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ARTICLE

Phase Morphology, Mechanical and Thermal Properties of Poly(trimethylene terephthalate) and Poly(ethylene terephthalate) Blends

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ABSTRACT

Poly(ethylene terephthalate)/poly(trimethylene terephthalate) (PET/PTT) blends were prepared and their phase morphology, mechanical and thermal properties were investigated by scanning electron microscopy (SEM), polarized optical microscopy (POM), universal material testing machine, differential scanning calorimetry (DSC), wide-angle X-ray diffraction (WAXD), respectively. The glass transition and SEM results suggest apparently that poly(ethylene terephthalate) and poly(trimethylene terephthalate) have good miscibility at amorphous state. The blends with more poly(ethylene terephthalate) content less likely undergo a melting/recrystallization process during DSC heating scan. In blends, poly(ethylene terephthalate) component with higher supercooling degree will crystallize first and then the crystallites of poly(ethylene terephthalate) will be the nucleating agents for poly(trimethylene terephthalate), which greatly improves the crystallization rate of poly(trimethylene terephthalate). Because of the interaction between poly(ethylene terephthalate) and poly(trimethylene terephthalate), there are much smaller spherulites formed in blends with increasing poly(ethylene terephthalate) component. The blend with more poly(ethylene terephthalate) contents has larger tensile strength and modulus.

KEYWORDS

Poly(ethylene terephthalate). Poly(trimethylene terephthalate), Blends, Crystallization, Morphology, Mechanical property.

INTRODUCTION

Polymer blending is a widely used way to extend the application fields of polymers, which is straightforward, versatile and relatively inexpensive for creating a new polymeric material. It has been proved that many physical and mechanical properties of the polymers can be significantly improved [1]. Several published articles related to various aspects of binary blends of polyesters are available in the literatures [2-9].

Poly(trimethylene terephthalate) (PTT) has been receiving much attention because of its outstanding properties, such as good tensile behaviour, resilience, outstanding elastic recovery, dyeability, *etc*. Moreover, it takes an unusual combination of the topping properties of poly(ethylene terephthalate) and processing characteristics of poly(butylene terephthalate) (PBT). These characteristics make poly(trimethylene terephthalate) highly suitable for being used in fiber, film and engineering thermoplastics applications [10,11]. There has been much research on the morphology [12-14], crystallization kinetics [13,14] and rheological properties [15] of poly(trimethylene terephthalate) and its blends or copolymers with other polyesters [16-23].

In the present study, various PET/PTT blends with different poly(trimethylene terephthalate) composition were prepared by co-rotating twin screw extruder under the same processing conditions. In order to decrease a maximum of the polycondensation reaction between poly(ethylene terephthalate) (PET) and poly(trimethylene terephthalate) (PTT), the limited time of melt-blending was within 2 min. The miscibility, crystal morphology, thermal and mechanical properties were investigated by using differential scanning calorimetry (DSC), scanning electron microscopy (SEM), polarized optical microscopy (POM), wide-angle X-ray diffraction (WAXD) and mechanical measurements.

EXPERIMENTAL

Poly(trimethylene terephthalate) homopolymer used in this study was supplied in pellet form by Shell Chemicals (USA) with weight-average molecular masses (M_w) of 100,400 and molecular polydispersity (M_w/M_n) of 2.1 and its intrinsic viscosity was 0.92 dL/g measured in a phenol/tetrachloroethane solution at 25 °C. Poly(ethylene terephthalate) homopolymer was supplied in pellet form by Tianjin Petrochemical Co. with M_w of 70,000 and M_w/M_n of 2.4 and its intrinsic viscosity was 0.66 dL/g measured in phenol/tetrachloroethane at 25 °C.

Binary blends preparation: The materials were dried in a vacuum oven at 140 °C for 12 h before the preparing blends. The dried pellet of poly(trimethylene terephthalate) and poly(ethylene terephthalate) were mixed together with different weight ratios of PET/PTT as follows: B1:0/100; B2:20/80; B3:40/60; B4:60/40; B5:80/20; B6:100/0 and then melt-blended for about 2 min in a ZSK-25WLE WP self-wiping, co-rotating twin-screw extruder, operating at a screw speed of 100 rpm and at a die temperature of 280 °C. The resultant blends' ribbons were cooled in cold water, cut up and re-dried before being used in measurements.

Differential scanning calorimetry: The crystallization and subsequent melting behaviours of various binary blends were performed by the Perkin-Elmer Diamond DSC instrument that was calibrated with indium prior to performing the measurements; the weights of all samples were approximately 6.0 mg. The sample was heated to 280 °C at 100 °C/min under a nitrogen atmosphere, held for 5 min and then cooled to -50 °C at a constant cooling rate of 100 °C/min to obtain the amorphous state (the Perkin-Elmer Diamond DSC instrument can ensure the rushed temperature lower than 0.1 °C at scanning rate of 100 °C/min, so little error and its effect on crystallization can be ignored). The amorphous blend was heated to 280 °C at a heating rate of 10 °C/min, held for 5 min and then cooled to 0 °C at a constant cooling rate of 10 °C/min; the heating and cooling processes were recorded.

Wide-angle X-ray diffraction: The blend pellets were compression-molded between metal plates at 280 °C for 5 min, quenched by ice water, dried at room temperature and then cut into strips (15 mm × 15 mm × 1 mm). A group of strips were put into an oven annealed at 120 °C for different times and another group of strips were annealed at 195 °C for different times and then final crystallized samples was immediately quenched into ice water and then dried for the WAXD measurements. WAXD patterns were recorded on a Rigaku D/MAX-2500UBZ+/PCD diffractometer system. Nickel-filtered CuK_α (λ = 0.15418 nm) radiation generated at 40 kV and 50 mA was used. The diffraction patterns were recorded from 2θ scans in the range of 5-50° at a scanning speed of 10 °/min using a step size of 0.02°.

Polarized optical microscopy: Polarized optical microscopy (Yongheng 59XA, China) with a digital camera system (Panasonic wv-CP240, Japan) was used for observation of the crystallites. Samples were prepared by sandwiching a tiny pellet of PET/PTT blend between two glass plates with a film thickness of about 200 ìm, compressing at 280 °C for 5 min and then annealing in an oven at 200 °C for 3 h and then quenched by ice-water and dried at room temperature.

Scanning electron microscopy characterizations The scanning electron microscopy (KYKY-2800B, Travor-Norther Co., USA) was also used to study the blend's phase morphology at an accelerating voltage of 25 kV. The sample was selected from the ribbons that were extruded from the extruder and cooled by the cold water. Prior to the observations, the fractured surface of the above samples was sputter-coated with gold.

Mechanical characterizations: The blend pellets were made into the standard splines using a microinjection molding machine (SZ-15, Wuhan Ruiming Machinery, China). Tensile strength testing was performed with a universal testing machine (WSM-20, Changchun Intelligent Instrument Co., Ltd.) with a crosshead speed of 10 mm/min at room temperature (25 °C) according to the ASTM D638-2000.

RESULTS AND DISCUSSION

Miscibility of poly(ethylene terephthalate) and poly(trimethylene terephthalate): Fig. 1 shows the thermograms of the pure poly(trimethylene terephthalate), poly(ethylene terephthalate) and their binary blends at a heating rate of 10 °C/min and the melting parameters are listed in Table-1. It is clearly seen from Fig. 1 and Table-1 that, for pure poly(ethylene terephthalate) and poly(trimethylene terephthalate), poly(ethylene terephthalate) exhibits a higher glass transition temperature (T_g) and melting peak (T_m) at about 77.4 °C and 253.2 °C, followed by poly(trimethylene terephthalate) with $T_g = 45.7$ °C and $T_m = 227.6$ °C. Generally, the value of T_g and T_m relates to the gentleness of the molecular chains that is determined by the number of methylene groups. The larger the number of methylene groups, the lower of the T_g and T_m. Additionally, the phenyl and two methylene groups in poly(ethylene terephthalate) molecular chains exist in a plane with a hackle shape, which made it more difficult to rotate than poly(trimethylene terephthalate) molecular chains [24]. Moreover, two cold-crystallization peaks are observed at about 72.1 °C and 151.9 °C for poly(trimethylene terephthalate) and poly(ethylene terephthalate), respectively, suggesting that poly(trimethylene terephthalate) is more flexible than poly(ethylene terephthalate) to crystallize at a lower temperature.

TABLE-1
KINETIC PARAMETERS OF HEATING AND COOLING PROCESS FOR VARIOUS SAMPLES

Sample	Heating process							Cooling process			
	T_g (°C)	T_{re} (°C)	ΔH_{re} (J/g)	T_{mI} (°C)	ΔH_{mI} (J/g)	T_{mII} (°C)	ΔH_{mII} (J/g)	T_{pI} (°C)	T_{pII} (°C)	ΔH_{cI} (°C)	ΔH_{cII} (J/g)
B1	45.7	–	–	227.6	49.4	–	–	176.4	–	-50.4	–
B2	56.9	203.2	-8.42	227.6	42.9	252.4	5.3	192.4	–	-46.0	–
B3	58.8	203.1	-7.74	227.6	39.4	252.4	6.2	193.2	–	-44.9	–
B4	64.6	202.6	-4.67	225.6	17.7	254.8	27.1	198.5	208.3	-9.8	-16.4
B5	68.8	202.5	-1.94	223.4	14.2	255.6	30.4	198.5	212.9	-6.7	-30.0
B6	77.4	–	–	–	–	253.2	36.3	–	192.6	–	-23.5

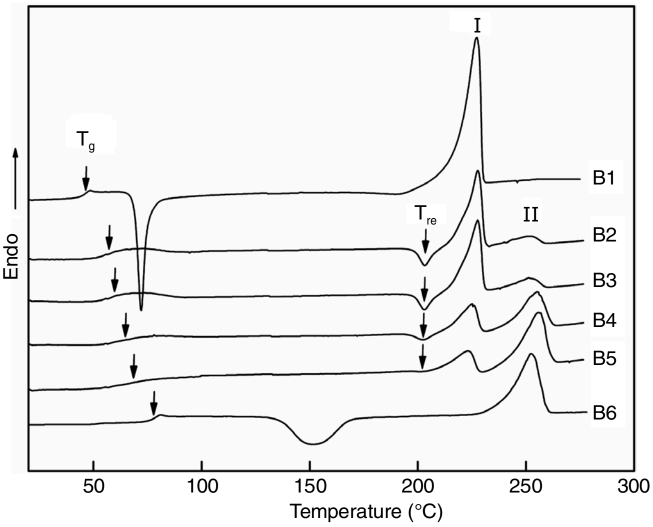

Fig. 1. Melting DSC curves of six samples

A phenomenon worth mentioning is the T_g for each binary blend of B2-B5 that a single and composition-dependent T_g is located between those of the pure components. In other words, the value of T_g increases monotonously from 65 to 73 °C with increasing the components of poly(ethylene terephthalate) from 20 to 80 wt %, although the transition is not as sharp as those of the pure poly(trimethylene terephthalate) and poly(ethylene terephthalate). The dependence of T_g on the blend composition can be predicted by Gordon-Taylor [25] and Fox [26] equations. In this paper, Fox equation is employed to describe the relationship between T_g and blend composition and the result is shown in Fig. 2.

$$\frac{1}{T_g} = \frac{w_A}{T_{gA}} + \frac{w_B}{T_{gB}} \tag{1}$$

where w_A and w_B are the weight fractions of the components of poly(trimethylene terephthalate) and poly(ethylene terephthalate), respectively. The T_{gA} and T_{gB} are the glass temperature values of the pure poly(trimethylene terephthalate) and poly(ethylene terephthalate), respectively. As seen from Fig. 2, the T_g value is in linear relationship with the poly(ethylene terephthalate) content. These results suggest a good miscibility between the two polymers in the amorphous state [27], which is also consistent with the results [4].

However, no cold-crystallization peak is observed in the DSC curves for all the binary blends (B2-B5) in Fig. 1. Although there is a good miscibility between poly(trimethylene terephthalate) and poly(ethylene terephthalate), some small domains of two polymers could exist due to micro phase separation. During the heating process, cold-crystallization (crystallization from the glassy state in the lower temperature region) will occur first in poly(trimethylene terephthalate) micro domains, followed by poly(ethylene terephthalate) crystallization in poly(ethylene terephthalate) micro domains because the chain mobility of poly(trimethylene terephthalate) is higher than that of poly(ethylene terephthalate). That is, poly(trimethylene terephthalate) chain segments diffuse to the front of the poly(trimethylene terephthalate) crystals to crystallize and poly(ethylene terephthalate) chains segments diffuse out from the front of poly(trimethylene terephthalate) crystals and the reverse process would take place in the front of the poly(ethylene terephthalate) crystals. The cold-crystallization would be controlled by diffusion rate and the crystallization was gradually completed in a broad temperature range during DSC scan, therefore, the cold-crystallization could not be observed in B2-B5.

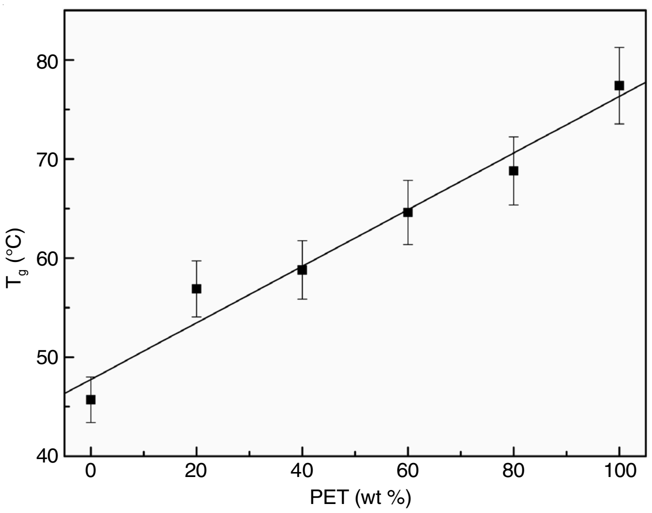

Fig. 2. T_g values of six samples as a function of blend composition. The solid line represents fits of the Fox equation.

The fracture surfaces of different blends are shown in Fig. 3(a-d). As shown in all the SEM micrographs, the fracture surface is so smooth that no dispersed phase can be observed in the blends, suggesting a good miscibility between the two polymers (at least on the micron scale) when they were extruded from the extruder and cooled by the cold water. This result is also consistent with the T_g conclusions.

Furthermore, an obvious recrystallization peak (T_{re} in Fig. 1) is observed in each curve of B2-B5. However, the peak intensity

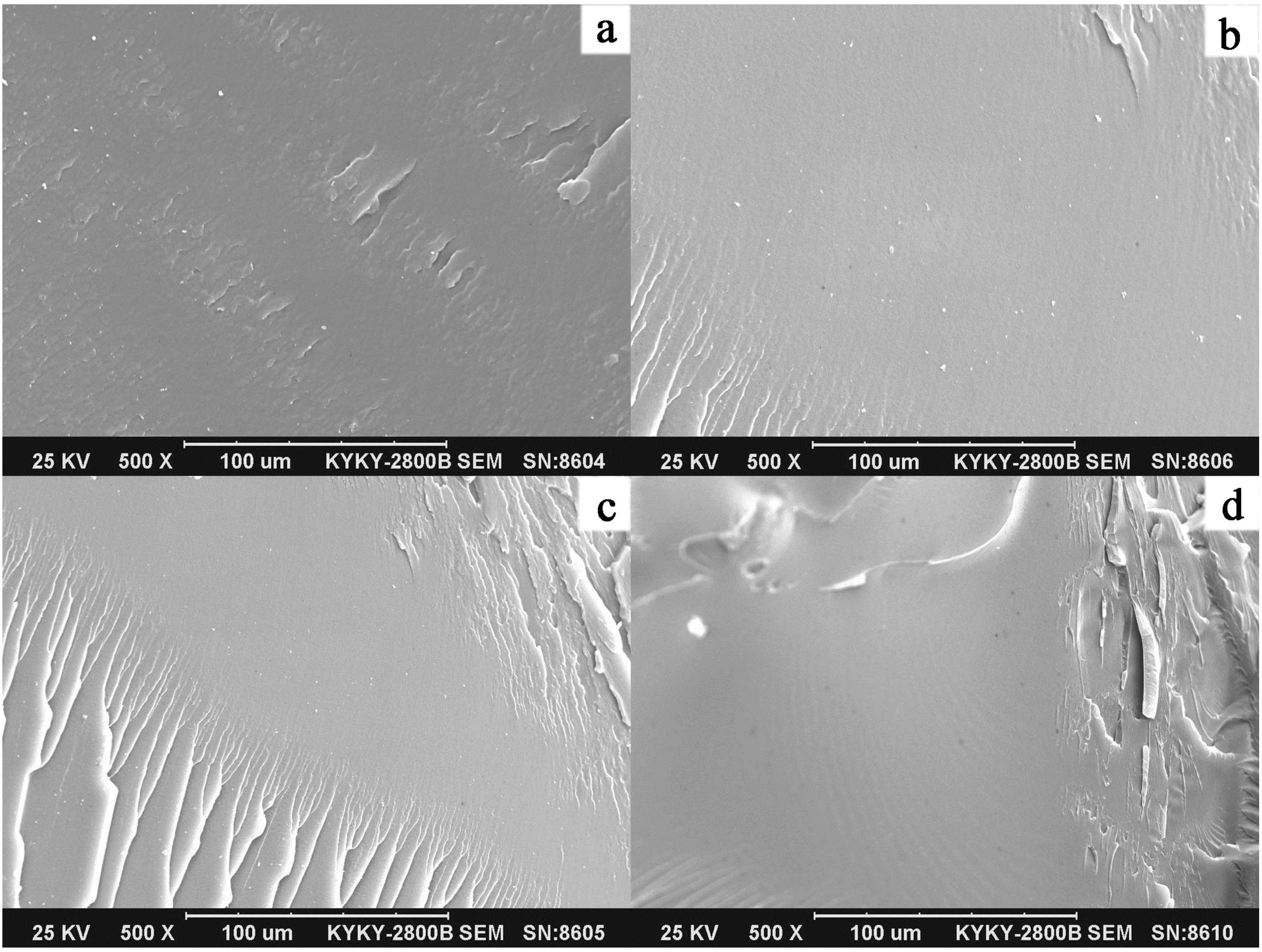

Fig. 3. SEM micrographs of the impact fracture surface of different blends, (a) B2, (b) B3, (c) B4 and (d) B5

of T_{re} becomes weaker and enthalpy of recrystallization (ΔH_{re} in Table-1) decreases as poly(ethylene terephthalate) component increases in blends. This result indicates that the blends with more poly(ethylene terephthalate) content less likely undergo a melting/recrystallization process during DSC heating scan [12]. Because the poly(ethylene terephthalate) crystal has a higher T_m and its molecular chain segments have a lower mobility, the reorganization or recrysta-llization becomes slower or difficult when the poly(ethylene terephthalate) component increases.

Melting, cold-crystallization and melt-crystallization behaviours: As shown in Fig. 1 and Table-1, the thermograms of blends exhibit two melting peaks: peak I at a lower temperature and peak II at a higher temperature, relatively. At this heating rate, the intensity of peak I and ΔH_{cI} decreases with decreasing of the poly(trimethylene terephthalate) component and that of peak II and ΔH_{cII} increases with increasing of the poly(ethylene terephthalate) component. Therefore, peak I at the lower temperature may be corresponding to the melting of the poly(trimethylene terephthalate) component and peak II at the higher temperature is the melting of the poly(ethylene terephthalate) component.

For further investigation of the cold-crystallization of the blends, WAXD is employed. Fig. 4(a) and (b) show the WAXD diffraction curves of B3 samples after isothermal annealed at 195 and 120 °C for different times, respectively. In Fig. 4(a), the diffraction curves of amorphous sample (B3, 0 min) show without sharp diffraction peaks, indicating that both poly-(ethylene terephthalate) and poly(trimethylene terephthalate) components in the blend are not crystallized. As the annealing time is 2 min, some weak diffraction peaks emerge. On prolonged annealing time to 4 min, the weak diffraction peaks become sharper. On further prolonged annealing time to 6 and 10 min, these peaks' intensity becomes sharper and stronger. Comparing the diffraction peaks of B3 annealed for 2-10 min with those of the pure poly(ethylene terephthalate) and poly-(trimethylene terephthalate) annealed at 195 °C for 10 min, they seems to be the combination peaks of pure poly(trimethylene terephthalate) and poly(ethylene terephthalate) peaks. The relative intensity of the specific peaks of poly-(trimethylene terephthalate) at about 15.4°, 17.1°, 19.6°, 23.7° and 24.9° and those of poly(ethylene terephthalate) at about 21.4°, 22.6° and 25.9° increase gradually with increasing annealing time; while the specific peaks of poly(ethylene terephthalate) at 16.2° and 17.4° are overlapped with the peaks of poly(trimethylene terephthalate). On the other hand, the specific peaks' intensity of poly(trimethylene terephthalate) is much stronger than those of poly(ethylene terephthalate), mainly because poly(trimethylene terephthalate) is the major component in B3 blend. These results suggest that the coexistence of the two type crystals of

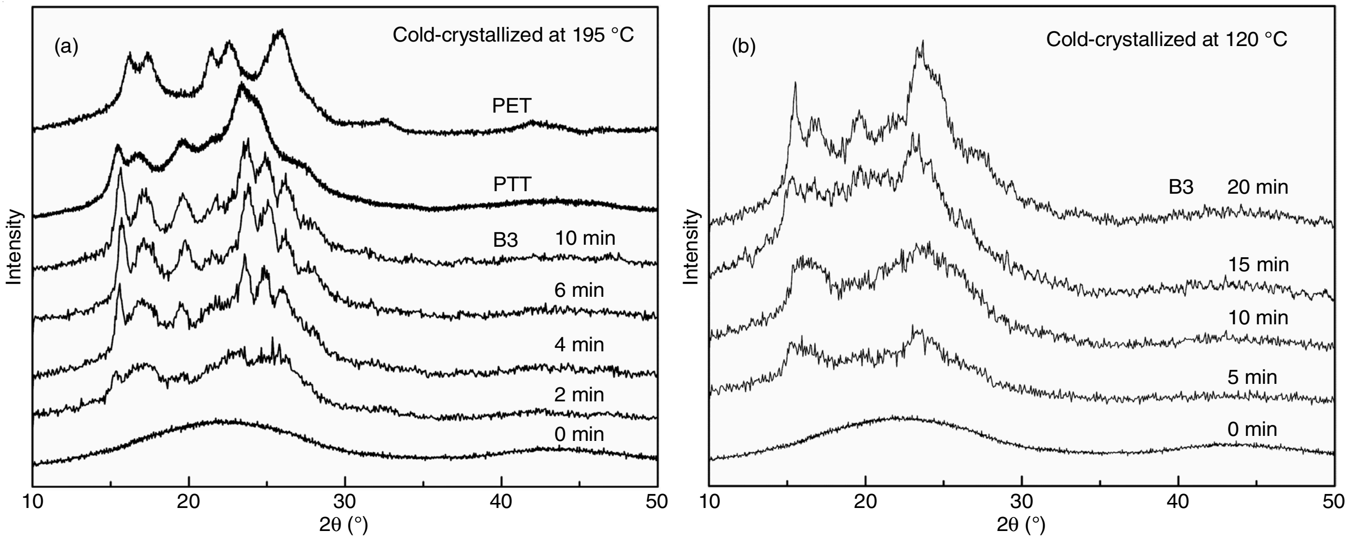

Fig. 4. WAXD profiles of B3 blend cold-crystallized at (a) 195 °C and (b) 120 °C for different times

poly(ethylene terephthalate) and poly(trimethylene terephthalate) in blend that formed simultaneously at higher annealing temperature of 195 °C.

There are some differences in diagram of Fig. 4(b) compared with Fig. 4(a), in which the intensity of the specific diffraction peaks of B3 becomes stronger with increasing annealing time from 0 to 20 min at annealing temperature of 120 °C. Comparing these diffraction curves with those of the pure crystalline poly(trimethylene terephthalate) and poly(ethylene terephthalate) [Fig. 4(a)], it is found that these specific peaks are similar to those of the pure poly(trimethylene terephthalate) and there is no specific peaks of poly(ethylene terephthalate) emerge. This phenomenon is dependent on the cold-crystallization temperature of 120 °C, which is much higher than the cold-crystallization temperature of the pure poly(trimethylene terephthalate) (as shown in Fig. 1, 63-85 °C) but lower than the cold-crystallization temperature of the pure poly(ethylene terephthalate) (130-170 °C). At 120 °C, poly(trimethylene terephthalate) molecular chain segments are easy to crystallize, while poly(ethylene terephthalate) molecular chain segments are difficult to crystallize, so the crystal of poly(trimethylene terephthalate) plays a predominant role in the WAXD diagrams.

The melt-crystallization behaviours of six samples at the cooling rate of 10 °C/min are shown in Fig. 5 and the parameters are also summarized in Table-1. In the case of crystallization from the melt at a fixed cooling rate, the temperature where the crystallization occurs is mainly determined by supercooling and thus by melting temperature (T_m) of polymers. Poly(ethylene terephthalate) has a higher T_m and therefore, it can crystallize at a higher temperature, while poly(trimethylene terephthalate) has a lower T_m and requires further drop in temperature for the occurrence of crystallization.

For each binary blend of B2 (PET20/PTT80) and B3 (PET40/PTT60), as poly(ethylene terephthalate) component is the minor one in blends, only a single crystallization exotherm is observed in the DSC curve. Moreover, we can find that the crystallization peak temperature (192.4 °C for B2, 193.2 °C for B3) is much higher than that of pure poly(trimethylene terephthalate) (176.4 °C) while it is closed to that of pure poly(ethylene terephthalate) (192.6 °C). These results suggest that the minor component of poly(ethylene terephthalate) can greatly enhance the crystallization temperature of poly(trimethylene terephthalate). As the blend melt was cooled, the poly(ethylene terephthalate) component with higher supercooling degree will crystallize first and then the crystallite of poly(ethylene terephthalate) will be the nucleating agent for poly(trimethylene terephthalate), which will greatly improve the crystallization ability of poly(trimethylene terephthalate) at a higher temperature.

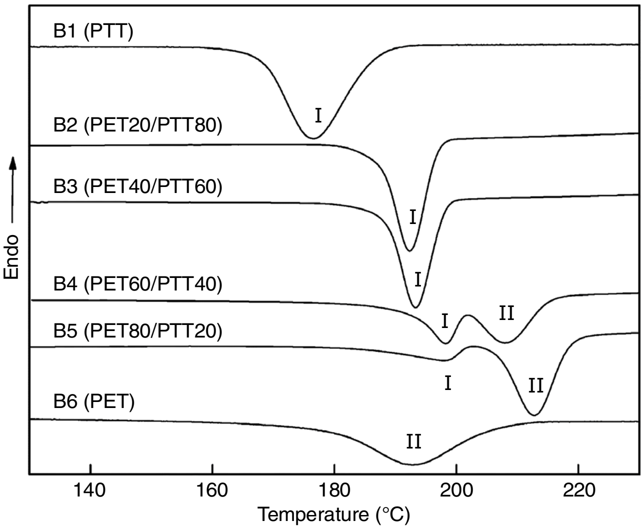

Fig. 5. Melt-crystallization DSC curves of six samples

While in the DSC curves of B4 and B5, as poly(ethylene terephthalate) component is the major one in blends, two crystallization exotherms are clearly observed: the primary II and subordinate I. Peak II and I are attributed to the crystallization behaviour of poly(ethylene terephthalate) and poly(trimethylene terephthalate), respectively, according to their crystallization temperatures. By careful observation, it is found that the temperature of peak II is much higher than that of neat poly(ethylene terephthalate) and peak I is much higher than that of neat poly(trimethylene terephthalate). The peak II shifting to higher temperature could be attributed to the diluent effect of poly(trimethylene terephthalate) in the melt, which increase the mobility of poly(ethylene terephthalate) molecular segments. The peak I shifting to higher temperature could be due to the crystallized poly(ethylene terephthalate), which act as nucleating agent and induce the crystallization of poly(trimethylene terephthalate) at subsequent higher temperature.

Crystal's morphology: Fig. 6(B1)-(B6) shows a series of polarized optical microscopy micrographs for poly(trimethylene terephthalate), poly(ethylene terephthalate) and the blends. Fig. 6(B1) reveals well-defined large spherulites with negative birefringence of pure poly(trimethylene terephthalate) and spherulites impinge on each other formed particular polygonal spherulites with the clear boundaries; While Fig. 6(B6) of pure poly(ethylene terephthalate) gives weaker maltese cross or birefringence, suggesting the poor crystal morphology because it has a weaker crystallization ability. As seen from Fig. 6(B2) to (B5), with increasing contents of poly(ethylene terephthalate), spherulites' size gradually decreases as well as with less perfection. From above results, it can be deduced that (1) poly(ethylene terephthalate) component can crystallize at a higher temperature and that increases the crystallization rate of the blends, therefore, more crystallites will form with smaller size in a limited space; (2) although poly(trimethylene terephthalate) has higher crystallization ability, the blend can only form poor crystal morphology due to the interaction between poly(trimethylene terephthalate) and poly(ethylene terephthalate).

Mechanical properties: The influences of poly(ethylene terephthalate) content on the mechanical properties of different blends are shown in Fig. 7. The tensile strength increases gradually with increasing poly(ethylene terephthalate) content, while the elongation at break decrease rapidly with increasing poly(ethylene terephthalate) content. Poly(ethylene terephthalate) has more rigid molecular chains than those of poly(trimethylene terephthalate), therefore, the blends show larger tensile strength and smaller elongation than that of poly(trimethylene terephthalate). The results also suggest that poly(ethylene terephthalate) and poly(trimethylene terephthalate) have good compatibility because of their gradually increased tensile strength.

Conclusion

The poly(ethylene terephthalate)/poly(trimethylene terephthalate) (PET/PTT) blends at amorphous state show good miscibility judged from the T_g and SEM results. The blend with more poly(ethylene terephthalate) content forms smaller or less perfect crystals, while it has higher tensile properties

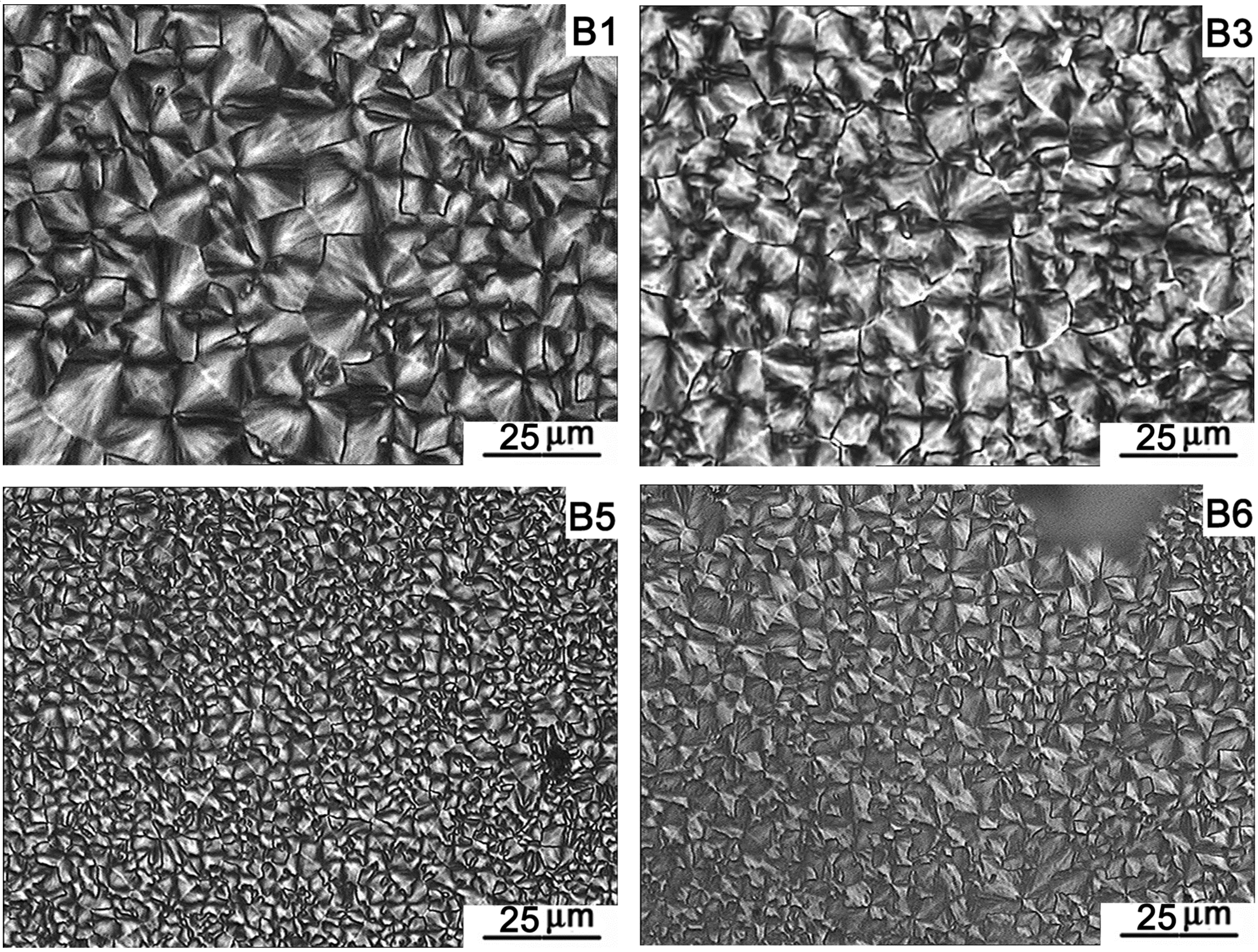

Fig. 6. Polarizing microscopic images of different crystallized samples

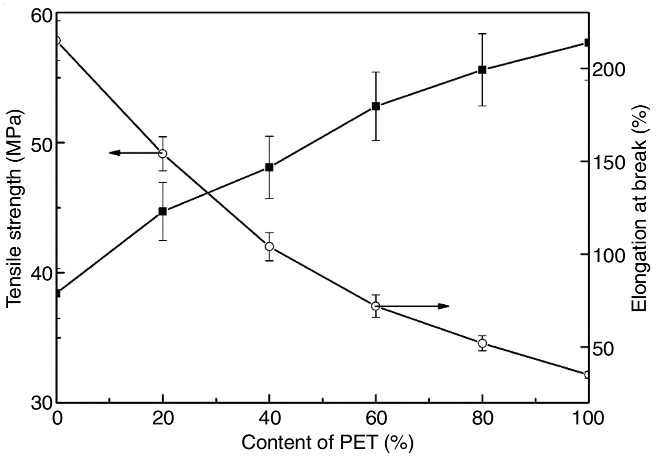

Fig. 7. Tensile strength and elongation at break of different samples

and less elongation at break. On the whole, the minor poly-(ethylene terephthalate) component in blends acting as nucleating agents improves the crystallization properties of the binary blend.

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