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THERMAL AND DYNAMIC MECHANICAL PROPERTIES OF PES/PPS BLENDS

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Abstract

Blends of poly(ether-sulfone) (PES) and poly(phenylene sulfide) (PPS) with various compositions were prepared using an internal mixer at 290°C and 50 rpm for 10 min. The thermal and dynamic mechanical properties of PES/PPS blends have been investigated by means of DSC and DMA. The blends showed two glass transition temperatures corresponding to PPS-rich and PES-rich phases. Both of them decreased obviously for the blends with PES matrix. On the other hand, T_g of PPS and PES phase decreased a little when PPS is the continuous phase. In the blends quenched from molten state the cold crystallization temperature of PPS was detected in the blends of PES/PPS with mass ratio 50/50 and 60/40. The melting point, crystallization temperature and the crystallinity of blended PPS were nearly unaffected when the mass ratio of PES was less than 60%, however, when the amount of PES is over 60% in the blends, the crystallization of PPS chains was hindered. The thermal and the dynamic mechanical properties of the PPS/PES blends were mainly controlled by the continued phase.

Keywords: blend, crystallization, poly(ether sulfone), poly(phenylene sulfide)

Introduction

The research in the area of polymeric blends, alloys, and composites has been very active in both academic and industrial fields. Polymer blends have now come to the fore as such a remarkable and major endeavor. Their current and potential technological importance is their ubiquitous presence in consumer products is testimony to their commercial importance [1].

Poly(phenylene sulfide) (PPS) is an important high performance engineering thermoplastic with wide applications in molding resins, fibers and matrices for thermoplastic composites. However, the resin has insufficient toughness and is brittle, which limits its application. In recent years, a great effort has been made to improve the above properties, hence, blending and block copolymerization are necessary [2–8].

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Poly(ether-sulfone) (PES) is an amorphous high-performance engineering plastic with good heat resistance and mechanical properties. So, blending PPS with PES is an important way to improve the toughness and impact strength of PPS resin.

Several studies have been reported on the thermal and mechanical properties [9, 10], melting behavior and crystallization of PPS/PES blend [11–12], but the conclusions should be discussed in detail. The behavior of the glass transition temperature on blending has been widely used as a criterion of miscibility in the polymer blends. The cooperative conformational rearrangements in a miscible polymer blend should involve the motion of polymer segments pertaining to both components of the blend. This also produces a single main dynamic mechanical or dielectric relaxation process with relaxation times that depend on the cooperation of the blend. Provided the glass transition temperatures shift inward comparing with that of the pure components, the polymer blend is partly miscible. Whereas in the polymer blends that are immiscible the individual relaxation processes of both phases can be observed experimentally, when the difference in the relaxation times corresponding to each component are high enough. Nevertheless, different experimental techniques can be sensitive to molecular motions in different characteristic length scales.

In general, miscibility and phase behavior of polymer blends were studied by using differential scanning calorimetry (DSC) [13, 14]. The DSC limits the resolution of the T_gs because of a lack of sensitivity, as pointed out previously by Brostow *et al*. [15, 16] for studying phase diagrams in polymer liquid crystal systems and by Aoki *et al*. [17] for investigating the miscibility of poly(ethylene terephthalate) and poly(ethylene-2,6-naphthalate) blends. Some contradictory results [18] are reported in the literature. Dynamic mechanical and dielectric spectroscopy are able to detect the microheterogeneity or localized concentration fluctuations of the blends in a small length scale. Certainly, nuclear magnetic resonance spectroscopy (NMR) seems to be even more sensitive to investigate molecular motion in polymer blends. Sometimes, we have to study the miscibility of polymer blends by means of a combination of two or more methods in order to confirm the experimental results.

Although several studies related to the characteristics of PPS/PES blends have been reported in the literature, no detailed reports have been seen on the relation between the dynamic mechanical properties and miscibility of PPS/PES blends. However, this technique is considered to be very sensitive to measure the physical properties of the glass transition region.

Shibata *et al*. [11], Ruan *et al*. [9], Zhang *et al*. [10] and Zeng *et al*. [12] have concluded that PPS and PES blends are partly miscible. The glass transition temperatures of PPS rich-phase and PES rich-phase shifted inward in the blends compared with one of the pure components and the dependence of the glass transition temperature on the composition in the blends was found. Also, Shibata *et al*. reported that the heat of crystallization (H_c) normalized to the pure PPS content decreased slightly with increasing PES content in the blends, indicating that the degree of crystallinity of PPS was lowered a little.

In this article some results that are different from those reported in the literatures as mentioned above are presented. The characteristic of the molecular motion and the factors affected the phase behaviors of the blends were discussed.

Experimental

Materials

PES powder was purchased from Xinghua Chemical Plant of Jilin University (Changchun, China). The inherent viscosity was 380 mL g^{-1} (293 K). PPS used was a commercial product (Model p-3) manufactured by Sichuan Factory of High Performance Engineering Plastics. All PES and PPS were dried at 90°C for 12 h in a vacuum oven to remove the absorbed water.

Samples preparation

The PES and PPS powders was melt blended in a Brabender internal mixer at 290°C, 50 rpm for 10 min. The plain PPS and PES used for comparison was also subjected to the same treatment. The torque is 5.6 Nm for neat PES and 0.4 Nm for pure PPS, respectively. The blends were cooled at room temperature, and then stored in a desiccator until testing.

Thermal analysis

Thermal behavior of the samples was measured with a Perkin Elmer DSC-7 differential scanning calorimeter under N_2 atmosphere in the temperature range of 50 to 300°C. The mass of samples was around 10 mg. The heating and cooling scans were always carried out at a rate of 10°C min^{-1}. The samples were firstly heated from room temperature to 300°C. After holding the specimens at 300°C for 5 min to destroy the crystalline nuclei completely, the cooling traces were recorded from 300 to 50°C, and then the second heating scan was recorded.

Dynamic mechanical measurements

All samples for the dynamic mechanical measurements were prepared by compression molding at 290°C to about 1 mm in thickness. The quench sheet was made by quickly transferring from hot press to ice water under pressure as soon as possible. All the films were cut into size about 20×5 mm, and the edges of each film were carefully smoothed using fine sandpaper. A Netzsch dynamic mechanical analyzer DMA-242 (Germany) was used to determine storage modulus E', loss modulus E'', and loss tangent delta (tanδ) in a temperature range from 20 to 240°C. All measurements were carried out under the N_2 atmosphere at a frequency of 3 Hz and heating rate of 3°C min^{-1}.

Results and discussion

Thermal behavior of PPS/PES blends

Figure 1 shows the DSC cooling and second heating curves of PPS/PES blends at 10°C min^{-1}. All parameters down from the thermal analysis curves, T_g, T_c and T_m for PPS and PPS/PES blends are summarized in Table 1. The detectable glass transition temperature related to PPS was not observed for all the blends because of its semicrystalline property, while the T_g of PES in blends were lower than that of neat PES and was affected a little by the composition.

For the blends with PPS matrix, the peak temperature of melting and crystallization and the heat of crystallization normalized to the PPS mass fraction were nearly unaffected by the PPS composition. However, the degree of crystallinity of PPS in these blends was lower than that of neat PPS. On the other hand, the blends showed lower melting and crystallization temperature when PPS content was less than 40%, and the degree of crystallinity also decreased obviously.

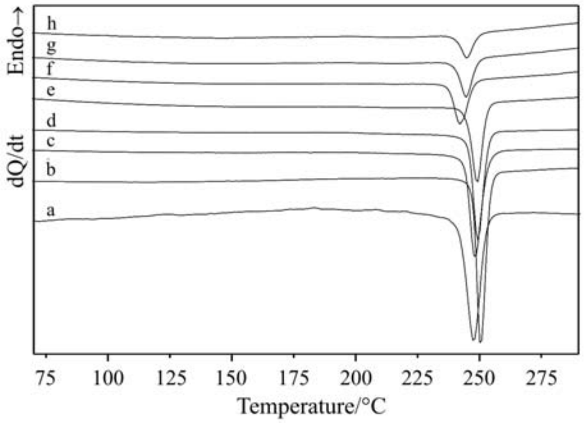

Fig. 1a The DSC trace of PPS/PES blends from the melt at a cooling rate of 10°C min^{-1}, PPS/PES mass ratio: a – 100/0; b – 80/20; c – 70/30; d – 60/40; e – 50/50; f – 40/60; g – 30/70; h – 20/80

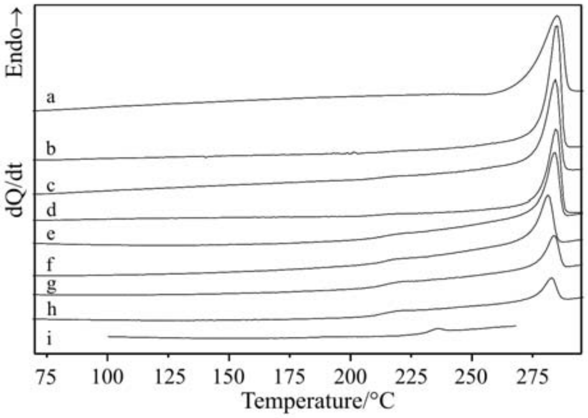

Fig. 1b The second heating scan of PPS/PES blends, PPS/PES mass ratio: a – 100/0; b – 80/20; c – 70/30; d – 60/40; e – 50/50; f – 40/60; g – 30/70; h – 20/80; i – 0/100

Table 1 Thermal transition parameters of the blends of PPS and PES measured by DSC

Sample (PPS/PES) mass ratio	$T_{g,PPS}$/°C	$T_{g,PES}$/°C	T_c/°C	T_m/°C	ΔH_c/J g^{-1}	$X_{d,c}$[a]
100/0	85	–	248.3	285.0	57.7	0.72
80/20	–	–	250.5	284.8	52.36	0.65
70/30	–	–	249.2	284.2	51.1	0.64
60/40	–	–	250.4	284.6	52.0	0.65
50/50	–	215.4	250.1	284.0	53.8	0.67
40/60	–	213.1	243.2	281.3	42.6	0.53
30/70	–	213.2	246.3	283.7	44.4	0.55
20/80	–	212.8	245.9	282.7	40.3	0.5
0/100	–	225.1	–	–	–	–

[a] $X_{d,e}$: The degree of crystallinity of PPS was calculated according to following equation: $X_c=\Delta H_c/\Delta H_c^0$. The heat of crystallization ΔH_c^0, for 100% crystalline PPS, 80 J g^{-1}, was extrapolated from the data of Brady [23].

These results indicated that although the crystallization of PPS was inhibited by the existence of PES, there was no sign of strong interaction between the PPS and PES macromolecules. The T_g of PES and the degree of crystallinity of PPS in blends were lower than that of bulk polymers, a clear shift of all the thermal parameters was observed between the blends with PPS matrix and with PES matrix. Obviously, the thermal properties of the blends were mainly controlled by the continuous phase. It could be postulated that the crystallization process of PPS domains should be restrained by the PES continuous phase remarkably. The crystallization temperature of PPS from melt, T_c, decreased to about 251°C that is close to the glass transition temperature of the PES rich-phase, 225°C. Then the PES continuous phase with high viscosity inhibited the crystallization of PPS phase. On the other hand, for the blends with PPS continuous phase, the PES domains were separated, even though the macromolecules of PES exhibit high viscosity, they could hardly restrain the mobility and the crystallization of PPS macromolecules.

Dynamic mechanical properties of PPS/PES blends

Dynamic mechanical analysis was an effective means to study the transition and relaxation behavior of polymer systems. The DMA curves illustrating the temperature dependence of tanδ and storage modulus E' of PPS/PES blends cooled at room temperature from melt were shown in Fig. 2, while Fig. 3 presented the DMA spectra of quenched samples. The dynamic mechanical analysis data were listed in Table 2. The phase structure of a blend is referred by the number of glass transition temperatures observed in the dynamic mechanical spectra. That is, the appearances of two glass transitions are a clear evidence of phase separation and that of a single glass transition at a temperature intermediate between those of the pure components indicates miscibility and the glass transition temperatures shift inward provided the components in the blend are partial miscible. Clearly, two glass transition temperatures, $T_{g,\ PPS}$ and $T_{g,\ PES}$, were observed in the spectra of Figs 2 and 3. The broad dynamic mechanical damping peak at about 100°C is due to the glass transition of PPS and the sharp peak near 220°C is associated with the T_g relaxation of PES component in the blends. The presence of two peaks in the blends confirms that the blends consist of two phases.

The blends exhibited two plateaus of storage modulus *E*' before the T_g of PES changing with the composition. For the unquenched samples, the value of first *E*' plateau decreased with the increasing PES content as presented in Fig. 2a while Fig. 3a showed totally opposite trend concerning the quenched samples. And the

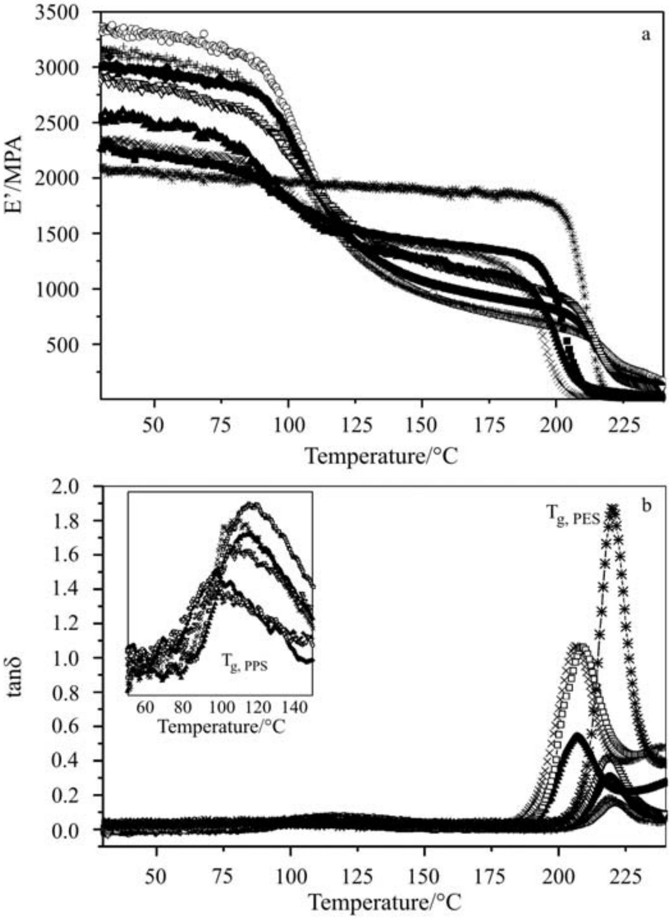

Fig. 2 a – storage modulus *E*' and b – loss tangent tanδ as a function of temperature for the unquenched PPS/PES blends, PPS/PES mass ration: ○ – 80/20, + – 70/30, □ – 60/40, ▽ – 50/50, ▲ – 40/60, × – 30/70, ▪ – 20/80, ∗ – 0/100

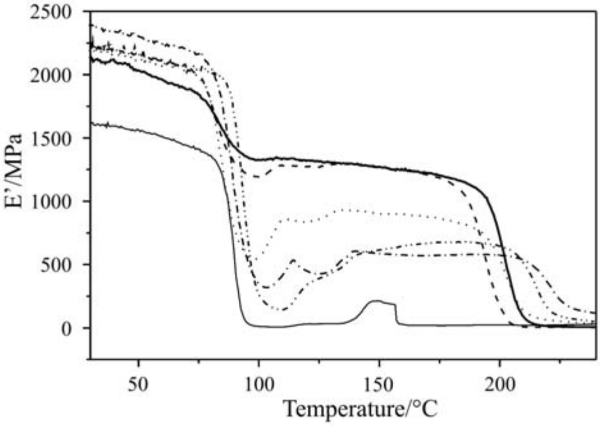

Fig. 3a Storage modulus *E*' as a function of temperature for the quenched PPS/PES blends, PPS/PES mass ratio: — – 100/0, –·– – 60/40, –··– – 50/50, ··· – 40/60, --- – 30/70, — – 20/80

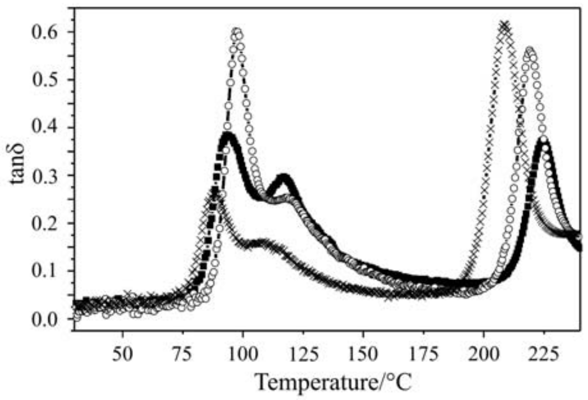

Fig. 3b Loss tangent tanδ as a function of temperature for the quenched PPS/PES blends, PPS/PES mass ration: ▪ – 60/40, ○ – 50/50, × – 40/60

value of the second *E*' plateau increased with the increasing of PES content in both quenched and unquenched samples. The unquenched PPS had higher glass modulus than PES resulting from its semicrystalline property, whereas the storage modulus of quenched PPS was much lower because of its amorphous properties and its bigger fractional free volume created during the quenching.

For all quenched samples with PPS ingredient, when the temperature was higher than its glass transition temperature, the PPS began to crystallize at about 110°C by showing a rapid increase in *E*'. The cold crystallization led the PPS to show a second peak of tanδ after T_g even though the PPS mass fraction was 20% only.

The T_g values (peak temperature of tanδ curves) of all samples were summarized in Table 2. Unlike Shibata [11], who reported that the $T_{g,\text{ PPS}}$ increased with increasing

Table 2 T_g Values of PPS/PES blends measured by DMA

Sample PPS/PES mass ratio	T_{g_1}/°C	T_{g_2}/°C	Sample quenched PPS/PES mass ratio	T_{g_1}/°C	T_{g_2}/°C
100/0	118.9	–	100/0	95.4	–
80/20	116.3	221.1	80/20	94.0	217.2
70/30	109.5	219.2	70/30	93.7	222.6
60/40	114.9	219.2	60/40	94.1	224.6
50/50	110.4	219.1	50/50	97.6	219.4
40/60	97.6	207.1	40/60	88.2	208.8
30/70	98.2	206.5	30/70	83.6	209.3
20/80	98.3	208.2	20/80	85.1	212.0
0/100	–	225.4	–	–	–

PES content, and the $T_{g,\mathrm{PES}}$ decreased with increasing PPS content. First of all, PPS and PES are miscible partly in the PES rich-phase in the blends; however, they are immiscible in PPS rich-phase. In another words, some of PPS molecules can penetrate into PES phase in the blends but PES molecules cannot go into PPS phase at all. The similar results for high performance polymer blends were conducted in [19]. For example, the content of nylon 66 mixed into poly(ether imide) phase was very low in the nylon 66/poly(ether imide) blends. The poly(ether imide) rich-phase consists of 99% of poly(ether imide) and 1% of nylon 66 over a wide composition range of the blends. On the other hand, the amount of poly(ether imide) contained in nylon 66 rich-phases was much higher, about 4~9%, depending on the composition in the blends. The decreasing of T_g of PPS rich-phase instead of shifting inward as mentioned in [11] was observed in the blends. Second, the continue phase controls the properties of the blends, such as glass transition temperatures, and the dependence of T_g on the compositions of the blends was not founded. Thirdly, the decreasing of crystallinity of PPS phase with increasing the PES content monotonously in the blends was not founded. The results showed that PPS phase exhibited similar degree of crystallinity in the blends when the mass ratio of PPS component was 50~80%. On the other hand, the decreasing of the crystallinity of PPS phase was observed when the content of PPS was less than 40 mass% in the blends.

The data in Table 2 can be divided into two groups according to the T_g values, one was concerning the blends with PPS matrix, from neat PPS to PPS/PES (50/50, mass ratio), and the other was concerning the blends with PES matrix. Obviously, in the first group, the T_g of PES was hardly affected by the composition while the T_g of PPS decreased a little with the increase of PES content. However, the lowest T_g of PPS phase appeared at the composition of PPS/PES (70/30, mass ratio). After phase inversion, comparing with the blends with PPS matrix, PPS as the dispersed phase showed a much lower T_g that little changed with the increase of the PES content. Apparently the phase inversion had some effect on the thermal behavior of PPS/PES blends, the matrix phase dominated the molecular motion. It was also observed that because of different thermal history, $T_{g,\,\mathrm{PPS}}$ of the quenched samples are lower than that of unquenched samples. PPS with high degree of crystallinity made the mobility of the segments in amorphous regions to be severely restricted and there is much higher fractional free volume in the quenching PPS that led to decrement of the glass transition temperature of quenched PPS phase. The crystallization of PPS was hindered, accompanied with the drop of the degree of crystallinity for the blends with PES matrix, the diluted effect of the amorphous component weaken the restriction of crystallized PPS to the molecular motion, and resulted in lower of $T_{g,\,\mathrm{PPS}}$ and $T_{g,\,\mathrm{PES}}$ comparing with that of blends with PPS matrix. Based on the explanation mentioned above, no matter what the composition of the blends is, the value of T_g will be close whenever the degree of crystallinity is similar. This conclusion was in agreement with the results of the crystallinity of PPS in Table 1.

The characteristics of macromolecular motion in polymer blends are very complicated. If the molecules of different components mix on a molecular scale, the phase morphology, structure and interaction between components will have some effect on the macromolecular motion of the blend. Blending PES with PPS made the T_g of PES in all blends shifted to lower temperature, especially for the blends with PES matrix, and we concluded that some amorphous molecules of PPS had diffused into the PES phase as a plasticizer. On the assumption that this part of PPS/PES blend are miscible, according to Fox relationship:

$$\frac{1}{T_g}=\frac{m_1}{T_{\mathrm{gPPS}}}+\frac{1-m_1}{T_{\mathrm{gPES}}} \tag{1}$$

where m_1 is the mass ratio of PPS in the blends. When we took the average value of $T_{g,\,\mathrm{PES}}$ in blends measured by DMA as T_g, the mass fraction of PPS in PES phase was about 5.8%. In this experiment, the minimal mass fraction of PPS in blends was 20%, that was the reason why $T_{g,\,\mathrm{PES}}$ was nearly unaffected by the composition of blends.

It should be emphasized here that the glass transition temperature of PPS phase in the blends decreased to lower temperature instead of shifting inward comparing with the T_g of plain PPS. It indicated that the decrease of $T_{g,\,\mathrm{PPS}}$ resulted from the drop of degree of crystallinity in blends with PES component.

Some information can be obtained from the further analysis on data of the quenched PES/PPS blends. As showed in Table 2, the varying trend of T_g dependence on composition was same for both quenched and unquenched samples, that is, an obvious shift appeared at the phase inverse region. The point is if T_g was dominated by the degree of crystallinity as we discussed above, why the same shift trend occurred in the amorphous samples. It should be attributed to the influence of PES phase on the molecular motion of PPS. During quenching, the first PES phase was congealed into polymer glass, and then alone with the falling of temperature, PPS chains vitrified in the circumscription of PES glass, especially when PES being the matrix. It was supposed that the higher fractional free volume leaded to the decrease of $T_{g,\,\mathrm{PPS}}$.

The main mechanism governing the immiscibility, phase behavior and phase morphology development in the immiscible polymer blends is believed to be the result of both droplet break-up and coalescence. The minimum obtainable droplet diameter in immiscible blend system can be estimated from the critical capillary number:

$$(C_\mathrm{a})_\mathrm{crit} = \frac{\eta_\mathrm{m}\dot{\gamma}R}{\sigma_{12}} \tag{2}$$

where η_m represents the matrix viscosity, $\dot{\gamma}$ the shear rate, R the average droplet radius and σ_{12} the interfacial tension. If the capillary number (Ca) is larger than the critical capillary number $(Ca)_\mathrm{crit.}$, droplets can further deform and break-up. Coalescence is possible when the droplet size is less than the steady-state droplet size given by Eq. (2).

Taylor derived a function for the value of $(Ca)_\mathrm{crit.}$ in the case of Newtonian systems under simple shear flow [20]:

$$(C_\mathrm{a})_\mathrm{crit} = \frac{1}{2}\frac{16p+16}{19p+16} \tag{3}$$

where p is the viscosity ratio $\eta_\mathrm{d}/\eta_\mathrm{m}$, with η_d being the viscosity of the dispersed phase. Wu [21] has established an empirical equation fitting the capillary master curve:

$$2C_\mathrm{a} = \frac{\eta_\mathrm{m}\dot{\gamma}D_\mathrm{n}}{\sigma_{12}} = 4\left(\frac{\eta_\mathrm{d}}{\eta_\mathrm{m}}\right)^{\pm 0.84} \tag{4}$$

In this equation, the exponent is positive for $p>1$ and negative for $p<1$, D_n represents the number average particle diameter. Everaert [22] concluded that highly viscous matrices ($p<<1$) enhance droplet break-up owing to their efficient shear stress transfer towards the dispersed phase, while low viscous matrices ($p>>1$) often act as a lubricant for the dispersed phase reducing the droplet break-up. Because the apparent melting viscosity η_a of the polymer is directly proportion to its torque in the internal mixer we can confirm that the viscosity of PPS is much lower than that of PES and the ratio is about 0.0714, while the viscosity ratio is 14 when the continuous phase consists of PPS. During melt-mixing, the low viscous phase PPS acts as a lubricant to minimize the energy of mixing, and hence retards the liquefaction process of the soft PES phase at higher temperature. The component with lower flow temperature, PPS, will first encapsulate the component with higher flowing temperature, PES, to form the matrix phase, only if a substantial amount of the higher temperature flowing component has softened, gradual phase inversion can proceed.

In general, coalescence during melt-mixing will be governed by the interfacial mobility. A relatively high coalescence rate is encountered in polymer blends. However, the latter can be reduced if the matrix becomes highly viscous. As expected by Eqs (2)–(4), PES can inhibit the coalescence of PPS domains, then the opportunity of diffusion of PPS macromolecules into PES matrix increases, while the dispersed phase PES with higher melt viscosity penetrates hardly into PPS matrix with lower viscosity during melt mixing. The prediction drawn from the polymer mixing theory mentioned above is dealt with our experimental observations, i. e., the phase behavior of PPS/PES blends with PPS matrices ($p>>1$) is quite different from those with PES matrices ($p<<1$).

Conclusions

PPS/PES blends prepared by melt-mixing showed two glass transition temperatures corresponding to PPS-rich and PES-rich phase, which were hardly affected by the composition, indicating the poor miscibility between PPS and PES. For the blends with PES matrix, the melting temperature, the crystallization temperature and the normalized heat crystallization were lower than that of the blends with PPS matrix, revealing that the crystallization of PPS was hindered by PES. In DMA measurement, the samples with higher degree of crystallinity showed higher value of storage modulus *E*’ and glass transition temperature. Although compatibility between PES and PPS was poor, the glass transition of PES decreased after blending with PPS, which indicated that a little part of amorphous PPS had diffused into the PES phase.

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