:

$$\frac{d\eta_m\dot{\gamma}}{\gamma_{12}} \approx (TR)^n \qquad (6)$$

Because $\dot{\gamma}$ and γ_{12} are the same for all the simple blends, a plot of $(d\eta_m)$ versus torque ratio can be used to calculate exponent n in eq. (6).

Figures 2 and 3 represent the number-average particle diameter (d) and ($d\eta_m$) versus TR, respectively.

Results shown in Figure 2 indicate that the EPDM particle size increases from 0.83 to 5.2 μm when the TR increases from 1 to 8. Figure 3 shows an exponential relation between ($d\eta_m$) and TR, from the curve fitting of the experimental data; the exponent n was found to be about 0.5141. By substituting $n = 0.5141$, eq. (4) can be rewritten in the following form:

$$\frac{d\eta_m\dot{\gamma}}{\gamma_{12}} \approx (TR)^{0.5141} \qquad (7)$$

Compatibilized blends

System a

Figure 4 shows that for these blends d decreases with increasing EMA–I content, reaching its minimum at about 6 wt % of ionomer, above which it remains constant. EMA–ionomers have been used as compatibilizing agents in many binary blends, even though the role of ionomers in these systems is not fully understood.[28–33] Some authors[31–33] believe that a physical interlocking could be occurring between blend components attributed to ionic crosslinking characteristics of ionomers. Kim et al.[32,33] showed that Na-neutralized EMA–ionomers can act as good compatibilizing agents for PP/EPDM (50/50 wt %) blends, through physical crosslinking.
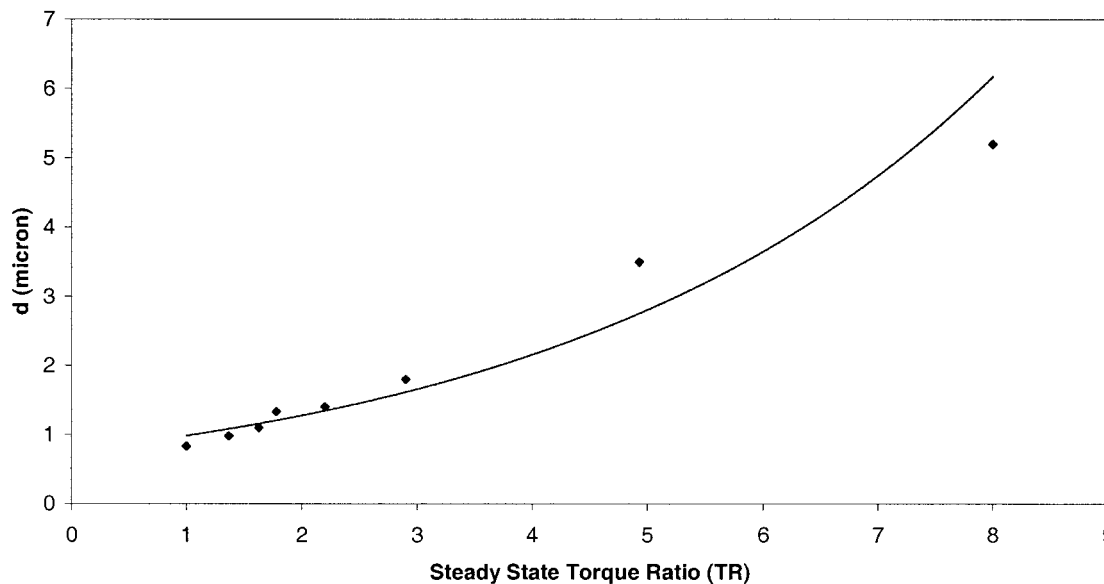
Figure 2 Number-average diameter of EPDM dispersed phase (micron) versus TR.

When a compatibilizer is added to a binary blend, it concentrates at the interface and induces chemical or physical interactions. These interactions reduce interfacial tension and blends with a smaller dispersed phase are obtained.[3] The size of the dispersed phase decreases as the concentration of compatibilizer increases, until the interface between the two phases is saturated. At this point an equilibrium particle size is observed. However, as the concentration of the compatibilizer reaches its saturation point, it can no longer act as an interfacial agent.[3] Thus the results shown in Figure 4 can be related to the compatibilizing action of

EMA–ionomer for PP/EPDM (80/20 wt %) during the melt–melt blending process. From the results shown in Figure 4, one may thus conclude that EMA–I up to 6 wt % acts as a compatibilizer, whereas with further increases of its content it can no longer affect the rubber particle size.

System b

As shown in Figure 5, d decreases with increasing DCP content, reaching its minimum at the DCP concentration between 0.02 and 0.04 wt %, above which it begins to
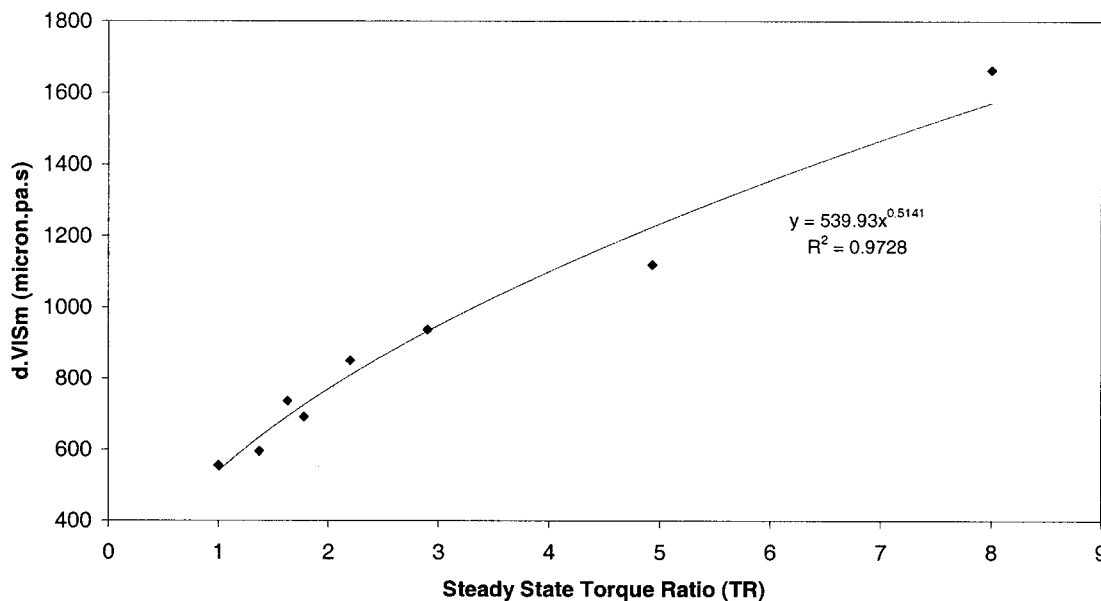

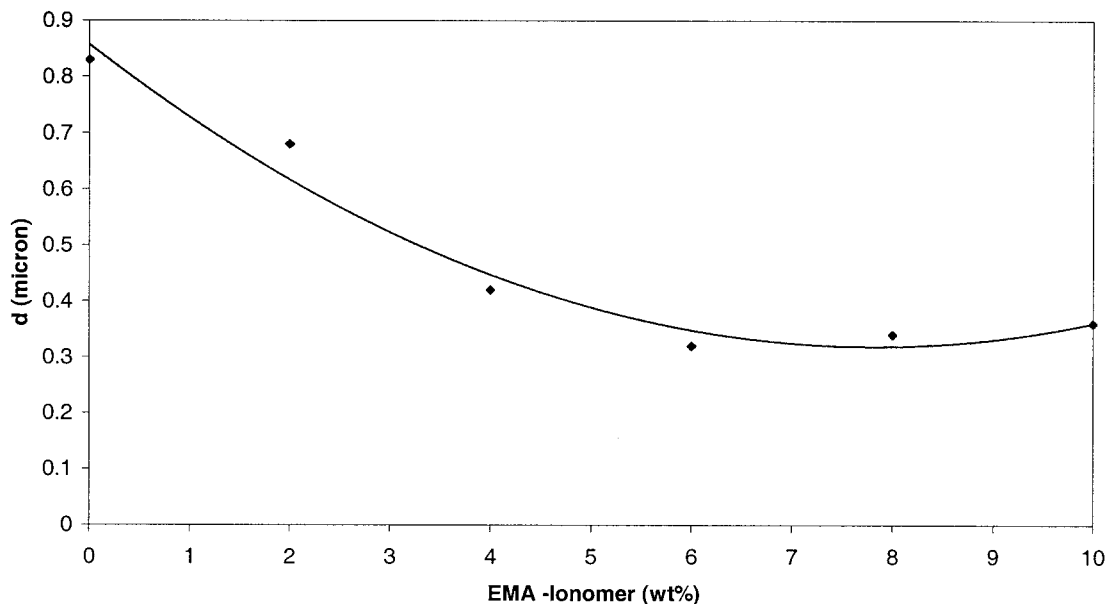**Figure 3** ($d\eta_m$) versus torque ratio (TR).

Figure 4 Number-average diameter of EPDM dispersed phase (micron) versus EMA–ionomer (wt %).

increase to a value even higher than that observed for the simple blend. Dicumyl peroxide is a well known cross-linking agent that has been used to compatibilize blends containing one crosslinkable constituent, through dynamic vulcanization.[12,13] However, when both blend components have the ability to react with the peroxide *in situ*-formed copolymers that are to be interposed at the interface, there is an improvement in the interfacial interaction.[13]

On the other hand, in the case of PP/rubber blends, melt mixing in the presence of DCP can lead to PP chain scission as a competitive reaction with the inter-copolymer formation,[34] giving rise to the PP melt viscosity reduction and thus increasing the TR values, as shown in Tables III and IV.

Thus, from these results it can be suggested that DCP, up to a concentration of 0.04 wt %, acts as an interfacial modification agent, whereas at higher con-
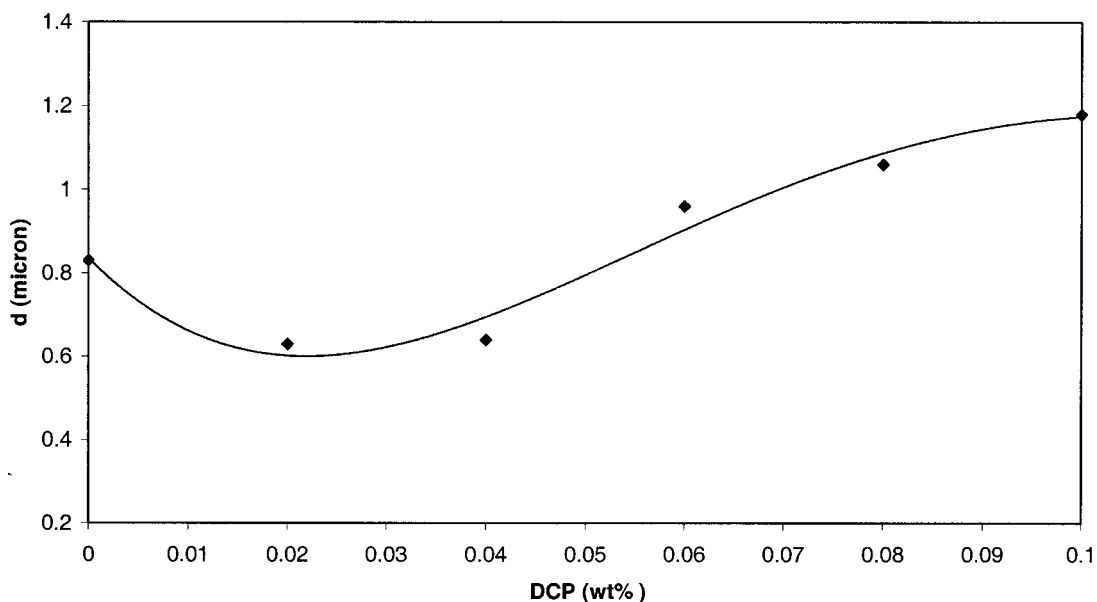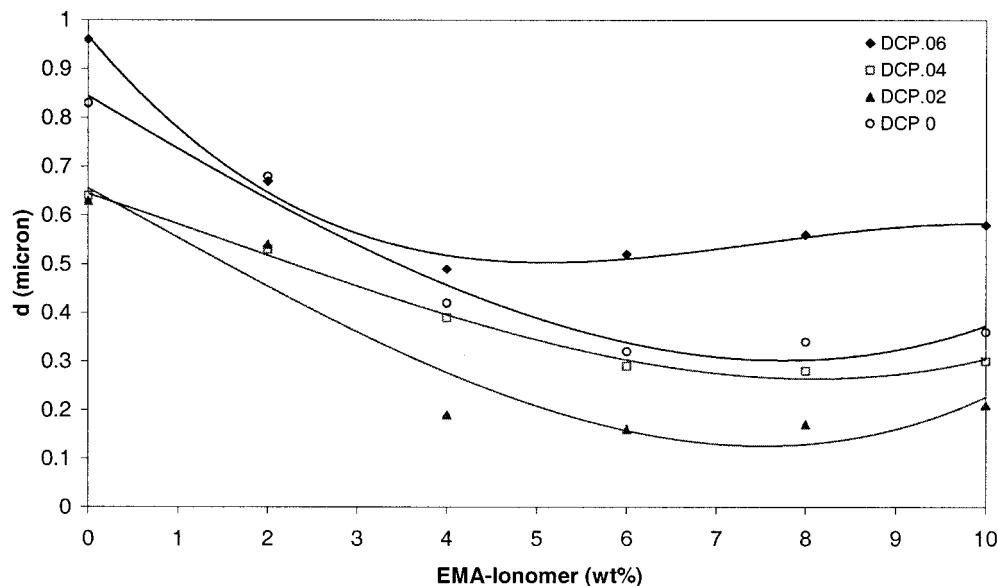

Figure 5 Number-average diameter of EPDM dispersed phase (micron) versus DCP (wt %).

Figure 6 Number-average diameter of EPDM dispersed phase (micron) versus EMA–ionomer (wt %) at various contents of DCP (wt %).

centrations it produces an increase in the torque ratio, which according to eq. (7) will have an adverse effect on controlling the rubber particle size.

System c

As shown in Figure 6 for these blend systems at constant DCP content, d first decreases with EMA–I content and remains unchanged at a concentration range of 6–10 wt % of EMA–I, exhibiting almost the same trend as that obtained for the blends containing only EMA–I. However, it can be seen that for the range of concentration of EMA–I under study the blends containing 0.02 and 0.06 wt % of DCP show the smallest and largest particle size, respectively. This can be explained by the same manner as that for the blends containing only DCP. Moreover, it is interesting to note that minimum particle size is obtained for the blends containing a combination of 0.02 wt % of DCP and 6 wt % EMA–I, suggesting a synergistic effect for these interfacial agents.

System d

The effect of incorporation of TMPTMA into system b as a coagent is illustrated in Figure 7. As mentioned before, in the presence of a high concentration of peroxide in the blends, chain scission becomes the dominant reaction for PP. However, when a multifunctional monomer (such as TMPTMA) is used as coagent, the rate of chain session reactions will be reduced because of stabilization of macroradicals through resonance. Therefore crosslinking reactions

are favored because the steric hindrance is resolved.[34-36] Thus, it is expected that the DCP–TMPTMA system has less effect on changing the TR of EPDM/PP blends than that of DCP alone. This idea is supported by the results given in Tables III and IV. This is the reason that the number-average diameter of EPDM is smaller for these blends compared to that of system b. Figure 7 shows that the optimum content of TMPTMA is 0.04 wt %.

Surface tension estimation of compatibilized blends

If we assume that eq. (7) is applicable for the compatibilized blends and that rewriting this equation for simple blend 1 as a reference in the form

$$\frac{d_1 \eta_{m1} \dot{\gamma}}{(\gamma_{12})_1} = (TR_1)^{0.5141} \qquad (8)$$

then eq. (9) can be derived by dividing eq. (7) into eq. (8), which can be used to estimate γ_{12} of the compatibilized blends,

$$\gamma_{12} = (\gamma_{12})_1 \times \frac{d}{d_1} \times \frac{\eta_m}{\eta_{m1}} \times \left(\frac{TR_1}{TR}\right)^{0.5141} \qquad (9)$$

where index 1 denotes the simple blend characteristic data.

Thus if $(\gamma_{12})_1$ is known, then γ_{12} can be calculated from eq. (9) using η_m, d, and TR given in Tables III and
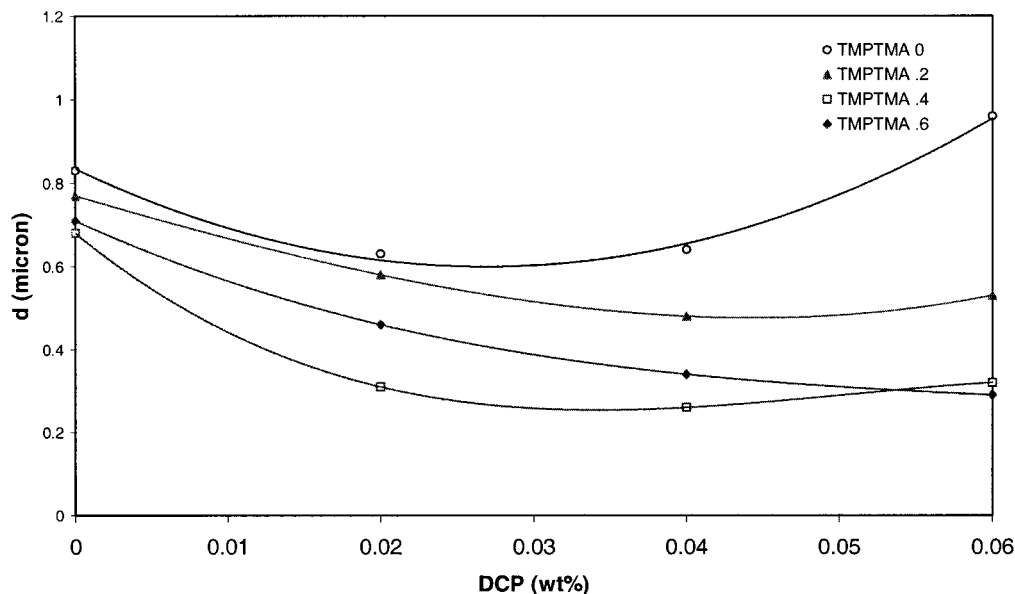
Figure 7 Number-average diameter of EPDM dispersed phase (micron) versus DCP (wt %) at various contents of TMPTMA (wt %).

IV for compatibilized blends and η_{m1}, d_1, and TR$_1$ given in Table II for simple blends.

To calculate $(\gamma_{12})_1$, the surface tension values of PP and PE at 190°C were calculated using the surface tension values reported[37] at 180°C in conjunction with the variations of surface tensions with temperature $[-(d\gamma/dT)]$[and polarities $[x_p = (\gamma_p/\gamma)]$, which is the ratio of the polar part of surface tension (γ_p) to γ. The surface tension of EPDM was calculated by means of the simple mixture rule based on surface tension of polyethylene norbornene (PENB), PE, and PP. Given the low ENB content in EPDM1, the surface tension of PENB was assumed to be the same as that for polybutadiene. Surface tension γ, its dispersive contribution γ_d, and its polar contribution γ_p for PP1, EPDM1 at 190°C are listed in Table V.

Thus, the interfacial tension between PP1 and EPDM1 in a simple blend can be calculated from the well-known harmonic mean equation[37] as

$$(\gamma_{12})_1 = \gamma_1 + \gamma_2 - \frac{4\gamma_{1p}\gamma_{2p}}{\gamma_{1p} + \gamma_{2p}} - \frac{4\gamma_{1d}\gamma_{2d}}{\gamma_{1d} + \gamma_{2d}} \quad (10)$$

TABLE V
Estimated Surface Tension of Polymers at Mixing Temperature of 190°C

Polymer	γ (190°C) (mN/m)	γ_p (190°C) (mN/m)	γ_d (190°C) (mN/m)
PP1	20.22	0.4	19.81
EPDM1	23.15	0.21	22.94

The value of $(\gamma_{12})_1$ at 190°C, obtained from eq. (10), was found to be about 0.32 mN/m. By substituting the value of $(\gamma_{12})_1$ in eq. (9), the interfacial tension (γ_{12}) of the compatibilized blends (listed in Tables III and IV) can be estimated.

Relationship between viscosity and interfacial tension of the blends

The SST values measured as a function of interfacial agents' content for systems a and b are shown in Figure 8, which shows that the SST increases with increasing the EMA–I content, reaching to a minimum, above which it begins to decrease. However, the extent of the variation in the SST of the blends varies depending on the DCP content. The maximum SST is obtained for the blend containing 6 wt % EMA–I and 0.02 wt % DCP, whereas the minimum SST is for the blend containing 0 wt % of EMA–I and 0.06 wt % DCP.

Figure 9 also shows that for the blend systems c and d the SST decreases with increasing DCP content, but increases with increasing TMPTMA concentration.

The SST of a blend is a measure of viscosity[26,27] and thus can be related to interfacial interaction between the phases. When there is a strong interfacial interaction (i.e., low interfacial tension between phases), a positive deviation from the mixture low is expected for the blend viscosity.[32,36] Figure 10 shows the variation of SST of the blends with estimated interfacial tension. As can be noticed, the data are scattered and therefore it is hard to define a logical relationship between SST and predicted interfacial tension values.
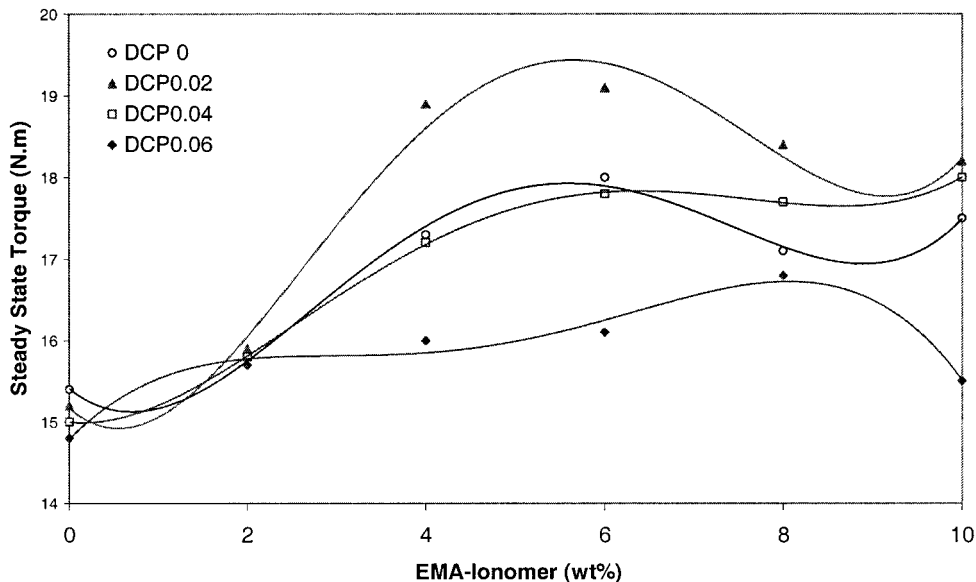
Figure 8 Steady-state torque ratio of the blends versus EMA–ionomer content (wt %) at various contents of DCP (wt %).

However, as discussed earlier, the compatibilizing systems used in these blends, apart from increasing the interfacial interaction between the phases, can influence the SST of the components through different mechanisms. To eliminate this effect, the SST of the blends was calculated on the basis of mixture law (SSTM) but using the SST of each component containing the same content of compatibilizing system. Then the difference between these values and corresponding SST measured for the blends was considered as a measure of positive deviation of viscosity of the blends. The results obtained from this procedure are presented in Figure 11, which can be used as a more realistic guide to examine the correlation between viscosity of the blends and the interfacial tension between the two phases. These results show that there is a certain amount of interfacial tension below which the extent of positive deviation of viscosity sharply increases with decreasing the interfacial tension (i.e., increasing the interfacial interaction). This is in accordance with the reported theoretical predictions.[32,36]

CONCLUSIONS

A power law relation between measured rubber particle size and the torque ratio was obtained for the
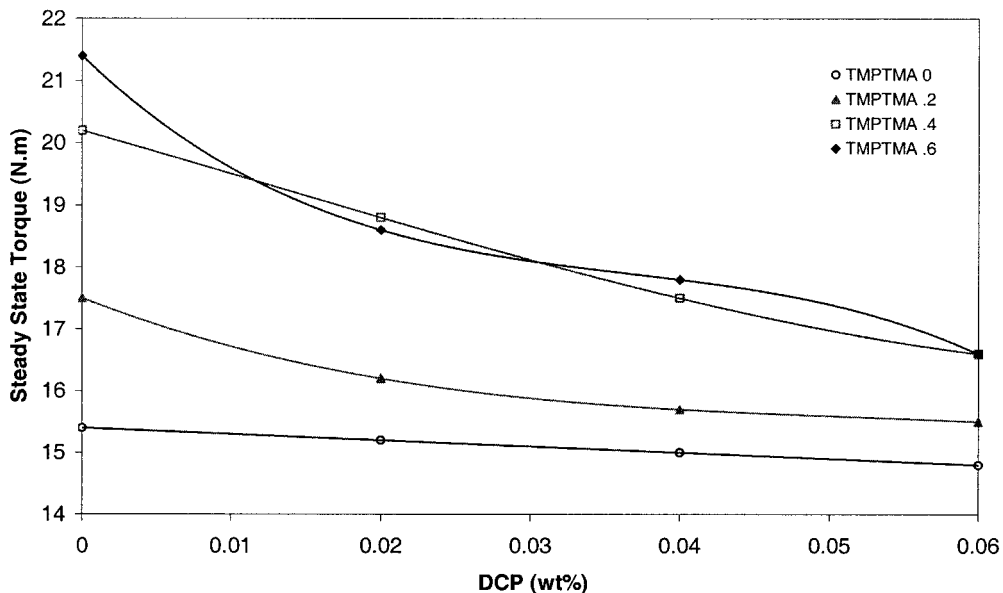

Figure 9 Steady-state torque ratio of the blends versus DCP content (wt %) at various contents of TMPTMA (wt %).
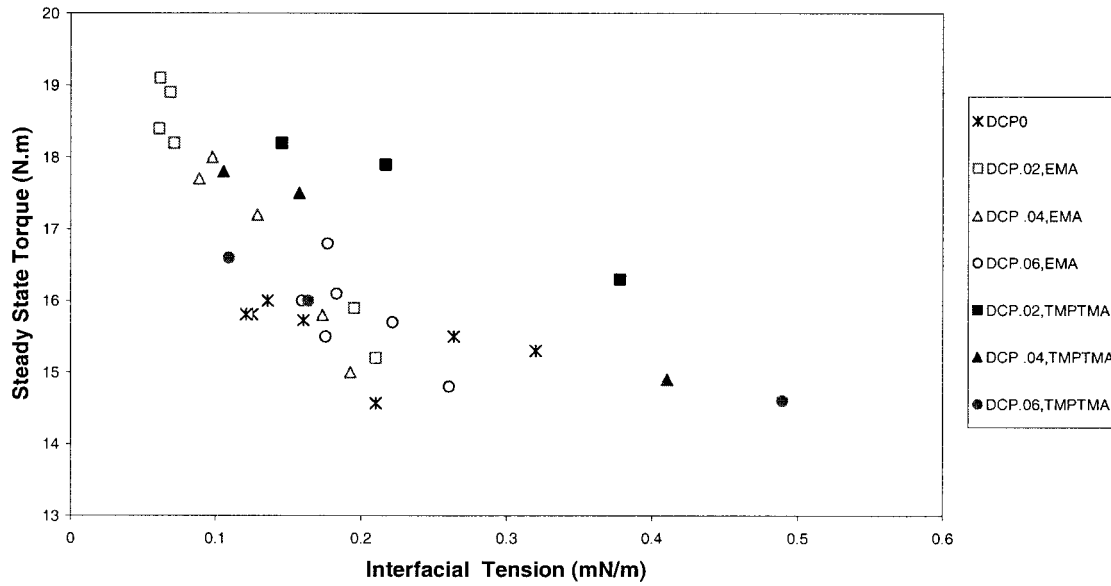
Figure 10   Steady-state torque of the blends (N·m) versus estimated surface tension (mN/m).

PP/EPDM (80/20) simple blends in an internal mixer. It was demonstrated that such a relationship can be used for PP/EPDM compatibilized blend systems to estimate the interfacial tension between the two phases.

The experimental results showed that for different compatibilizing systems used in this study, there is an optimum concentration of interfacial agents above which they can no longer enhance the interfacial interaction between two phases. It was also shown that a compatibilizing system consisting of EMA–I and DCP has a stronger, more influential effect compared to that of the other systems. The most compatibilizing effect and therefore the smallest particle size was obtained for the blends containing 6 wt % of EMA–I and 0.02 wt % of DCP.

A small amount of DCP combined with interfacial agents enhanced the compatibilizing efficiency in the blends. However, using a higher concentration of DCP had an adverse effect because it accelerated the degradation of PP, thus leading to an increase in viscosity ratio and increased rubber particle size in the blends. This was the reason that particle size in the blends containing only DCP was larger.
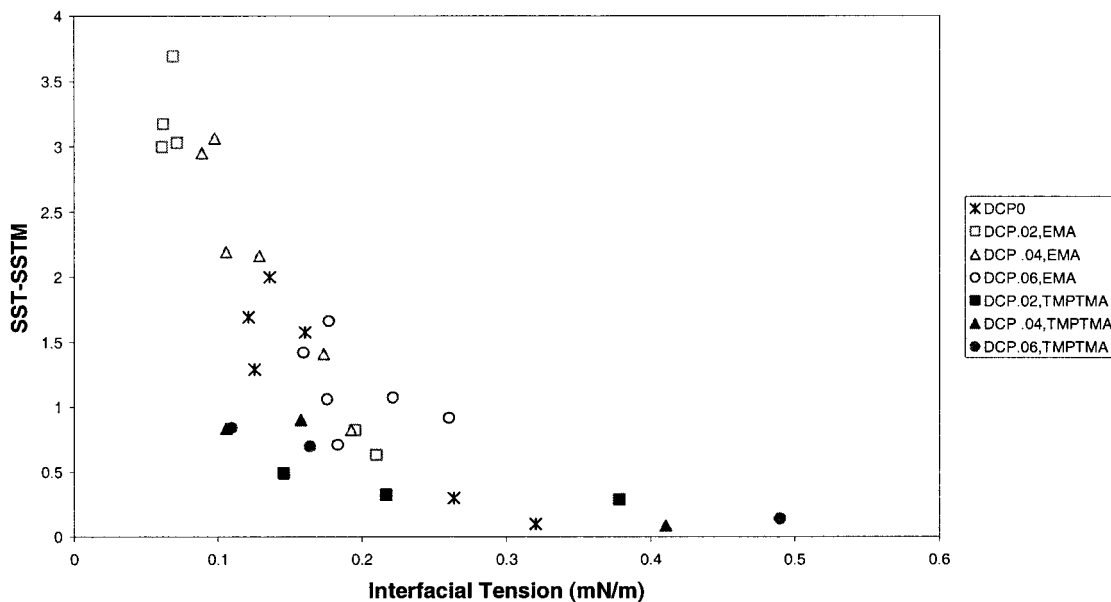

Figure 11   Difference between steady-state torque of the blends and steady-state torque from the mixture law (N·m) versus estimated surface tension (mN/m).

Using TMPTMA as a multifunctional coagent in combination with DCP decreases the PP degradation, resulting in smaller particle size in the blends.

Finally, when the amount of interfacial interaction exceeds a certain value, there was a sharp increase in the extent of positive deviation of blend viscosity.

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