

Processability of Polysulfone Improved by Adding Polyamide 6 and Polysulfone-Polyamide 6 Block Copolymer

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ABSTRACT: Although polysulfone (PSU) is a potential thermoplastic engineering plastic with high heat resistance, good dimensional stability and excellent mechanical properties, its poor processability has greatly restricted its application in electrical, aerospace, and medical fields. In this work, polyamide 6 (PA6) and PSU-PA6 block copolymer (PSU-*b*-PA6) were used to improve the processability and formability of PSU depending on their excellent fluidity and good compatibility between two components. Furthermore, the fluidity, thermal and mechanical properties of the blends were carefully investigated. It was found that, melt flow index of PSU could be increased above 10 times, and strength and toughness could be enhanced by 4–10% with the introduction of 10 wt % PA6 and PSU-*b*-PA6 without compromising the heat resistance of PSU obviously. The processing conditions of PSU could be improved while maintaining a decent comprehensive performance. Thus, the method has great potential for extending the applications of PSU. © 2014 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* **2014**, *131*, 41139.

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INTRODUCTION

High-performance thermoplastics like polysulfone (PSU) are attracting much attention owing to its superior heat resistance, outstanding dimensional stability, and excellent mechanical properties.^{1–7} The favorable properties make PSU suitable for many applications, such as electrical and electronic components, medical equipment, household food utensils, automotive parts and aviation devices.⁸ However, just as other high performance polymers, poor processability of PSU greatly limits its application because of its rigid molecular chains. In addition, its high melt viscosity makes processing performance worse. Extrusion temperature of PSU generally maintains in 302–332°C and the injection molding temperature is as high as 320–360°C.⁹ Harsh processing conditions set forth stricter requirements for the common machines. Furthermore, some chemical changes like decomposition or crosslinking of PSU at high temperature also make the forming processes become more difficult. Due to these processing defects, most applications of PSU were restricted in polymer electrolyte membranes,¹⁰ hemodialysis, and apheresis membranes,¹¹ filtration membranes¹² in recent years.

Such processing drawbacks could be overcome by blending PSU with other polymers. Liquid-crystalline polymer (LCP) has exceptional stability at high processing temperature without post-processing migration. Therefore, several studies added LCP as processing aid to the PSU.^{13–15} However, the blends showed

some undesirable properties like poor weld strength and strong anisotropy in mechanical properties. Moreover, the cost of liquid crystal materials was much expensive. M. García et al.¹⁶ prepared glass fiber (GF)-reinforced copolyester LCP (gLCP) to promote the desired properties of the LCP. They found that adding either 10% or 20% gLCP to PSU led to an increase in the melt-flow index of 11%, but the presence of GF increased the viscosity of blends. Compared with the pure PSU, Young's modulus and notched impact strength of the composite increased by adding 10% gLCP, but ductility and tensile strength decreased. Ran and Jiang¹⁷ reported that 5% acrylonitrile-butadiene-styrene copolymers and 3% Polypropylene (PP) could improve the fluidity and stress cracking resistance of PSU, but the impact strength of PSU was also decreased. Chen et al.¹⁸ studied the rheological behavior of PSU/polyphenylene sulfide (PPS) blend in melt processing. They found that the fluidity of PSU/PPS blend was improved obviously due to the good fluidity and thermal stability of PPS. However, the thermal and mechanical properties of the blend were not reported.

Polyamide 6 (PA6) is an important engineering plastics with good mechanical properties, self lubrication, excellent chemical resistance, and good fluidity above the melting point.^{19,20} Blending PSU with PA6 will improve the processability of PSU and obtain the good combination property.²¹ However, most

Table I. Compositions of PA6/PSU-*b*-PA6/PSU Blends

Sample code	PA6 (g)	PSU (g)	Block copolymer (g)
B0	80	720	0
B2	78.4	705.6	16
B5	76	684	40
B8	73.6	662.4	64
B10	72	648	80
A10	36	684	80
C10	144	576	80
D10	216	504	80

blends are thermodynamically immiscible including PSU and polyamide.^{22,23} In these blends, phase separation appeared due to the poor compatibility between the two components and the phase interface usually became the weakness for the material resulting in the decrease of responding performances. The addition of PA6 improved the processability of PSU, but PSU/PA6 blends displayed poor properties owing to their unstable morphology and poor interfacial adhesion.^{24–26}

A compatibilization strategy was usually used to solve this problem by adding a block copolymer as the compatilizer to improve the miscibility between the homopolymers.^{27–30} Charoensirisomboon et al. synthesized a series of block copolymers as the compatilizers of PSU/PA blends.^{31–34} It was found that the block copolymers could play an emulsification effect to decrease the particle size of PSU.

Several researchers synthesized PSU-PA block copolymers by anionic polymerization techniques and researched the performances.^{35–37} But using PA6 and PSU-PA6 block copolymer as processing aids to improve the processability of PSU has not been reported to our knowledge. In this article, we blended PSU with PA6 to decrease the melt viscosity of PSU. In order to enhance the interfacial interaction of PSU/PA6 blends, a block copolymer comprising PSU and PA6 components was employed to improve the compatibility between PSU and PA6 phases. The fluidity, thermal, and mechanical properties of PA6/PSU-*b*-PA6/PSU blends were studied. The role of PA6 and PSU-PA6 block copolymer in the performance improvement of PSU was carefully investigated.

EXPERIMENTAL

Materials

Bisphenol-A PSU powders (P-7303, Dalian PSU Plastic, China, $\eta = 0.52\text{--}0.65$, the density = 1.24 g/cm^3) were dried at 120°C and PA6 pellets (cm1017, Toary Industries, Japan) were dried at 100°C to remove moisture. Caprolactam used for synthesizing block polymer was supplied by Petroleum Chemical Plant (Hunan, China).

Synthesis of Block Copolymer

Caprolactam (300 g) was dissolved at 170°C with vacuum pumping and stirring. Subsequently, 12 g PSU was added and kept vacuuming for 1 h, and then NaOH catalyst was introduced. The reaction temperature was raised to 180°C and kept vacuuming for further 0.5 h, after that, the melt was quickly

poured into a mould at 170°C , and kept heat preservation for another 45 min. The sample was taken out of the mould after cooling. The sample need to be characterized was purified using soxhlet extraction with chloroform, aqueous solution of formic acid to remove unreacted raw materials. And then the product was washed with methanol and dried for 48 h in a vacuum oven.

Processing of Blends

PSU, PA6, and PSU-PA6 block copolymer were blended in a high-speed mixer setting rotor speed at 600 rpm. Then the pre-mixed materials were prepared by melt blending in a single screw extruder (TE-34, Nanjing Keya Chemical Equipment, China). The extrusion temperature ranged from 263 to 293°C and the screw speed was 20 Hz/min. The extrusive pellets were prilled and dried in a vacuum oven at 100°C before molding. The pellets were injection molded into standard samples using injection molding machine (YJ400-II, Ningbo Jiangbei Machinery, China) and the heating temperature ranged from 300 to 360°C . The compositions of PA6/PSU-*b*-PA6/PSU blends were listed in Table I.

Characterization of Block Copolymer

FTIR spectrum was carried out on a Nicolet company IS10 FTIR spectrometer (United States) at a resolution of 2 cm^{-1} . Sample was diluted with KBr and prepared by the compression method. Nuclear magnetic resonance spectrum was obtained using a 400 MHz Bruker AVANCE III NMR spectrometer with tetramethylsilane as a reference. The NMR solvent of block copolymer was deuterated trifluoroacetic acid.

Dynamic Mechanical Analysis

Dynamic mechanical measurements were performed by DMA 242 dynamic mechanical analyzer (NETZSCH, German) with a flat sample at a heating rate of $3^\circ\text{C}/\text{min}$. The operating temperature ranged from -110 to 200°C , and the frequency was 1 Hz. Loss tangent ($\tan\delta$) curves of each sample were automatically recorded.

Fluidity Test

Melt flow index (MFI) measurements were measured using a melt indexer (XRZ-400, Shenzhen Sans, China) and the test condition was set at 343°C with a 2.16 kg load.

Thermogravimetric Analysis

Thermogravimetric analysis (TGA) was carried out with a DTG-60H thermogravimetric analyzer (SHIMADZU Company, Japan) from room temperature to 750°C at a heating rate of $10^\circ\text{C}/\text{min}$ under nitrogen atmosphere.

Scanning Electron Microscopy

The tensile fracture surfaces were coated with a thin layer of gold to observe the damage morphology of blends under scanning electron microscopy (HITACHI S-4800 field emission scanning electron microscope, Japan). The accelerating voltage was 5 kV.

Mechanical Properties Test

Tensile strength and elongation at break of blends were measured using electronic universal testing machine (CMT6104, Shenzhen Sans, China) at a tensile rate of $2\text{ mm}/\text{min}$ on $150 \times$

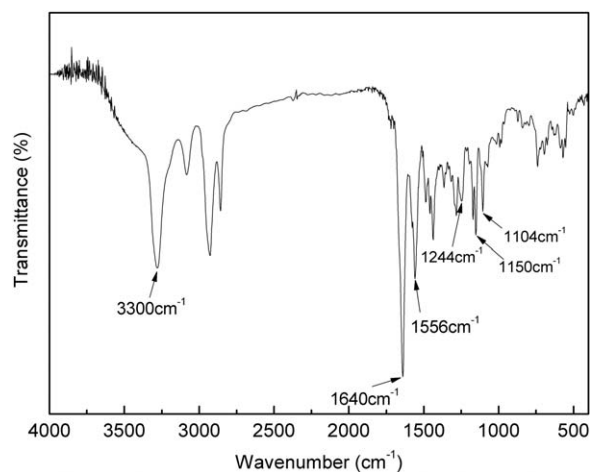


Figure 1. FTIR spectrum of PSU-PA6 block copolymer.

10 × 4 mm specimens. Notched impact strength of blends was measured using pendulum impact testing machine (ZBC-4, China) on 80 × 10 × 4 mm specimens at an average notch depth of 1.0 mm. Seven samples for each composition were tested and the average values were reported.

RESULTS AND DISCUSSION

Characterization of PSU-PA6 Block Copolymer

FTIR spectrum of PSU-PA6 block copolymer was showed in Figure 1. The absorption peak at 3300 cm^{-1} corresponded to the N—H stretching vibration of PA6. There were strong absorption bands at 1640 and 1556 cm^{-1} , which were attributed to the vibration of amide group. The peak at 1244 cm^{-1} was assigned to the C—O—C band asymmetric stretching vibration of PSU. The peaks at 1150 and 1104 cm^{-1} were characteristic of O=S=O group and S—C band stretching vibration of PSU, respectively.^{38,39} The characteristic absorption peaks of PA6 and PSU existed simultaneously in the spectrum after purifying, indicating that the block copolymer had been synthesized.

The structure of PSU-PA6 block copolymer was further confirmed by $^1\text{H-NMR}$ analysis and $^1\text{H-NMR}$ spectrum was pre-

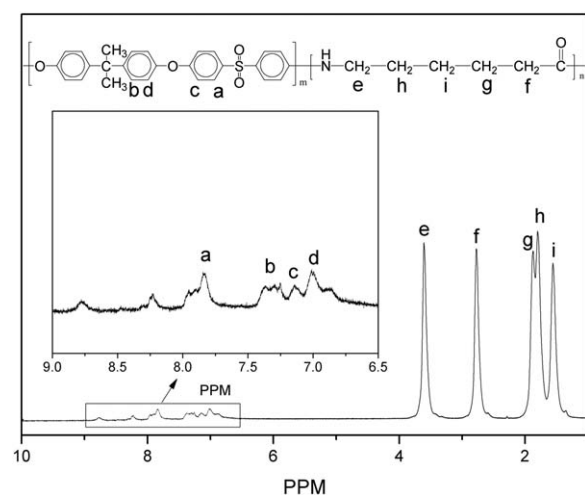


Figure 2. ^1H NMR spectrum of PSU-PA6 block copolymer.

Table II. Parameters for the PSU-PA6 Block Copolymer

$[\eta]^a$	M_n^a	$M_n(\text{PSU})^b$	$M_n(\text{PA6 block})$	$T_g(\text{°C})^c$	$T_m(\text{°C})^d$
200.4	70,902	25,861	45,041	79.6	216.2

^a Measured at 25°C in concentrated sulfuric acid using Ubbelohde viscometer.

^b Measured at 25°C in chloroform using Ubbelohde viscometer.

$M_n(\text{PA6 block})$ was calculated under the assumption that the molecular weight of the PSU was preserved without chain scission during polymerization.

^c Determined by DMA.

^d Determined by DSC.

sented in Figure 2. The characteristic $^1\text{H-NMR}$ peaks of PSU-PA6 block copolymer at 7.84, 7.30, 7.13, and 7.01 ppm were assigned to aromatic protons of the PSU backbone. The characteristic peaks of block copolymer at 3.64, 2.77, 1.89, 1.79, and 1.55 ppm were assigned to the protons of PA6 chain. The results showed that the final product included PSU and PA6 structures after purifying, further confirming the synthesis of block copolymer. And intrinsic viscosity, viscosity-average molecular weight, glass transition temperature, and melting temperature of the block copolymer were also gathered in Table II.

Dynamic Mechanical Analysis

To confirm the compatibilization of PSU-*b*-PA6, the loss tangent ($\tan\delta$) variation curves of ternary blends as a function of temperature were shown in Figure 3. Two main relaxations in the investigative temperature interval were observed. The $\tan\delta$ peaks appearing between 50 and 70°C were assigned to the α -relaxation (glass transition) of PA6 main chains, and the peaks appearing between 170 and 180°C were assigned to the glass transition of amorphous PSU phase.^{40,41} The presence of two transition peaks indicated that PA6 and PSU phases were not molecularly miscible.

Glass transition temperatures (T_g s) of blends are directly related to the compatibility of polymer components. To exhibit the compatibility more intuitively, plots of T_g of each phase as a function of block copolymer and PA6 content were depicted. As shown in Figure 3 and Table III, T_g of two components converged each other with the addition of copolymer. T_g of PSU was shifted to a lower temperature (from 180.1 to 175.9°C) with increasing the content of copolymer, whereas T_g of PA6 was shifted to a higher temperature (from 59.6 to 70.5°C). The shift in the T_g peaks of the blends implied the better compatibility between the PA6 and PSU components.^{42,43} Block copolymer could reduce interfacial tension of each domain resulting in a fine dispersion of PA6. The concentration of PA6 also revealed an effect on the compatibility as shown in Figure 3(b). Further addition of PA6 component impaired the compatibilization, since the excess PA6 might agglomerate together leading to a bad dispersion. Sample B10 (PA6/PSU-*b*-PA6/PSU = 9/10/81) displayed the better compatibility in the blends.

Fluidity of Blends

The effects of PA6 and PSU-PA6 block copolymer on the processability of the blends were studied by means of MFI measurements and the results were depicted in Figure 4. It was evident

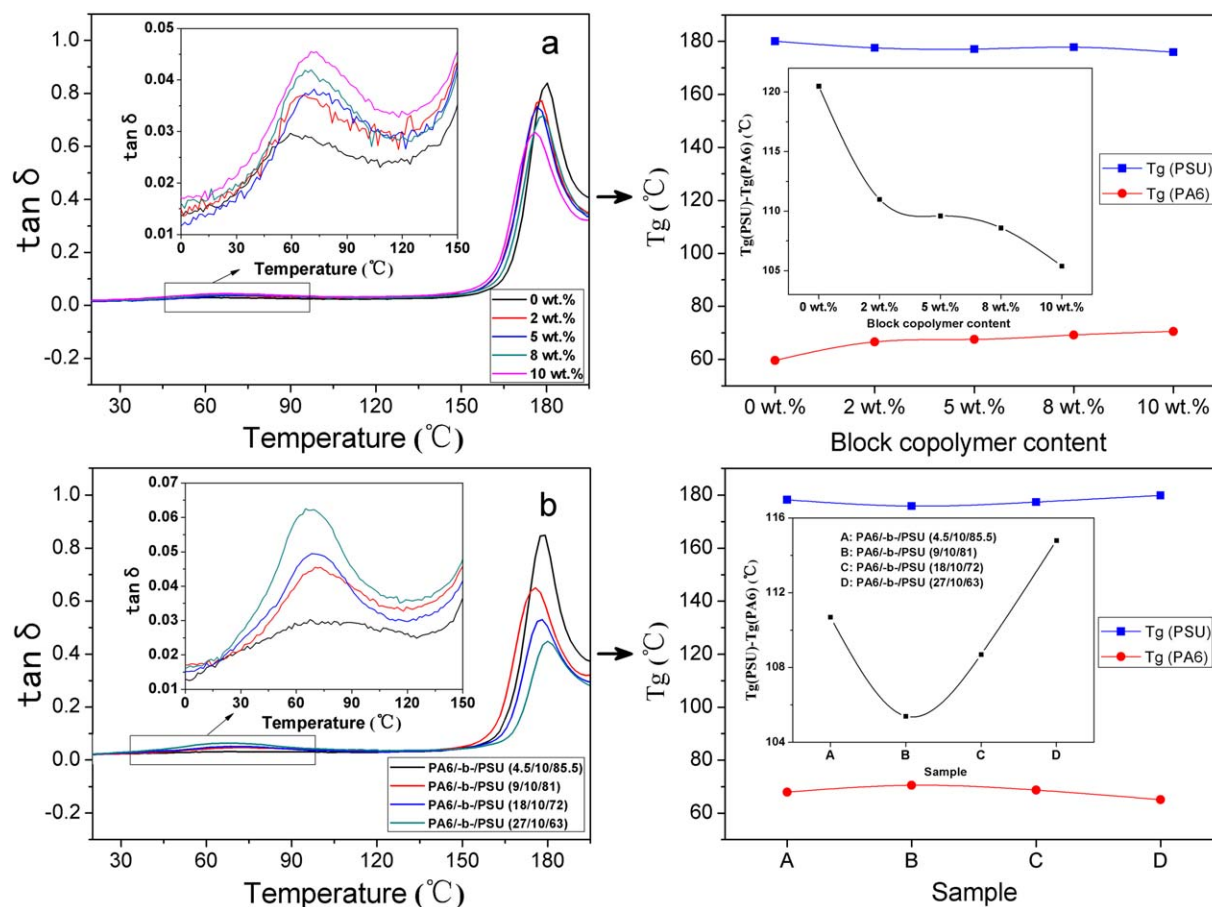


Figure 3. $\tan \delta$ variation curves and corresponding plots of T_g of PSU and PA6 in the blends: (a) different content of block copolymer in blends (PA6/PSU = 1/9); (b) different mass ratio of PA6/PSU (10 wt % block copolymer). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

that PSU represented poor fluidity owing to its rigid molecular chain. Whereas a remarkable improvement in the MFI of PSU was achieved after adding PA6 and block copolymer. In Figure 4(a), MFI of blends appeared to increase monotonically with increasing copolymer content, and the addition of 10 wt % copolymer led to a sharp increase. In addition, MFI of ternary blends improved more observably as PA6 content increased (b). MFI of Sample E (PA6/PSU-*b*-PA6/PSU = 27/10/63) increased nearly 40 times in contrast to the pure PSU, indicating that PA6 and PSU-*b*-PA6 could decrease melt viscosity of PSU and improve the processability.

To express the effect of PA6, a control experiment without PA6 component (Sample A: PA6/PSU-*b*-PA6/PSU = 0/10/90) was arranged. The result showed the irreplaceable role of PA6 in the blends considering the incorporation of block copolymer solely had a little effect on the improvement of fluidity. Several reasons could be speculated for the viscosity reduction of PSU matrix. PA6 exhibited a low viscosity above the melting point because of its flexible chain, which could play a lubricating role in the reduction of PSU's viscosity. It was believed that PA6 phases dispersed more uniformly in the PSU matrix owing to the addition of block copolymer, and the even PA6 phase reduced the friction between the PSU phases resulting in the increase of MFI. Thus, the viscosity of these blends decreased apparently.

TGA of Blends

To investigate the thermal stability of blends, the curves of TGA and the corresponding 5% weight-loss degradation temperatures ($T_{d5\%}$) were showed in Figure 5 and Table III, respectively. The $T_{d5\%}$ of pure PSU was 487.9 $^{\circ}\text{C}$, whereas the $T_{d5\%}$ of pure PA6 was 379.2 $^{\circ}\text{C}$. The degradation temperatures of blends were

Table III. Thermal Properties of PA6/PSU-*b*-PA6/PSU Blends

Sample code	T_g (PSU) in block ($^{\circ}\text{C}$)	T_g (PA6) in block ($^{\circ}\text{C}$)	T_g (PSU) - T_g (PA6) value ($^{\circ}\text{C}$)	$T_{d5\%}$ ($^{\circ}\text{C}$)
PSU				487.9
PA6				379.2
B0	180.1	59.6	120.5	421.2
B2	177.6	66.6	111.0	423.6
B5	177.1	67.5	109.6	421.0
B8	177.8	69.2	108.6	415.9
B10	175.9	70.5	105.4	410.9
A10	178.3	67.6	110.7	420.0
C10	177.4	68.7	108.7	396.0
D10	179.9	65.1	114.8	399.0

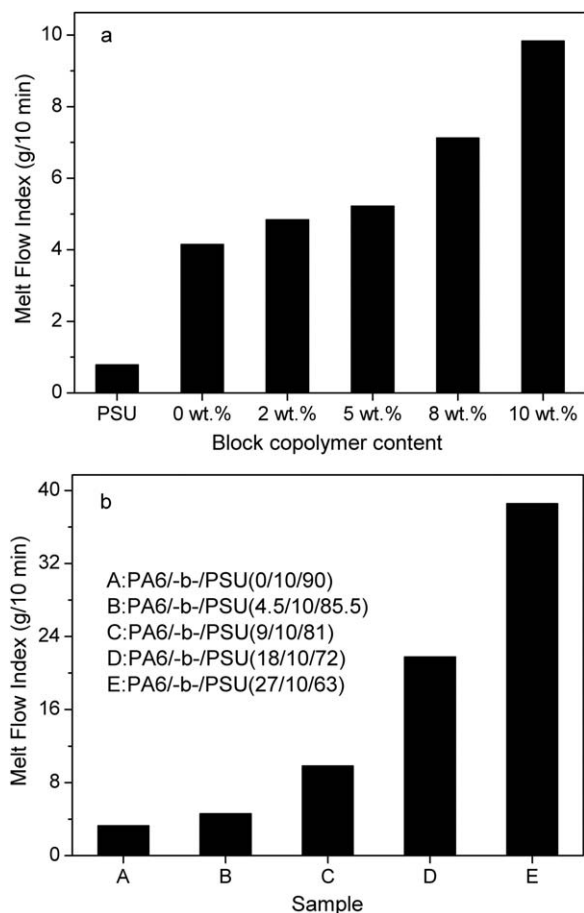


Figure 4. MFI of PSU, PA6/PSU, and PA6/PSU-*b*-PA6/PSU blends. (a) different content of block copolymer in blends (PA6/PSU = 1/9); (b) different mass ratio of PA6/PSU (10 wt % block copolymer).

somewhere between the parent components. In addition, two obvious degradation processes were found in the curves. The first step was due to the thermal degradation of PA6 chains, and the second step was due to the PSU chains. The curves with different content of block copolymer showed the same trend of degradation process, which indicated that the addition of block copolymer exhibited little effect on the degradation behavior of PSU/PA6 blend. Although $T_{d5\%}$ s of ternary blends were not as high as that of the PSU, the blends remained decent thermal stability since a significant weight loss below 410°C was not observed.

PSU possessed the better thermal stability due to the aromatic ring structure on its backbone. Compared with PSU, the thermal stability of PA6 showed much weaker owing to the amide groups. Meanwhile, amide groups were sensitive to polar material, which gave rise to the poor thermal stability.⁴⁴ Therefore, the introduction of PA6 segments was bound to drop the initial decomposition temperature of blends.

Micrographs of the Blends

The morphology of PSU/PA6 and PA6/PSU-*b*-PA6/PSU blends was observed in SEM images of the tensile fracture surfaces (Figure 6). With respect to the PA6/PSU binary blend [Figure 6(A)], it exhibited a smooth fracture surface between the matrix

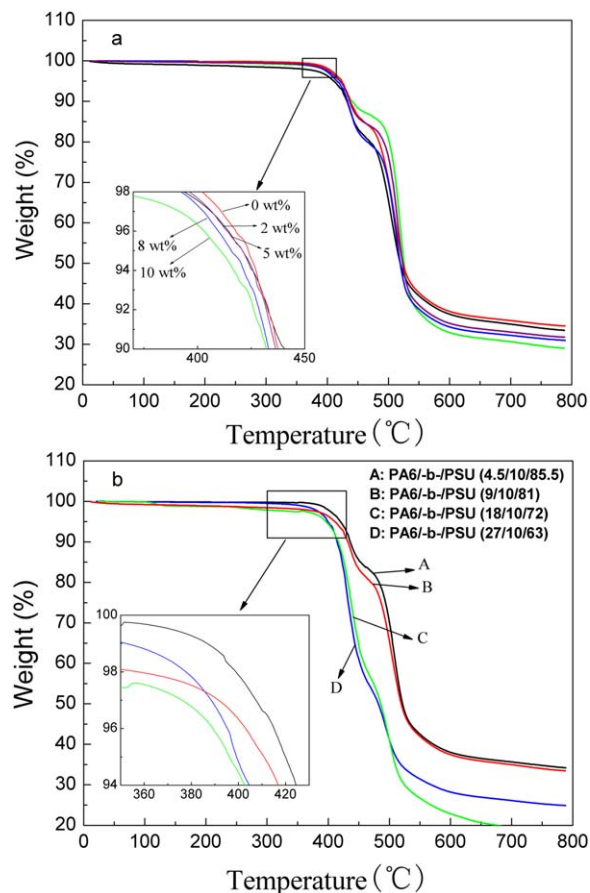


Figure 5. Thermogravimetric curves of PA6/PSU and ternary blends. (a) different content of block copolymer in blends (PA6/PSU = 1/9); (b) different mass ratio of PA6/PSU (10 wt % block copolymer). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

and dispersed phase. And the boundary was clear after the PA6 phase was pulled out. Distinct phase interface demonstrated the weak interfacial adhesion and poor compatibility between the

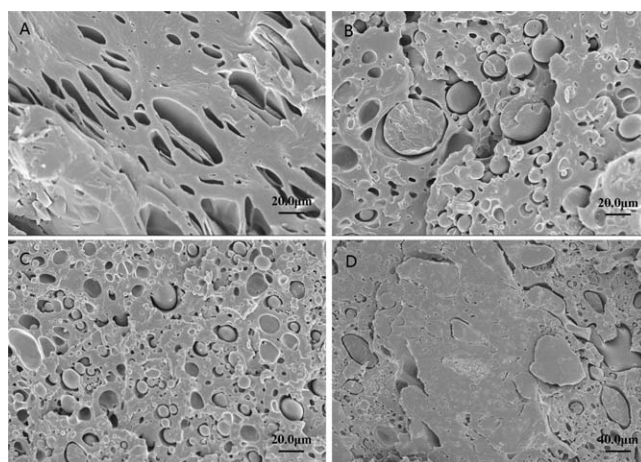


Figure 6. The tensile fracture surfaces SEM images of PA6/PSU and PA6/PSU-*b*-PA6/PSU blends. (A) Sample B0: PA6/PSU-*b*-PA6/PSU = 10/0/90; (B) Sample B2: PA6/PSU-*b*-PA6/PSU = 9.8/2/88.2; (C) Sample B10: PA6/PSU-*b*-PA6/PSU = 9/10/81; (D) Sample C10: PA6/PSU-*b*-PA6/PSU = 18/10/72.

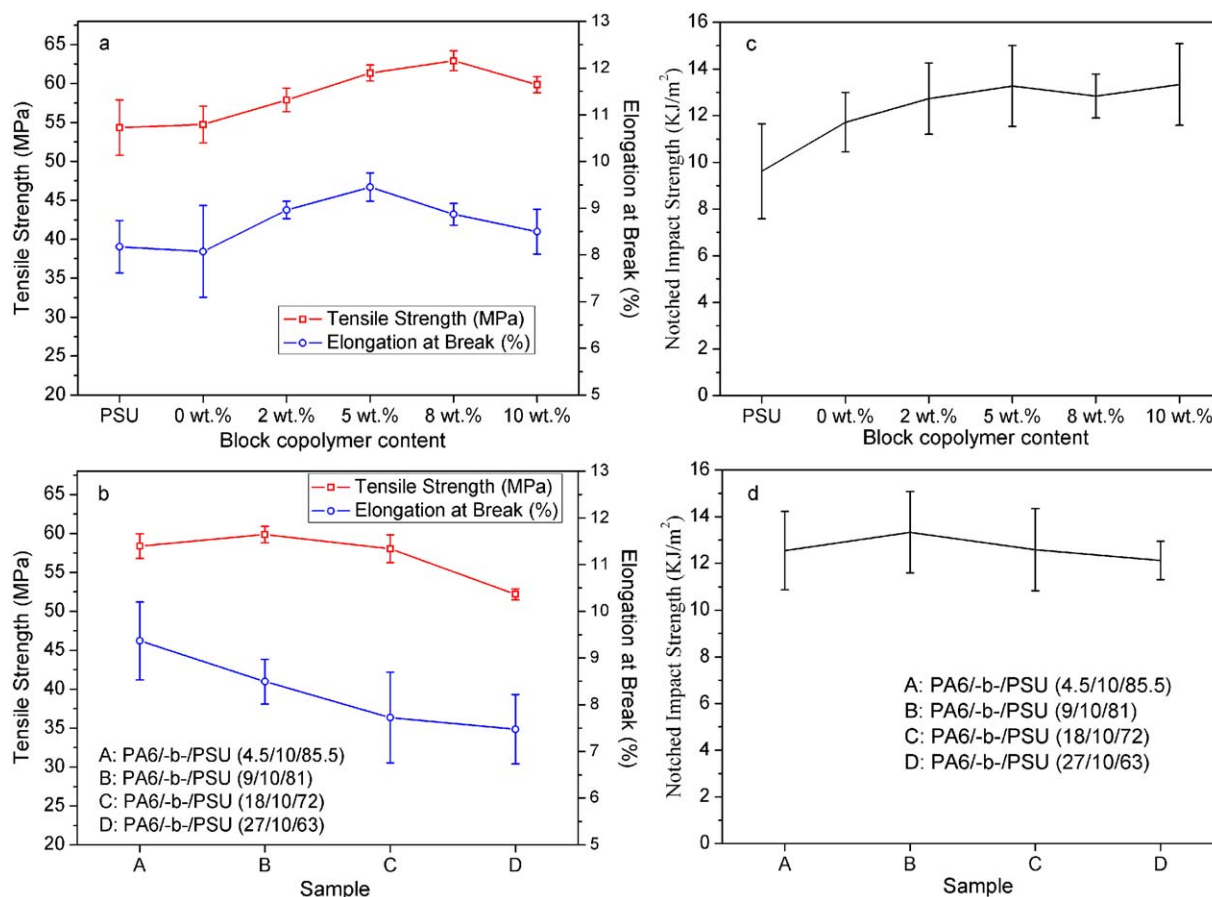


Figure 7. Mechanical properties of PSU, PA6/PSU, and PA6/PSU-*b*-PA6/PSU. (a, c) with different content of block copolymer in blends (PA6/PSU = 1/9); (b, d) with different mass ratio of PA6/PSU (10 wt % block copolymer). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

two phases. Due to the different polarities between two components, the blend will lead to the formation of unstable morphologies and macro-phase separation.⁴⁵

The residual PA6 phases appeared in the PSU matrix after the addition of block copolymer [Figure 6(B,C)]. It showed a trend that the average size of PA6 particle was reduced as copolymer content increased, indicating the better compatibility between the phases. The incorporation of 10 wt % copolymer led to a strong decrease of particle size [Figure 6(C)]. The copolymer was believed to play a crucial role in reducing interfacial tension and preventing particle coalescence between the immiscible polymer phases resulting in a fine dispersion.^{31–34} Additionally, with further addition of PA6 content, the size of the particles increased, and PA6 aggregates appeared [Figure 6(D)]. The formation of aggregated PA6 domains impaired the interfacial adhesion of two phases.

Mechanical Properties of Blends

The mechanical parameters (tensile strength, elongation at break, and notched impact strength) of PSU, PA6/PSU and PA6/PSU-*b*-PA6/PSU were presented in Figure 7. It could be found that the pure PSU and PA6/PSU binary blend exhibited the similar mechanical properties. The introduction of PA6 made a negligible contribution to the improvement of PSU's

mechanical properties (or even worse to elongation at break), which was attributed to the poor interfacial adhesion between PSU and PA6 phases. By contrast, the addition of copolymer caused a variation in its mechanical properties. The results in Figure 7 indicated that the blends with block copolymer were capable to withstand higher stress in tensile mode. Compared with the pure PSU, the blend containing 8 wt % copolymer induced an increase of 15% in tensile strength and the blend containing 5 wt % copolymer led to an increase of 16% in elongation at break.

Additionally, the copolymer concentration had a positive effect on the notched impact strength of blends [Figure 7(c)]. Notched impact strength improved by 40% compared to that of pure PSU with the addition of 10 wt % copolymer. The ductility of PSU was improved by the introduction of polyamide and block copolymer owing to the presence of flexible methylene chain in the polyamide. The improvement in mechanical properties might be attributed to the better interfacial adhesion and phase dispersion in the presence of block copolymer, facilitating the stress transfer between the phases.⁴⁶

The effect of PA6 content on the mechanical properties of blends was showed in Figure 7(b,d). It was found that the tensile strength and notched impact strength of blends decreased

with more addition of PA6 component. The reduction of the ductility of blends resulted from the presence of aggregated PA6 domains, leading to the stress concentration of material. Sample B10 (PA6/PSU-*b*-PA6/PSU = 9/10/81) showed the maximum tensile strength as well as the maximum notched impact strength over other blends.

CONCLUSIONS

PSU-PA6 block copolymer was synthesized via caprolactam anionic polymerization. And PA6/PSU-*b*-PA6/PSU ternary blends with different content of PA6 and PSU-PA6 block copolymer were prepared by extrusion. Fluidity, thermal and mechanical properties of blends were investigated. The results indicated that the compatibility of the PA6 and PSU components in the blends was improved with the incorporation of copolymer. Fluidity of PSU improved obviously with the introduction of PA6 and block copolymer. When the mass ratio of PA6/PSU-*b*-PA6/PSU was 9.5/5/85.5, tensile strength and elongation at break of the blends led to an increase of 15% compared with the pure PSU. Although thermal decomposition temperature of PSU dropped with the addition of PA6 and copolymer, ternary blends remained decent thermal stability.

Compared with the pure PSU, MFI of the blend was improved above 10 times when 10 wt % PA6 and block copolymer was introduced. Moreover, strength and toughness of the blend were enhanced 4–10% without compromising the heat resistance of PSU obviously. Extrusion temperature of PSU was declined from 302–332°C to 263–293°C and injection temperature was declined from 320–360°C to 300–330°C. In general, the harsh processing conditions of PSU were relieved without damaging its performance.

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REFERENCES

- Zodrow, K.; Brunet, L.; Mahendra, S.; Li, D.; Zhang, A.; Li, Q. L.; Alvarez, P. J. *J. Water. Res.* **2009**, *43*, 715.
- Matsumoto, K.; Higashihara, T.; Ueda, M. *Macromolecules* **2009**, *42*, 1161.
- Dang, Z. M.; Zhou, T.; Yao, S. H.; Yuan, J. K.; Zha, J. W.; Song, H. T.; Li, J. Y.; Chen, Q.; Yang, W. T.; Bai, J. *Adv. Mater.* **2009**, *21*, 2077.
- Wang, J. H.; Li, S. H.; Zhang, S. B. *Macromolecules* **2010**, *43*, 3890.
- Zhu, J. H.; Wei, S. Y.; Chen, X. L.; Karki, A. B.; Rutman, D.; Young, D. P.; Guo, Z. H. *J. Phys. Chem. C.* **2010**, *114*, 8844.
- Milliman, H. W.; Miguel, S. S.; Arostegui, A.; Schiraldi, D. A. *J. Appl. Polym. Sci.* **2012**, *125*, 2914.
- Li, X. H.; Guo, Y. C.; Zhang, J.; Zhang, L. *J. Appl. Polym. Sci.* **2013**, *128*, 2994.
- Wu, Z. W. *Special engineering plastics and its application*. Beijing: Chemical Industry Press, **2011**; p 152.
- Shi A. F.; Gong, Y. B. *Manual of engineering plastics*. Shanghai Scientific and Technical Publishers: Shanghai, **2001**; p 712.
- Miyatake, K.; Chikashige, Y.; Higuchi, E.; Watanabe, M. *J. Am. Chem. Soc.* **2007**, *129*, 3879.
- Park, J. Y.; Acar, M. H.; Akthakul, A.; Kuhlman, W.; Mayes, A. M. *Biomaterials* **2006**, *27*, 856.
- Celik, E.; Park, H.; Choi, H. *Water. Res.* **2011**, *45*, 274.
- Golovoy, A.; Kozlowski, M.; Narkis, M. *Polym. Eng. Sci.* **1992**, *32*, 854.
- Magagnini, P. L.; Paci, M.; La Mantia, F. P.; Surkova, I. N.; Vasnev, V. A. *J. Appl. Polym. Sci.* **1995**, *55*, 461.
- Hong, S. M.; Kim, B. C.; Kim, K. U.; Chung, I. *J. Polym. J.* **1991**, *23*, 1347.
- García, M.; Eguiazábal, J. I.; Nazábal, J. *J. Macromol. Sci. Phys.* **2004**, *B43*, 489.
- Ran, R. C.; Jiang, S. *J. China Plast. Ind.* **1982**, *02*, 29.
- Chen, X. Y.; Lu, A.; Wang, G.; Yu, X. *J. China Plast. Ind.* **2007**, *35*, 241.
- Nevin, G. K.; Ayse, A. *Compos. B* **2013**, *51*, 270.
- Han, K.; Zhang, X.; Zhan, S. *J. Macromol. Sci. Phys.* **2011**, *50*, 225.
- Charoensirisomboon, P.; Weber, M. *Polymer* **2001**, *42*, 7009.
- Gaymans, R. J.; Borggreve, R. J. M.; Spoelstra, A. B. *J. Appl. Polym. Sci.* **1989**, *35*, 47.
- Deng, Z.; Zhang, Y.; Ding, Y. J.; Luo, C. Q. *Acta Polym. Sin.* **1984**, *8*, 288.
- McGrath, J. E.; Robeson, L. M.; Matzner, M. *Polym. Prepr.* **1973**, *14*, 1032.
- Charoensirisomboon, P.; Chiba, T.; Solomko, S. I.; Inoue, T.; Weber, M. *Polymer* **1999**, *40*, 6803.
- Maréchal, Ph.; Chiba, T.; Inoue, T.; Weber, M.; Koch, E. *Polymer* **1998**, *39*, 5655.
- Omonov, T. S.; Harrats, C.; Moussaif, N.; Groeninckx, G.; Sadykov, Sh. G.; Ashurov, N. R. *J. Appl. Polym. Sci.* **2004**, *94*, 2538.
- Du, L. B.; Yang, G. S. *J. Appl. Polym. Sci.* **2008**, *108*, 3419.
- Dizman, C.; Kahveci, M. U.; Yagci, Y. *Polym. Bull.* **2013**, *70*, 2097.
- Jiang, Y.; Shi, H. Y.; Cai, M.; Liang, Y.; Li, B.; Zhang, H. W.; Song, R. G. *J. Appl. Polym. Sci.* **2013**, *129*, 247.
- Charoensirisomboon, P.; Inoue, T.; Weber, M. *Polymer* **2000**, *41*, 4483.
- Charoensirisomboon, P.; Chiba, T.; Inoue, T.; Weber, M. *Polymer* **2000**, *41*, 5977.
- Charoensirisomboon, P.; Inoue, T.; Weber, M. *Polymer* **2000**, *41*, 6907.
- Charoensirisomboon, P.; Inoue, T.; Solomko, S. I.; Sigalov, G. M.; Weber, M. *Polymer* **2000**, *41*, 7033.
- Ding, Y. J.; Tian, X. S. *Polym. Commun.* **1980**, *5*, 297.
- Koning, C. E.; Fayt, R.; Bruls, W.; Vondervoort, Lvd.; Rauch, T.; Teysse, P. *Macromol. Symp.* **1993**, *75*, 159.

37. Kaufhold, W.; Schnablegger, H.; Kumpf, R. T.; Pielarzik, H.; Cohen, R. E. *Acta. Polym.* **1995**, *46*, 307.
38. Silverstein, R. M.; Bassler, G. C.; Morrill, T. C. *Spectrometric Identification of Organic Compounds*. New York: John Wiley & Sons, Inc, **1981**; p 95.
39. Rotter, G.; Ishida, H. *J. Polym. Sci. Part B: Polym. Phys.* **1992**, *30*, 489.
40. Otsuki, T.; Kakimoto, M. A.; Imai, Y. *J. Appl. Polym. Sci.* **1990**, *40*, 1433.
41. Kim, K. J.; Hong, D. S.; Tripathy, A. R.; Kyu, T. *J. Appl. Polym. Sci.* **1999**, *73*, 1285.
42. Huang, P.; Zheng, S. X.; Huang, J. Y. *Polymer* **1997**, *38*, 5565.
43. Ahn, T. O.; Hong, S. C.; Jeong, H. M.; Kim, J. H. *Polymer* **1997**, *38*, 207.
44. Luo, Y. R. *Chemical Bond Energy Data Handbook*; Science press: China, **2005**; p 257.
45. Laoutid, F.; Estrada, E.; Michell, R. M.; Bonnaud, L. *Polymer* **2013**, *54*, 3982.
46. Gao, C.; Zhang, S. L.; Li, X. J.; Zhu, S. S.; Jiang, Z. H. *Polymer* **2014**, *55*, 119.