

Morphology and Mechanical Properties of Immiscible Polyethylene/Polyamide12 Blends Prepared by High Shear Processing*

Wen-jin Yu, Shu-man Xu, Li Zhang and Qiang Fu**

College of Polymer Science and Engineering, State Key Laboratory of Polymer Materials Engineering,
Sichuan University, Chengdu 610065, China

Abstract In this work, completely immiscible polyethylene/polyamide12 (PE/PA12) blends were prepared by high shear extruder. The morphology and mechanical properties of the blends were investigated as a function of rotation speed. It was found that the high shear processing is an effective method to improve the dispersion of the PA12 phase in PE matrix when PA12 contents are 5 wt% and 10 wt%, and the dispersed phase particle size is reduced with the increase of rotation speed from 100 r/min to 500 r/min. However, with further increase of PA12 content to 20 wt%, high shear processing has no effect on the phase morphology of the blends. Accordingly, a largely increased elongation at break and impact strength are observed for PE/PA12/95/5 and PE/PA12/90/10 blends obtained at high rotation speeds but no effect on the property of PE/PA12/80/20. Annealing experiment demonstrated that the obtained phase morphology is not stable thus compatibilizer should be introduced in the future work. This work could provide a guideline for the application of high shear processing in the preparation of polymer blends with huge polarity difference.

Keywords Immiscible polyethylene/polyamide12 (PE/PA12) blends; High shear processing; Phase morphology

INTRODUCTION

Polymer blending is an efficient method to develop new materials with specifically desired properties^[1–3]. However, most of the polymer blends are immiscible and form a multiphase structure due to the great difference between chain structures^[4]. Consequently, apparent interphase boundary is obtained between the two components, which is a weak region and will result in relatively lower strength of blends compared with constituent bulk polymers. It is commonly established that the final properties of immiscible polymer blends with spherical morphology rely heavily on the particle size and size distribution of the dispersed phase as well as the interfacial interaction between two phases.

To improve the interfacial interaction between the two phases or control the particle size of dispersed phase is the essence to prepare high performance polymer blends. For this reason, compatilizer is widely used in polymer blending due to its specific properties. Generally, compatilizer has similar chain structures to the two components of polymer blends or can react with one component *via* the functional groups^[5]. When the compatilizer is added into immiscible polymer blends, it decreases the particle size of dispersed phase by reducing the interfacial tension of the two phases. For example, polyethylene/polyamide (PE/PA), exhibiting great potential in practical interest, is one of the typical immiscible polymer blends that have been studied in many works^[6–11]. Massive researches have reported that different kinds of compatibilizers can be used to reduce the dispersed particles diameter and stabilize the interface of PE/PA blends^[12–16]. For instance, Sumana *et al.*^[14]

* This work was financially supported by the National Natural Science Foundation of China (No. 51421061).

** Corresponding author: Qiang Fu (傅强), E-mail: qiangfu@scu.edu.cn

Received February 13, 2017; Revised March 14, 2017; Accepted March 19, 2017

doi: 10.1007/s10118-017-1954-1

reported that the average diameter of dispersed phase could be reduced from 3.76 μm to 2.48 μm when 2 wt% compatilizer (maleic anhydride grafted polyethylene) was added to polyethylene/polyamide6 (80/20, *W/W*) blends. Even though compatilizer shows an advantage in tuning morphology and properties of polymer blends, there are still some problems on the usage of compatilizer. For example, it is not always possible to find a good compatilizer for every polymer blends. What is more, addition of compatilizer will increase the cost for industrial manufacturing.

For polymer blends, their properties also depend on processing method, as proper processing method improves the properties as well as enhances industrial application prospects^[17, 18]. Recently, a “high shear processing” technique has been developed and used to fabricate high performance polymer blends, which provides a possibility to the development of nanostructured blends with properties not possible to reach by classical processing techniques^[19–28]. The “high shear processing” technique refers to that polymer blends are obtained under a high shear field by using a special high speed extruder which can reach a maximum screw rotation speed more than 1000 r/min while traditional processing is carried out under screw rotation speed of 150–300 r/min. For a qualitative point of view, the effect of high rotation during processing is usually expressed by SME, which is used to quantify the level of energy transferred to the blends. SME can be calculated by using the following equation:

$$\text{SME} = \frac{\tau \times N}{Q} \quad (1)$$

where τ represents the torque (N·m), N is the screw rotation speed (min^{-1}) and Q is the feed rate (kg/min). SME is so important because it takes both the screw rotation speed and the feed rate into consideration. As we all know, the immiscible blends during processing exhibit a competition between breakup and coalescence of particles. The high rotation speed leads to a high SME which is helpful for the break-up of dispersed particles. Bouaziz *et al.*^[20] investigated the contribution of high shear processing to (polypropylene/ethylene propylene rubber)/polyethylene ternary blends, and reported a decrease in the particle size of (polyethylene/ethylene propylene rubber) domains when increasing rotation speed from 300 r/min to 600 r/min. Teyssandier *et al.*^[26] demonstrated that high shear processing could lead to improved mechanical properties of polyamide12 (PA12)/plasticized starch (TPS) blends without any additives. They attributed these findings to the finer dispersion of TPS within PA12 matrix and a good adhesion at the interface by strong hydrogen bonding between plasticized starch and polyamide12 phases, as induced by high shear rate. Furthermore, polymer blends with nanostructure can be obtained when appropriate system and processing condition are designed. For example, Li and Shimizu^[22] achieved high transparent polycarbonate/poly(methyl methacrylate) blends containing domains with a size of less than 50 nm when increasing rotation speed from 300 r/min to 2250 r/min. An improvement of the mechanical properties of the high-shear-processed blend was clearly observed compared with a low-shear-processed blend.

As outlined above, processing under high shear flow is considered to be a very effective technique for controlling the particle size of dispersed phase. Up to now, the reported researches of high shear processing mainly focus on polymer blends with similar polarity, such as (polypropylene/ethylene propylene rubber)/polyethylene and polycarbonate/poly(methyl methacrylate) blends, or blends which may have interaction between the components during processing, such as PA12/TPS blends. But very less work has been done on polymer blends with huge polarity difference, so called completely immiscible blends, such as polyethylene/polyamide blends which have been widely used in industry. Therefore, it is necessary to clarify the effect of high shear processing on morphology and property of such polymer blends. In our work, we studied the influence of high shear processing on phase morphology of polyethylene/polyamide12 (PE/PA12) blends, a typical immiscible polymer blend without the addition of any additives. Since the two components are completely immiscible and the dispersion is mainly achieved *via* strong shear rate, its stability is the main concern. For this reason, the morphology stability of PE/PA12 blends obtained *via* high shear processing was investigated by high temperature annealing. Finally, the influence of phase morphology on mechanical and

rheological properties was discussed and compared with those obtained *via* conventional processing or low shear processing.

EXPERIMENTAL

Materials

High density polyethylene (HDPE) (5000S) with a melt flow rate (MFR) of 1.0 g/10min (2.16 kg, 190 °C) was purchased from Lanzhou Petrochemical Co., Ltd., China. PA12 (VESTAMID L1670), with a viscosity number of 112–130 cm³/g, was supplied by Evonik Industries (Germany).

Sample Preparation

All the materials were dried at 80 °C for 8 h prior to extrusion and injection. Three kinds of binary blends were prepared by varying the PA12 content in PE matrix, with the PA12 contents of 5 wt% (PE/PA12/95/5), 10 wt% (PE/PA12/90/10) and 20 wt% (PE/PA12/80/20). The melt blending was carried out on a high shear co-rotating twin-screw extruder (Leistritz ZSE 18 MAXX) at barrel temperature from 170 °C to 210 °C and the throughput rate around 3 kg/h. The screw diameter and the length/diameter (*L/D*) ratio are respectively about 18 mm and 40. The screw rotation speeds (*N*) were set to 100, 300, 500 and 700 r/min. After drying, the blends were injected into a mold using the mini-injected machine (Haake MiniJet) with a barrel temperature of 190 °C for mechanical test. Specimens with a thickness of 1 mm for rheology property measurements were prepared by hot pressing at 190 °C.

Viscosity Analysis

The viscosity of pure PE and PA12 was measured at various rotation speeds carried out on a piston mode Rosand RH70 (Malvern, Bohlin Instruments) capillary rheometer with the measurement temperature of 190 °C. The selected shear rate for measurement was in the range of 50–1000 s⁻¹.

Scanning Electron Microscopy (SEM)

Scanning electron microscopy (SEM) experiments were performed using an FEI Inspect F SEM instrument with an acceleration voltage of 20 kV. For morphological observations of the blends, samples were first cryo-fractured in liquid nitrogen vertical to the shear flow direction. The particle size of PA12 phase dispersed in PE matrix was analyzed using an Image-Pro Plus software. For each specimen, at least 300 particles from several independent SEM micrographs were measured. The number-average particle size (*d_n*) was calculated from the following equation.

$$d_n = \frac{\sum_{i=1}^N n_i d_i}{\sum_{i=1}^N n_i} \quad (2)$$

Mechanical Properties

Mechanical properties were measured on an Instron 5567 universal testing machine by uniaxial tension. The samples were tested with a cross-head speed of 50 mm/min and with a gauge length of 30 mm. The specimen dimensions were 70 mm in length, 4 mm in width, and 2 mm in thickness. Notched Izod impact strength was measured according to ISO180/179 using an impact tester (VJ-40, China). The measurements were carried out at room temperature (23 °C). All the mechanical parameters reported were derived from at least five specimens.

Dynamic Rheological Analysis

The melt rheology properties of PE/PA12 blends were measured in a strain-controlled dynamic rheometer (Bohlin Gemini 2000, Malvern, British). For the measurements, disk-like samples obtained from the compression molding of the extrudates were used with diameter of 25 mm and thickness of 1 mm. The strain amplitude was set as 1% with the frequency sweep performed in the range of 0.06–628 rad/s. All the measurements were carried out at 190 °C under nitrogen atmosphere to prevent thermo-oxidative degradation.

RESULTS AND DISCUSSION

The Effect of Rotation Speed on the Degradation of PE and PA12

As mentioned before, the high value of SME is helpful for the break-up of dispersed particles. Thus, the values of SME at different rotation speeds are calculated and shown in Table 1. It can be seen that τ almost keeps a constant while SME shows a rapid increase as increasing the rotation speed, which is very helpful for the dispersion of polymer blends. However, such a high SME can also lead to the degradation of polymer. For this purpose, the possible degradation of pure PE and PA12 processed at various rotation speeds was roughly evaluated by viscosity measurements. As shown in Fig. 1, PE does not exhibit a significant change in viscosity while PA12 shows a decrease of viscosity after processed at 700 r/min. This decrease suggests a reduction of the molecular weight, according to the relation $\eta_0 = KM_w^{3.4}$. In order to avoid the degradation of PA12, the rotation speed up to 500 r/min was used for the preparation of PE/PA12 blends.

Table 1 SME values under different rotation speeds

Rotation speed (r/min)	τ (N·m)	SME (kJ/kg)
100	35.51	142
300	33.37	400
500	32.95	639
700	33.59	855

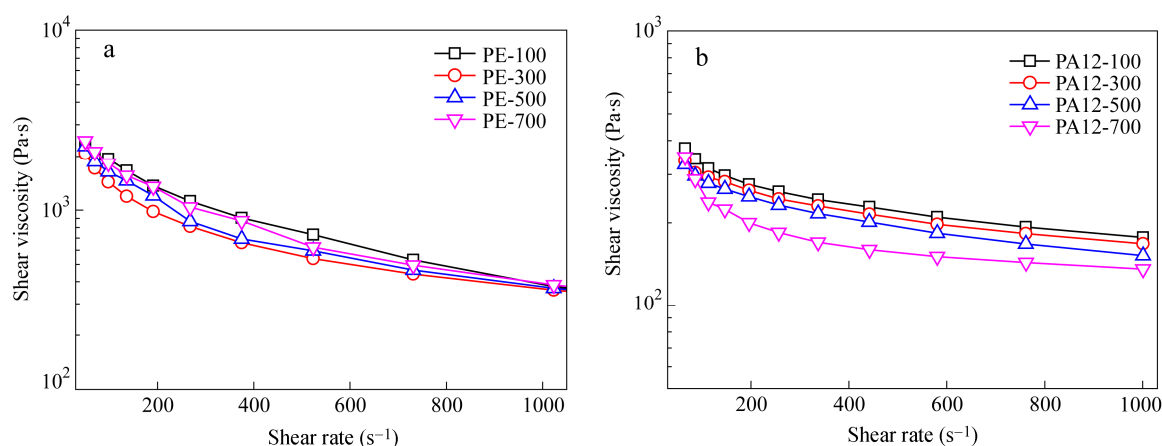


Fig. 1 Shear rate dependence of (a) melt viscosity of PE and (b) melt viscosity of PA12

Phase Morphologies of the Blends

In order to have a clear understanding of the morphology of the PE/PA12 blends, SEM observations of all the samples were carried out and the results are shown in Fig. 2. As expected, all of the samples exhibit a typical sea-island structure in which spherical PA12 droplets are dispersed in the PE matrix. It can be seen from Fig. 2 that the increase of rotation speed has great influence on the dimension of the dispersed PA12 droplets in PE matrix when PA12 content at 5 wt% and 10 wt%. The dimension of dispersed phase is decreased sharply along with the increase of rotation speed from 100 r/min to 500 r/min. For the PE/PA12 blends containing 20 wt% PA12, however, there is no significant decrease of average particle size, indicating that component percentage also influences the morphology of the PE/PA12 blends processed by high shear extruder. To quantitative characterize the precise dimension and distribution of PA12 particles, diameters of PA12 particles were calculated and the results are shown in Fig. 3. PE/PA12 blends processed at 100 r/min exhibit the biggest PA particle sizes, about 1.05 μm for PE/PA12/95/5 and 1.11 μm for PE/PA12/90/10 blends, respectively. With rotation speed increased to 500 r/min, the sizes of the dispersed particles of PA are greatly decreased and the average diameter is decreased to 0.53 μm for PE/PA12/95/5 and 0.60 μm for PE/PA12/90/10 blends,

respectively. The PE/PA12/80/20 blend does not exhibit a significant change when rotation speed increases from 100 r/min to 500 r/min, which may be due to the fact that collision probability between particles is increased with the increase of dispersed phase content. The small particles caused by high shear coalesce to form large particles quickly. Nevertheless, the particle size distribution is also improved along with the decrease of particle size, which is beneficial for the mechanical properties of the blends.

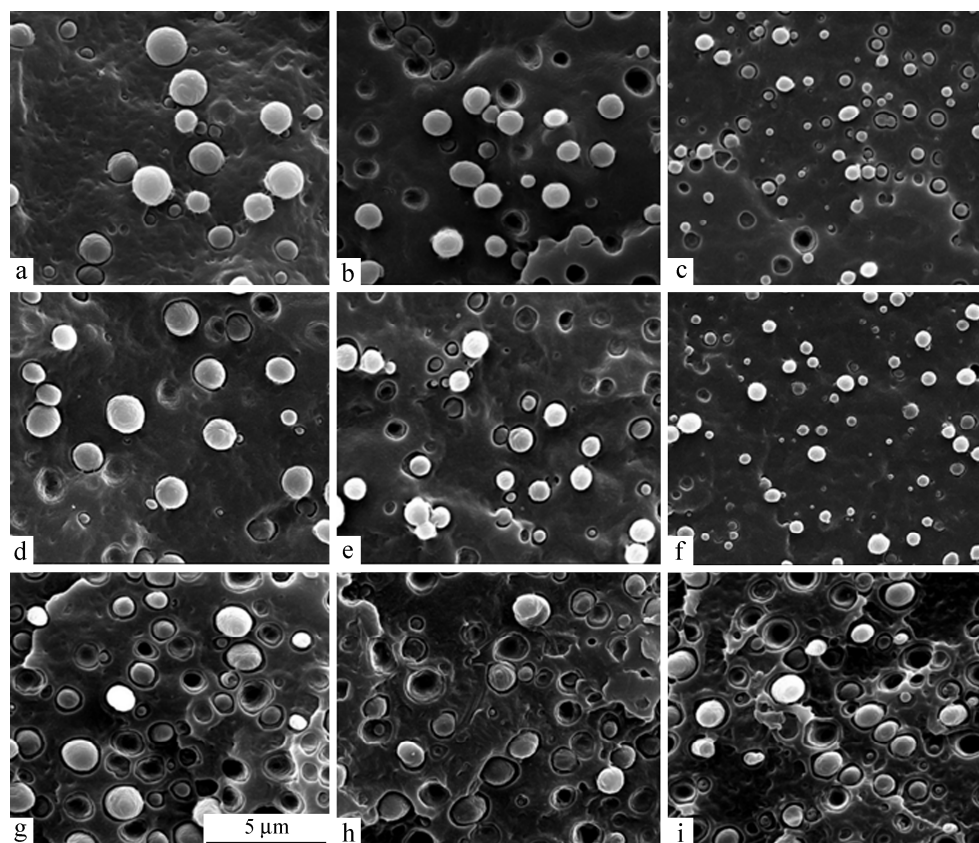


Fig. 2 SEM images of PE/PA12/95/5 blends prepared at (a) 100 r/min, (b) 300 r/min, (c) 500 r/min; PE/PA12/90/10 blends prepared at (d) 100 r/min, (e) 300 r/min, (f) 500 r/min and PE/PA12/80/20 blends prepared at (g) 100 r/min, (h) 300 r/min and (i) 500 r/min

Mechanical Properties

In general, mechanical properties of polymer blends are mainly determined by their morphology. Based on this, experiments on the mechanical properties were carried and the results are shown in Fig. 4 and listed in Table 2, including tensile strength, elongation at break, Young's modulus and impact strength.

As shown in Fig. 4 and Table 2, Young's modulus and tensile strength do not exhibit an obvious variation for PE/PA12 blends obtained at different rotation speeds. However, the elongation at break of the blends is obviously increased with the increase of rotation speed when PA12 content is at 5 wt% and 10 wt%, but does not show a big difference when the PA12 content is at 20 wt%. The small elongation at break of PE/PA12 blends processed at 100 r/min is attributed to the big domain size observed previously by SEM. By increasing rotation speed from 100 r/min to 500 r/min, the elongation at break increases from 90.2% to 306.9% for PE/PA12/95/5 (Fig. 4a) and from 70.3% to 224.2% for PE/PA12/90/10 (Fig. 4b), respectively, where an approximately 240% and 220% improvement has been achieved, respectively. In other words, the elongation at break increases from 90.2% to 306.9% as the PA12 particles dimension decreases from 1.05 μm to 0.53 μm for PE/PA12/95/5 blend and increases from 70.3% to 224.2% for PE/PA12/90/10 blend as PA12 particle sizes decreases from 1.11 μm to

0.60 μm . However, for PE/PA12/80/20 blend, the elongation at break is always low, which is only about 50%–60%, mainly due to its big domain size.

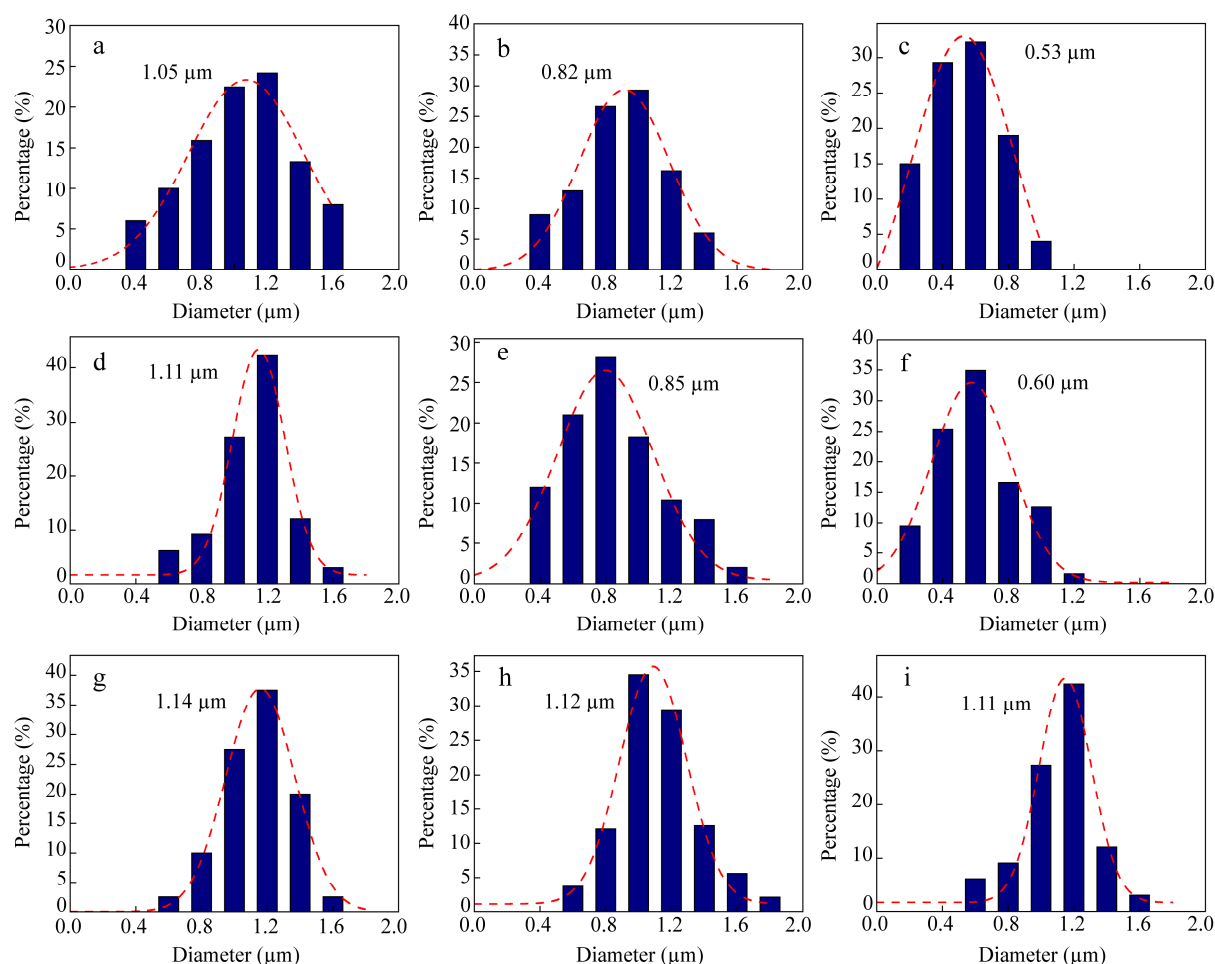


Fig. 3 The particle size distribution of PE/PA12/95/5 blends prepared at (a) 100 r/min, (b) 300 r/min, (c) 500 r/min; PE/PA12/90/10 blends prepared at (d) 100 r/min, (e) 300 r/min, (f) 500 r/min and PE/PA12/80/20 blends prepared at (g) 100 r/min, (h) 300 r/min, (i) 500 r/min (The datum in each graph indicates the average diameter of PA12 particles.)

Table 2 Mechanical properties of PE/PA12 blends at different rotation speeds

Sample name	Tensile stress (MPa)	Elongation at break (%)	Young's modulus (MPa)	Impact strength (kJ/m ²)
PE/PA12/95/5-100	28.9 ± 1.2	90.2 ± 8.7	694 ± 12	14.5 ± 0.6
PE/PA12/95/5-300	28.8 ± 1.5	203.3 ± 12.1	654 ± 13	17.8 ± 0.9
PE/PA12/95/5-500	28.6 ± 0.6	306.9 ± 17.2	649 ± 15	22 ± 1.1
PE/PA12/90/10-100	28.7 ± 1.1	70.3 ± 6.9	671 ± 11	9.3 ± 0.5
PE/PA12/90/10-300	29.4 ± 1.5	160.1 ± 12.0	704 ± 14	10.7 ± 0.7
PE/PA12/90/10-500	29.8 ± 0.7	224.2 ± 13.6	714 ± 13	12.7 ± 0.6
PE/PA12/80/20-100	28.1 ± 0.6	64.6 ± 2.4	727 ± 12	7.6 ± 0.3
PE/PA12/80/20-300	28.0 ± 0.4	52.7 ± 5.7	719 ± 25	7.0 ± 0.4
PE/PA12/80/20-500	28.0 ± 0.4	54.7 ± 6.5	724 ± 19	7.4 ± 0.2

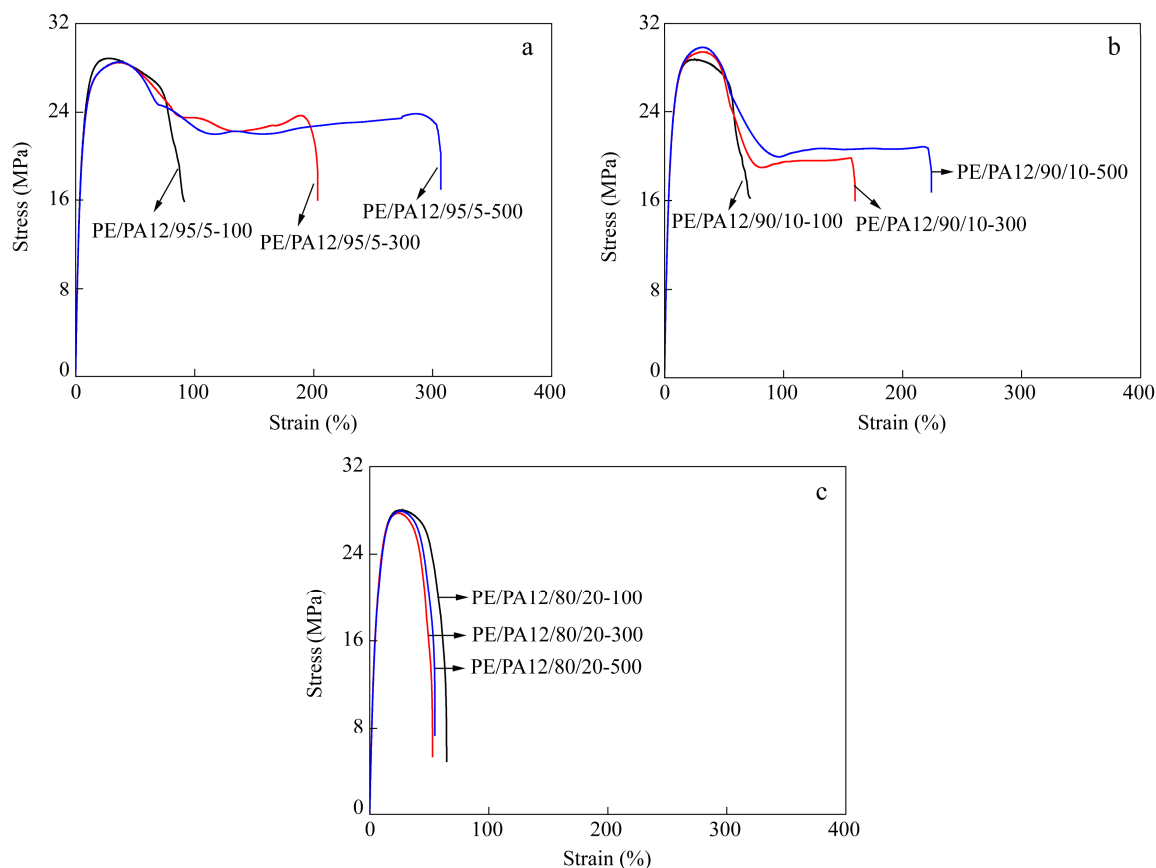


Fig. 4 Typical strain-stress curves of (a) PA/PA12/95/5 blends, (b) PE/PA12/90/10 blends and (c) PE/PA12/80/20 blends prepared at different rotation speeds

Figure 5 shows the comparison of notched Izod impact strength of all samples. As expected, the blends exhibit inferior impact strength due to the poor interfacial adhesion between PE and PA12 components. With increasing rotation speed from 100 r/min to 500 r/min, the impact strength of PE/PA12/95/5 blends increases from 14.5 kJ/m² to 22 kJ/m², where a 51% improvement is obtained, while it is increased from 9.3 kJ/m² to 12.7 kJ/m² for PE/PA12/90/10. However, the impact strength of PE/PA12/80/20 blends processed at different rotation speed does not show big difference due to the big dispersed phase as observed by SEM. In our work,

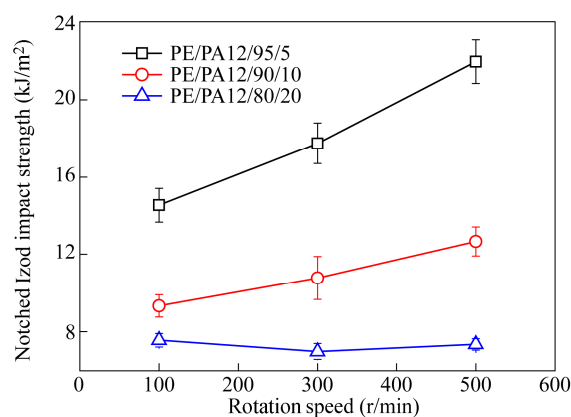


Fig. 5 Notched Izod impact strength of samples as a function of rotation speed

sample prepared at 100 r/min exhibits larger sizes for the dispersed particles than that prepared at 500 r/min and the distribution of particles diameters is also wider than that prepared at 500 r/min. Therefore, the sample prepared at 100 r/min with large particles tends to induce premature crack propagation due to the coalescence of crazes, following craze initiation and propagation at the early stage of the impact testing, and then inevitably lead to a brittle fracture. Furthermore, the wider distribution of the particles leads to an inhomogeneous distribution of the particle sizes which may also contribute to the inferior fracture toughness.

Rheology Property

In order to evaluate the influence of dispersed particle size on rheology property, variation of the elastic modulus (G') versus frequency of blends PE/PA12/95/5 was investigated and is shown in Fig. 6(a). As we can see from Fig. 6(a), G' increases with the increasing frequency for all samples. Actually, at lower frequencies, the increase of G' from PE/PA12 blends processed at 100 r/min to 500 r/min should be attributed to the relaxation of the shape of the dispersed phase when enduring high shear, where the shape deformation of phase interface is converted into interface energy and eventually becomes storage elastic energy. Since PE/PA12 blends processed at higher rotation speed have a smaller average size of PA12 particles, more interface is expected between PE matrix and PA12 dispersed phase, which induces a higher elastic modulus.

Figure 6(b) shows the dynamic viscosity of PE/PA12/95/5 blends as a function of frequency. The viscosity decreases with increasing frequency, indicating that the PE/PA12 blends exhibit shear-thinning behavior. Again an increase of viscosity of blends obtained from 100 r/min to 500 r/min is possibly due to the enhanced dispersion promoted by the high shear processing.

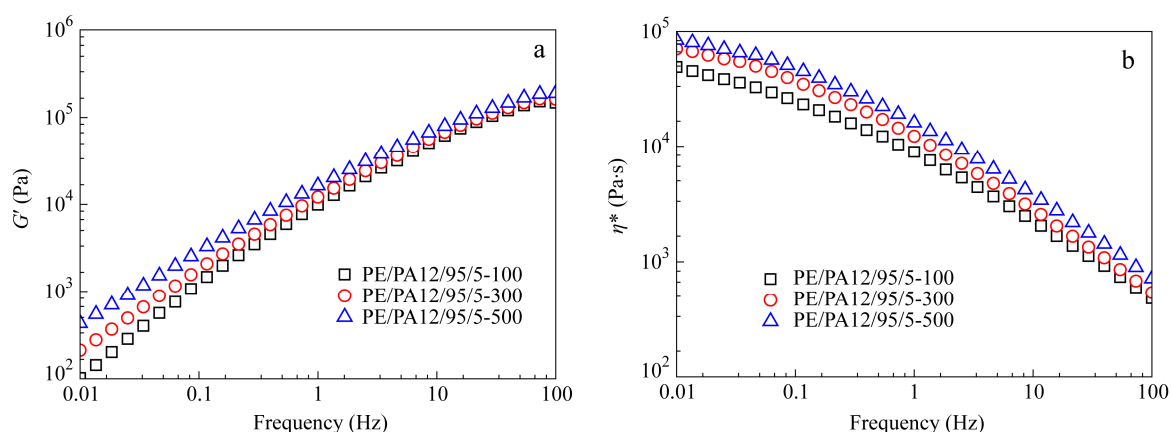


Fig. 6 Linear viscoelastic properties of PE/PA12/95/5 blends: (a) storage modulus G' and (b) absolute complex viscosity η^*

The Thermal Stability of Morphology

Reduced size of PA12 particles was obtained during high shear processing; however, as a consequence of no compatilizer being used in PE/PA12 blends, the morphology should be instable. Massive works have reported that the addition of a compatilizer in blends could reduce the interfacial tension and enhance the interfacial adhesion leading to a quite stable morphology. In our case, the instable morphology of PE/PA12 blends without any interfacial interaction may lead to droplet coalescence during a second processing step, which would strongly increase the size of the dispersed phase and consequently affect all the properties of the blends. To study the morphology stability of the PE/PA12 blends, the PE/PA12/95/5 samples prepared at 500 r/min were annealed at 190 °C, which is the same as barrel temperature of mini-injection, for 5, 10 and 60 min. The morphologies of the annealed samples after different annealing times are shown in Fig. 7 and its particle size distribution is shown in Fig. 8. As can be seen, the size of the dispersed PA12 particles keeps unchanged during annealing for 5 min (Figs. 7b and 8b), and only a little increase when annealed for 10 min (Figs. 7c and 8c). This result means that almost no droplet coalescence of PA12 particles has occurred during the first 10 min of annealing time. Thus we know the morphology of PE/PA12 blends during the rheological measurement does not change significantly

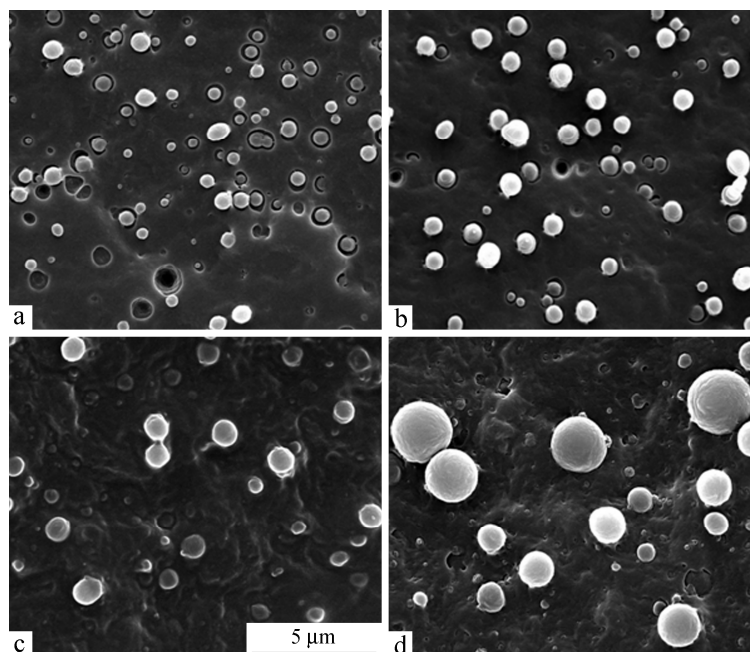


Fig. 7 SEM images of PE/PA12/95/5-500 (a) as prepared and after annealed at 190 °C for (b) 5 min, (c) 10 min and (d) 60 min

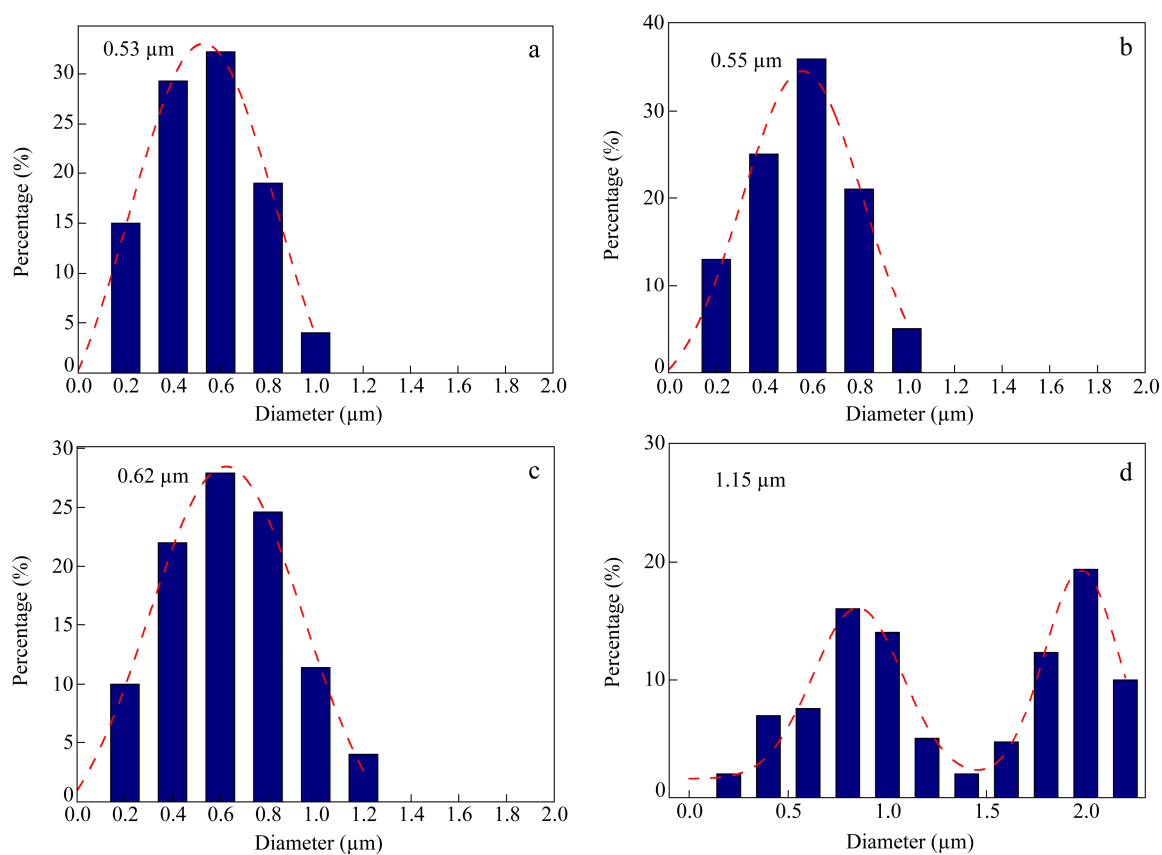


Fig. 8 The particle size distribution of PE/PA12/95/5-500 (a) as prepared and after annealed at 190 °C for (b) 5 min, (c) 10 min, (d) 60 min (The datum in each graph indicates the average diameter of PA12 particles.)

because the rheological test of each sample only lasts about 10 min. So, the rheological measurement can reflect the different morphology of PE/PA12 blend. However, the PA12 particle size of the sample after annealing for 60 min exhibits a significant increase (Figs. 7d and 8d), which becomes larger and more regular compared with the as-prepared samples (Figs. 7a and 8a). Many works have reported that the dispersed phase morphology is coarsened by coalescence through which the free energy of the system is minimized. Therefore, Fig. 7(d) shows PE/PA12/95/5 samples prepared at 500 r/min and annealed for 60 min exhibit a more spherical shape due to minimization of energy. Meanwhile, static coalescence is influenced by the volume fraction of dispersed phase and average particle size. In our case, the PE/PA12/95/5-500 sample with a low volume fraction of PA12 and a smaller particle size leads to a longer coarsening time. It is suggested that though high shear processing is effective also to reduce the size of dispersed phase for blends with huge polarity difference, the resulted morphology is not thermodynamically stable. Thus to prepare high performance polymer blends with a quite stable structure, the compatilizer seems necessary, particularly for blends with huge polarity difference. The combined effect of compatilizer and high shear processing on the phase morphology and its stability of polymer blends is currently investigated in our group.

CONCLUSIONS

In summary, high shear processing has a great influence on morphology of PE/PA12 blends, a typical completely immiscible blend system when PA12 contents are 5 wt% and 10 wt%. However, the morphological change as induced by high shear processing is reduced while the PA12 content is increased to 20 wt%. Accordingly, largely increased elongation at break and impact strength are observed for PE/PA12/95/5 and PE/PA12/90/10 blends obtained at the high rotation speeds. However, the resulted morphology is not thermodynamically stable and the obtained small particle size will be coarsened after annealed at high temperature for longer time. Thus how to prepare high performance polymer blends with a quite stable structure by high shear processing still needs further study. We firmly believe that this work could provide a guideline for the application of high shear processing in the preparation of polymer blends with huge polarity difference.

REFERENCES

- 1 Phetwarotai, W., Tanrattanakul, V. and Phusunti, N., *Chinese J. Polym. Sci.*, 2016, 34(9): 1129
- 2 Yao, H., Niu, J.L., Zhang, J., Ning, N.Y., Yang, X.Q., Tian, M., Sun, X.L., Zhang, L.Q. and Yan, S.K., *Chinese J. Polym. Sci.*, 2016, 34(7): 820
- 3 Zhou, M., Li, X.P., Jin, M., Xia, C., Shen, K.Z. and Zhang, J., *Chinese J. Polym. Sci.*, 2016, 34(8): 1001
- 4 Leibler, L., *Prog. Polym. Sci.*, 2005, 30(8-9): 898
- 5 Xu, Y.W., Loi, J., Delgado, P., Topolkaraev, V., McEneaney, R.J., Macosko, C.W. and Hillmyer, M.A., *Ind. Eng. Chem. Res.*, 2015, 54(23): 6108
- 6 Pernot, H., Baumert, M., Court, F. and Leibler, L., *Nat. Mater.*, 2002, 1(1): 54
- 7 Xiang, F.X., Wang, Y.X., Shi, Y.Y., Huang, T., Chen, C., Peng, Y. and Wang, Y., *Polym. Int.*, 2012, 61(8): 1334
- 8 Filippone, G., Dintcheva, N.T., Acierno, D. and Mantia, F.P., *Polymer*, 2008, 49: 1312
- 9 Araujo, J.R., Vallim, M.R., Spinace, M.A.S. and Paoli, M.A., *J. Appl. Polym. Sci.*, 2008, 110: 1310
- 10 Freluche, M., Iliopoulos, I., Flat, J.J., Ruzette, A.V. and Leibler, L., *Polymer*, 2005, 46: 6554
- 11 Minkova, L., Yordanov, H., Filippi, S. and Grizzuti, N., *Polymer*, 2003, 44: 7925
- 12 Argoud, A., Trouillet-Fonti, L., Ceccia, S. and Sotta, P., *Eur. Polym. J.*, 2014, 50: 177
- 13 Chen, J., Chen, J.W., Chen, H.M., Yang, J.H., Chen, C. and Wang, Y., *Compos. Part B Eng.*, 2013, 54: 422
- 14 Mallick, S., Kar, P. and Khatua, B.B., *J. Appl. Polym. Sci.*, 2012, 123(3): 1801
- 15 Hallden, A., Deriss, M.J. and Wesslen, B., *Polymer*, 2001, 42: 8743
- 16 Sinthavathavorn, W., Nithitanakul, M., Magaraphan, R. and Grady, B.P., *J. Appl. Polym. Sci.*, 2008, 107(5): 3090

- 17 Bai, D.Y., Wang, K., Bai, H.W., Zhang, Q. and Fu, Q., *Acta Polymerica Sinica* (in Chinese), 2016, (7): 843
- 18 Deng, S., Qi, X.D., Zhu, Y.L., Zhou, H.J., Chen, F. and Fu, Q., *Chinese J. Polym. Sci.*, 2016, 34(10): 1270
- 19 Sinthavathavorn, W., Nithitanakul, M., Magaraphan, R. and Grady, B.P., *J. Appl. Polym. Sci.*, 2008, 107(5): 3090
- 20 Bouaziz, A., Massardier, V., Louizi, M. and Jaziri, M., *Polym. Eng. Sci.*, 2015, 55(10): 2328
- 21 Chen, G.X., Li, Y. and Shimizu, H., *Carbon*, 2007, 45(12): 2334
- 22 Li, Y. and Shimizu, H., *Eur. Polym. J.*, 2006, 42(12): 3202
- 23 Li, Y. and Shimizu, H., *Polymer*, 2007, 48(8): 2203
- 24 Li, Y. and Shimizu, H., *Polym. Eng. Sci.*, 2011, 51(7): 1437
- 25 Louizi, M., Massardier, V. and Cassagnau, P., *Macromol. Mater. Eng.*, 2014, 299: 674
- 26 Teyssandier, F., Cassagnau, P., Gérard, J.F., Mignard, N. and Mélis, F., *Mater. Chem. Phys.*, 2012, 133: 913
- 27 Louizi, M., Massardier, V., Melis, F., Alcouffe, P. and Cassagnau, P., *Int. Polym. Proc.*, 2014, 18: 118
- 28 Li, Y. and Shimizu, H., *Macromolecules*, 2009, 42: 2587
- 29 Shimizu, H., Li, Y.J., Kaito, A. and Sano, H., *Macromolecules*, 2005, 38: 7880