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## MODIFICATION OF POLYMERS

# Branching and Cross-linking of Poly(ethylene terephthalate) and Its Foaming Properties[1]

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**Abstract**—The branching and cross-linking of poly(ethylene terephthalate) were investigated using two chain extenders: glycidyl methacrylate-styrene copolymer (GS) and poly(butylene terephthalate)-GS (PBT-GS) in order to improve the melt viscosity and melt strength of poly(ethylene terephthalate). An obvious increase in torque evolution associated with chain extending, branching and cross-linking was observed during the process. The properties of modified poly(ethylene terephthalate) were characterized by intrinsic viscosity and insoluble content measurements, rheological and thermal analysis. The intrinsic viscosity and rheological properties of modified PET were improved significantly when using PBT-GS, indicating that PBT-GS should be a better chain extender. Good foaming of poly(ethylene terephthalate) materials were obtained using supercritical $CO_2$ as blowing agent. The average cell diameter and cell density were 61 μm and $1.8 \times 10^8$ cells/cm$^3$, respectively.

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## INTRODUCTION

Polyethylene terephthalate (PET) has been widely used as fiber, bottle, packaging film, foaming materials and engineering materials due to its excellent mechanical strength, good electrical insulation, good chemical resistance, high temperature resistance, recyclability and low cost [1, 2]. However, PET is limited in some applications such as blow-molding or foaming due to its inherently poor melt viscosity and melt strength caused by the low molecular weight and linear structure [3–5]. On the other hand, the use of recycled PET is becoming more and more important due to the requirement of environmental protection. One key challenge is the deterioration of mechanical performance due to the degradation of molecular weight after reprocessing of PET [6, 7]. Therefore, several methods have been employed to increase its melt viscosity and melt strength such as melt polycondensation, solid-state polymerization (SSP) and chain extension. Due to the high viscosity of melt polycondensation, it is rather difficult to remove $H_2O$ generated from reaction system and increase molecular weight of PET. The high investment and operation costs associated with SSP (the main approach) have rendered uneconomical in the application due to the rigorous reactional parameters [8, 9].

In the last decade, introducing multi-functional chain extenders have been proved an easy, fast reacting and suitable way for industrial production. Chain extensions using di- and multi-functional epoxides [10–12], diisocyanates [13–15], dianhydrides [16, 17], or bis(oxazoline)s [18–20] have shown the increase in melt viscosity and melt strength through reacting with terminal groups of PET. Diisocyanates can react with hydroxyl (preferentially) and carboxyl end group of PET, but its application is limited due to its toxicity. Torres found that chemical modification of recycled PET by diisocyanates lead to an increase in the molecular weight from $3.0 \times 10^4$ to $5.1 \times 10^4$ g/mol and the intrinsic viscosity from 0.60 to 0.84 dL/g [15]. The reaction mechanism of PET and dianhydride is the hydroxyl end group of PET attacks the anhydride group of dianhydride leading to coupling them and forming two carboxyl groups which can later reacted with PET [21]. However, the high carboxyl content leads to less thermal and hydrolytic stability. Incarnato [22] improved the intrinsic viscosity of virgin PET from 0.48 to 0.74 dL/g with pyromellitic acid dianhydride (PMDA) as chain extender and increased the branched degree with the increase of PMDA concentration. Bis-oxazolines possess a high reactivity toward

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[1] The article is published in the original.

**Table 1.** Recipes of modified PET samples

| PET samples | PET + GS | | PET samples | PET+(PBT-GS) | | GS content, wt % |
|---|---|---|---|---|---|---|
| | PET, g | GS, g | | PET, g | PBT-GS, g | |
| PET-GS-1 | 49.5 | 0.5 | PET-(PBT-GS)-1 | 46.5 | 3.5 | 1 |
| PET-GS-2 | 49.0 | 1.0 | PET-(PBT-GS)-2 | 43.5 | 6.5 | 2 |
| PET-GS-3 | 48.5 | 1.5 | PET-(PBT-GS)-3 | 40.0 | 10.0 | 3 |
| PET-GS-4 | 48.0 | 2.0 | PET-(PBT-GS)-4 | 36.5 | 13.5 | 4 |

carboxyl groups and lead to the formation of stable esteramide linkages [23]. Karayannidis [24] modified the recycled PET with 2,2'-(1,4-phenylene)bis(2-oxazoline) and increased its intrinsic viscosity from 0.78 to 0.85 dL/g. Cavalcanti and Nascimento [25, 26] investigated the modified PET using organic phosphites as modifiers. Their results showed that triphenyl phosphite was the best chain extender in this family under the optimum processing conditions with a temperature of 260°C and 1 wt % of chain extender. Epoxide has been identified as one of the suitable chain extenders due to its efficiency upon reacting with carboxyl (preferentially) and hydroxyl end groups [27]. Raffa et al. [28, 29] investigated the effect of multifunctional isocyanate and epoxy chain extenders on the molecular structure and rheological properties of PET. The reactivity of di-isocyanate was higher than di-epoxide according to the results. Meanwhile they successfully combined an epoxide commercial modifier with pentaerythritol to cause simultaneous hyperbranching and controlled chain scission of PET. Dhavalikar modified PET with a multifunctional epoxide-triglycidyl isocyanurate and demonstrated the long chain branched structure by rheological analysis [30]. Japon used tetraglycidyl diamino diphenyl methane which contains tetra-functional epoxy for obtaining branched molecules [10]. Xiao proved the presence of long-chain branching structure through the modification of PET by epoxide modifier according to rheological analysis [31]. Zhong obtained branched PETs with PMDA in in-situ polymerization-modification process and homogeneous cell structures with cell diameter range from 38–75 μm and cell density from $2.8 \times 10^7$–$5.8 \times 10^6$ cells/cm$^3$ [32].

In this paper, a chain extender a copolymer of glycidyl methacrylate and styrene (GS) containing epoxy groups, which can react with terminal groups of PET, was applied to prepare branched and cross-linked PET to improve its melt viscosity and melt strength. Considering the better processability and compatibility with PET in the melting reaction, a new chain extender named as poly(butylene terephthalate)-GS (PBT-GS) which was GS chemically bonded on PBT was also used. Intrinsic viscosity, rheological and thermal properties of the modified PET samples were investigated. Some PET samples were also foamed in the molten sate using supercritical carbon dioxide (ScCO$_2$) as blowing agent.

## EXPERIMENTAL

PET (BG80, intrinsic viscosity 0.76 dL/g) and PBT (S610SF NC010) were supplied from Sinopec Yizheng Chemical fibre Co., Ltd., China and DuPont Company, respectively. Chain extender GS and PBT-GS (containing 15% GS) were supplied from Fuyuan Plastics Science and Technology Co., Ltd., Shanghai, China. Phenol, tetrachloroethane and NaOH were all analytical purity and purchased from Lingfeng Chemical Reagent Co., Ltd., Shanghai, China.

PET resins were vacuum dried over 8 h at 140°C before use. Then a certain amount of dried PET resins and chain extender (Table 1) were added into a Torque Rheometer (Rheocord 300P + Rheomin 600P, Thermo Haake) at 270°C and 80 rpm. The reaction maintained for 6 min.

A high-pressure vessel loaded with PET samples was saturated by 15 MPa CO$_2$ at 260°C for 20 min, then decreased temperature to 230°C and stayed for 5 min, finally the pressure was immediately released. After the depressurization, the vessel was opened and cooled in the water-ice bath.

Molecular weight of GS was determined by GPC (Waters Model 1515) connected to a small-angle light scattering detector (DAWN EOS, Wyatt Technology). The measurement range of the molecular weight was from $10^3$ to $10^6$. THF was used as the eluent with a sample content of 5 mg/mL at 25°C.

FTIR spectrometer (Nicolet 5700) was used for analysis of GS and PBT-GS. The FTIR spectra were acquired by scanning the specimens in the wave number range from 400 to 4000 cm$^{-1}$.

The epoxy group content of the chain extender GS was measured by the hydrochloric acid/acetone titration. 0.3 g of the sample was added into a 150 mL conical flask with 30 mL acetone. After the sample was completely dissolved, 10 mL of hydrochloric acid (0.35 N) were added in the flask to react with epoxy groups. After 30 minutes, the solution was titrated with 0.1 N sodium hydroxide alcohol solution, using phenolphthalein as the indicator until the color turned red. Besides, 10 mL of hydrochloric acid should be

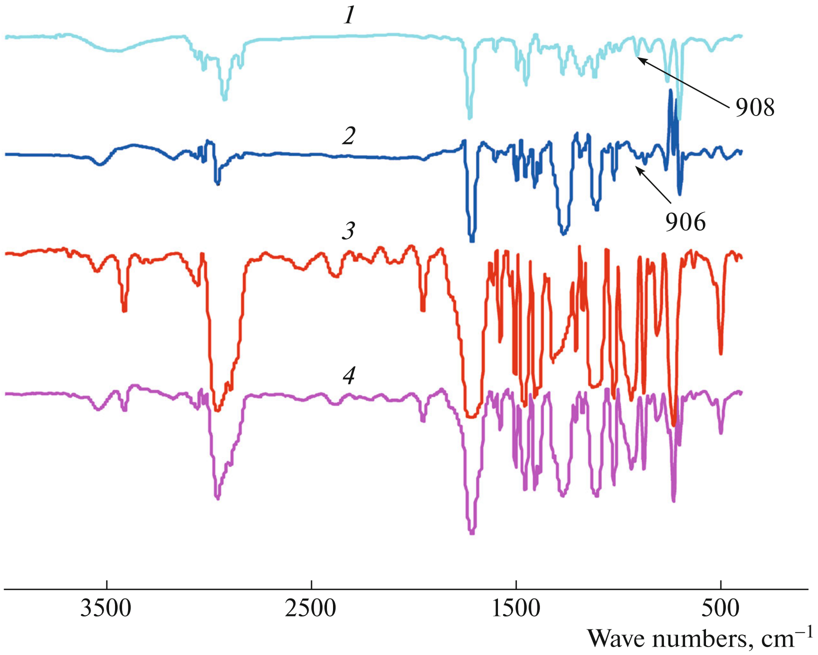


**Fig. 1.** (Color online) FTIR spectra of chain extenders: (*1*) GS, (*2*) (PBT-GS)-PBT, (*3*) PBT, (*4*) PBT-GS.

titrated as a blank control. The epoxy group content was calculated using Eq. (1):

$$E = \frac{C(V_1 - V_2)}{10m}, \quad (1)$$

where $V_1$, $V_2$ (mL) is the titrated volume of sodium hydroxide solution in control sample and the GS sample, respectively; $C$ (mol/L) is the concentration of sodium hydroxide alcohol solution; $m$ (g) is the mass of GS [33].

Intrinsic viscosity was measured using an Ubbelohde viscometer at 25°C in a mixed solvent of phenol and tetrachloroethane (60/40, w/w). The samples were stirred in the mixed solution at 120°C for 1 h for completely dissolving, then were cooled to room temperature and filtered. Intrinsic viscosity of each sample was calculated using the Schulz-Blaschke's Eq. (2):

$$\frac{\eta_{\text{rel}}}{c} = [\eta] + K_{SB}[\eta](\eta_{\text{rel}} - 1), \quad (2)$$

where $K_{SB}$ is 0.2345 at 25°C [34].

Some samples were partially dissolved in the above solvent mixture. After removing the insoluble part using filtration, washing these samples using solvent, drying them in a vacuum oven at 140°C for 8 h, and then measuring the intrinsic viscosity. Meanwhile, the insoluble content was calculated by the difference in the weight.

The rheological properties associated with melt viscosity and melt strength were characterized by a Rotational Rheometer (MARS III, Thermo Haake) under nitrogen atmosphere. The shear rheological tests were carried out with a parallel plate (20 mm in diameter, 1.0 mm in gap) and the oscillation frequency sweep mode. The linear domain of viscoelasticity was confirmed by dynamic strain sweep from 0.1 to 100% (260°C, 1 Hz). Complex viscosity ($\eta^*$), storage modulus ($G'$), and loss modulus ($G''$) as a function of frequency from 0.1 to 100 rad/s were recorded under 260°C at a constant strain amplitude.

Differential scanning calorimeter (Modulated DSC 2910) was used to analyze the thermal properties. All the samples were heated at a rate of 20 grad/min from 25 to 270°C and were held at this temperature for 3 min to eliminate heat history, then cooled to 25°C at 10 grad/min, stayed for 3 min and re-heated up to 270°C at 10 grad/min. The crystallization temperature $T_c$, crystallization enthalpy $\Delta H_c$, the melting temperature $T_m$, and melting enthalpy $\Delta H_m$, were obtained from the cooling scan and the second heating stage, respectively. The crystallinity ($\chi$) of PET was calculated by Eq. (3):

$$X = \frac{\Delta H_m}{\Sigma W_i \times \Delta H_{mi}} \times 100\%, \quad (3)$$

where $\Delta H_{mi}$ is the melting heat of 100% of crystallinity of PET and PBT, and the corresponding values are 140 and 142 J/g, respectively. $W_i$ represents the weight fraction of PET or PBT [35, 36].

The cell morphologies of the foamed PET samples were characterized by scanning electron microscopy (SEM, JSM-6360LV). The average cell diameter and cell density were calculated by the same method as described in paper [32].

## RESULTS AND DISCUSSION

The FTIR spectra of GS, (PBT-GS)-PBT, PBT and PBT-GS are shown in Fig. 1, among which (PBT-

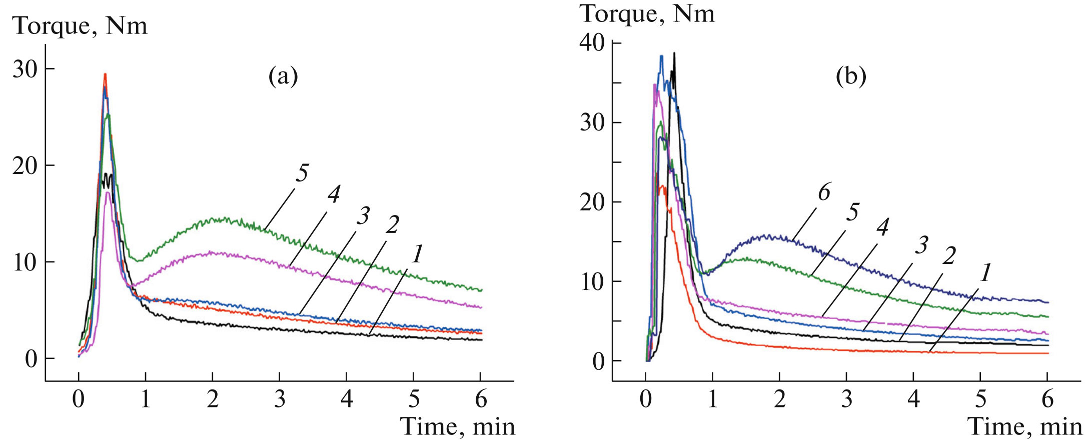
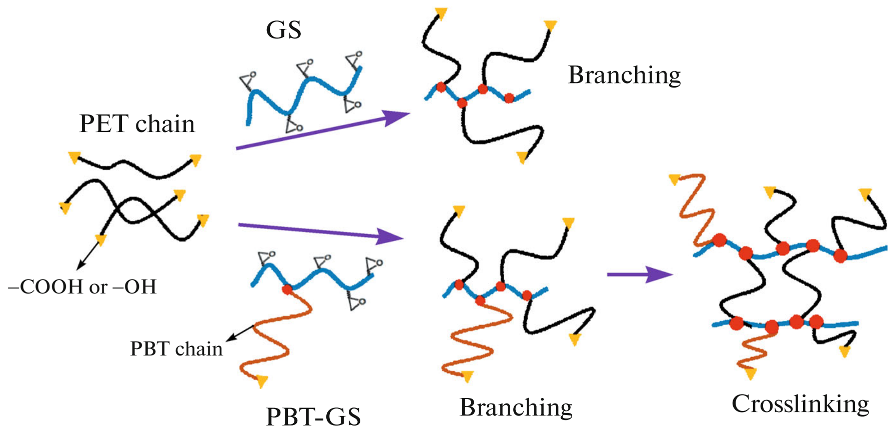


**Fig. 2.** (Color online) Torque vs. residence time in the Torque Rheometer (a) PET with GS: (*1*) PET-0, (*2*) PET-GS-1, (*3*) PET-GS-2, (*4*) PET-GS-3, (*5*) PET-GS-4; (b) PET with PBT-GS: (*1*) PET-PBT, (*2*) PET-0, (*3*) PET-(PBT-GS)-1, (*4*) PET-(PBT-GS)-2, (*5*) PET-(PBT-GS)-3, (*6*) PET-(PBT-GS)-4.

GS)-PBT is the subtractive spectrum between PBT-GS spectrum and PBT spectrum. The characteristic bands at 1726 and 908 cm$^{-1}$ in GS spectrum indicate the existence of the ester groups and the epoxy groups, and the bands at 3030, 757 and 697 cm$^{-1}$ owe to C—H groups and mono-substituted groups on the benzene ring. In PBT-GS FTIR spectrum, the epoxy group at 908 cm$^{-1}$ cannot be observed for the low content. However, the obtained subtractive spectrum (PBT-GS)-PBT based on the band at 729 cm$^{-1}$ (referred to para-substituted benzene in PBT) showed a new band at 906 cm$^{-1}$, confirming the existence of epoxy group in the PBT-GS sample.

Furthermore, based on the GPC results, the number average molecular weight ($M_n$) and molecular weight distribution (PDI) of GS are equal to 2.9 × 10$^3$ and 1.8, respectively. The epoxy group content of GS is 0.216 mol/100 g according to the titration result. According to the $M_n$ and the epoxy group content, the calculated average number of epoxy group per GS molecule is 6.5.

GS and PBT-GS were used as the chain extender to increase the molar mass and introduce branched structure of PET. The possible scheme of reaction between PET and chain extenders is shown below. The melt reactions were conducted in a Torque Rheometer. The recipes are listed in Table 1. The curves of torque vs residence time and torque values at 6 min are shown in Fig. 2 and Table 2, respectively. For the pure PET, after an initial melting stage, the torque of the sample continuously decreased for the molecular chain relaxation and the possible chain scission due to mechanical and thermal degradation. However, after the introduction of GS or PBT-GS, the torques of modified samples first increased, then gradually levelled off. With the increase of the amount of chain extenders, the reactive peaks became remarkable and the torque values at 6 min increased, especially in the samples PET-GS-3, PET-GS-4, PET-(PBT-GS)-3 and PET-(PBT-GS)-4.



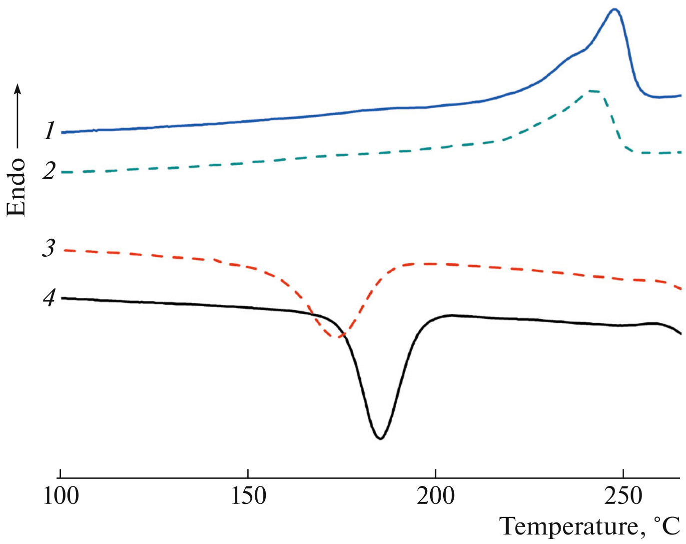


**Fig. 3.** (Color online) DSC curves of (*1*), (*4*) branched PET and (*2*), (*3*) crosslinked PET. (*1*) and (*2*) heating scan; (*3*) and (*4*) cooling scan.

Intrinsic viscosity and the gel content of samples are listed in Table 2. Intrinsic viscosity of PET-0 was lower than that of PET-virgin, which indicated that the chain scission and decomposition took place in the melt processing. With GS dosage increased, intrinsic viscosity of the samples increased and then exceeded that of PET virgin. Furthermore, intrinsic viscosity of all samples using PBT-GS as the chain extender were higher than those samples using GS under the same content of GS, which indicated PBT-GS had the better effect in the chain extension and branching than GS. These results clearly indicate GS chemically bonded on PBT molecular chains are easier to bond with PET than using GS directly, even the cross-linking was found in some samples, such as PET-(PBT-GS)-3 and PET-(PBT-GS)-4.

The DSC curves of the insoluble part in modified PET samples (cross-linked PET) as well as PET-(PBT-GS)-2 (branched PET) are shown in Fig. 3. The obvious crystallization and melting peak of insoluble part indicated that the degree of cross-linking was

**Table 2.** Torque values at 6 min, intrinsic viscosity and gel content of PET samples

| PET samples | Torque values at 6 min, Nm | [η], dL/g | Gel content, wt % |
|---|---|---|---|
| PET-virgin | – | 0.76 | 0 |
| PET-0[a] | 1.80 | 0.57 | 0 |
| PET-GS-1 | 2.50 | 0.58 | 0 |
| PET-GS-2 | 2.80 | 0.65 | 0 |
| PET-GS-3 | 5.20 | 0.72 | 0 |
| PET-GS-4 | 7.00 | 0.80 | 0 |
| PET-PBT[b] | 1.00 | 0.56 | 0 |
| PET-(PBT-GS)-1 | 2.60 | 0.79 | 0 |
| PET-(PBT-GS)-2 | 3.40 | 0.86 | 0 |
| PET-(PBT-GS)-3 | 5.60 | 0.94 | 4% |
| PET-(PBT-GS)-4 | 7.50 | 0.95 | 16% |

[a] PET-0 is the PET-virgin after being processed in a Torque Rheometer.

[b] PET-PBT is the sample blending 70 wt % of PET and 30 wt % of PBT.

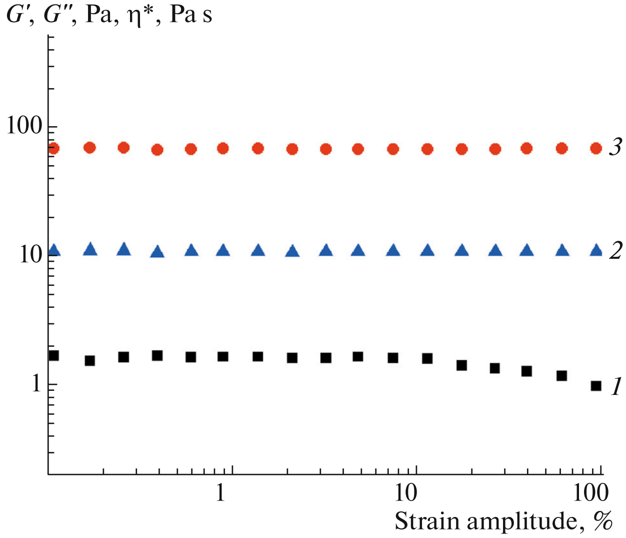


**Fig. 4.** (Color online) The dynamic strain sweep curve of PET-0: (*1*) $G'$, (*2*) $G''$, (*3*) $\eta^*$.

minor, which would not affect the melt processability of modified PET but improve its foamability.

Figure 4 shows the dynamic strain sweep curve of PET-0. It indicated the storage modulus started to decrease when the strain amplitude was more than 10%. Therefore, 1% strain was chosen to ensure all the samples were in the linear domain of viscoelasticity.

Figure 5 shows the dependence of the complex viscosity ($\eta^*$), storage modulus ($G'$) and loss modulus ($G''$) on the frequency of PET samples with different GS contents. $\eta^*$, $G'$ and $G''$ of all modified PET samples are higher than of unmodified one, especially in the low frequency region. As shown in Figs. 5b and 5c, $G'$ and $G''$ of PET-GS-4 at low frequency of 0.1 rad/s were 8.1 and 7.6 times higher than that of unmodified PET. Furthermore, the PET-GS-4 displayed a distinct shear thinning behavior which is the typical feature of long chain branched structure as shown in Fig. 5a. The development of $\eta^*$, $G'$, $G''$ and the shear thinning behavior demonstrate branched structures have been introduced in PET samples.

Figure 6 shows the frequency dependence of $\eta^*$, $G'$ and $G''$ for PET samples with different contents of PBT-GS. The trends of frequency dependence of $\eta^*$ were similar to the PET modified by GS and the shear thinning behavior was more obvious. $G'$ and $G''$ showed more significant increase than the unmodified counterpart. It was noted that $\eta^*$, $G'$, $G''$ of the PET sample blended with PBT were lower than those of virgin PET. However, $\eta^*$, $G'$ and $G''$ of the PET samples modified by PBT-GS were much higher than those of PET-PBT and PET-0, and also were higher than those of PET modified by GS under the same content of GS. The reason should be that the PBT molecular chains chemically bonded on GS are beneficial for the bonding reaction of PET with epoxy groups due to the good compatibility with PET.

DSC measurements of PET samples were carried out before and after the modification. Table 3 lists $T_c$, $\Delta H_c$, $T_m$, $\Delta H_m$ and $\chi$ of PET samples, and the corresponding DSC curves are shown in Fig. 7. $T_c$ and $\chi$ of PET decreased after the chain extenders were introduced, indicating that the crystallization of PET became more difficult because of the branching, cross-linking and the less regularity of the structure. In addition, $T_m$ decreased, which became significant when more chain extenders were introduced. It was noted that compared with GS, PBT-GS resulted in the lower $T_m$ of PET, which implied the larger change of PET's structure, indicating that PBT-GS is a kind of better extender in chain extending and branching than GS.

Figure 8 displays the cellular structures of unmodified PET, PET-GS-4 and PET-(PBT-GS)-4 samples under the same foaming condition. No distinct cell structures could be discerned in the unmodified PET due to the low melt viscosity and melt strength. PET-GS-4 appeared many visible and un-uniform cells,

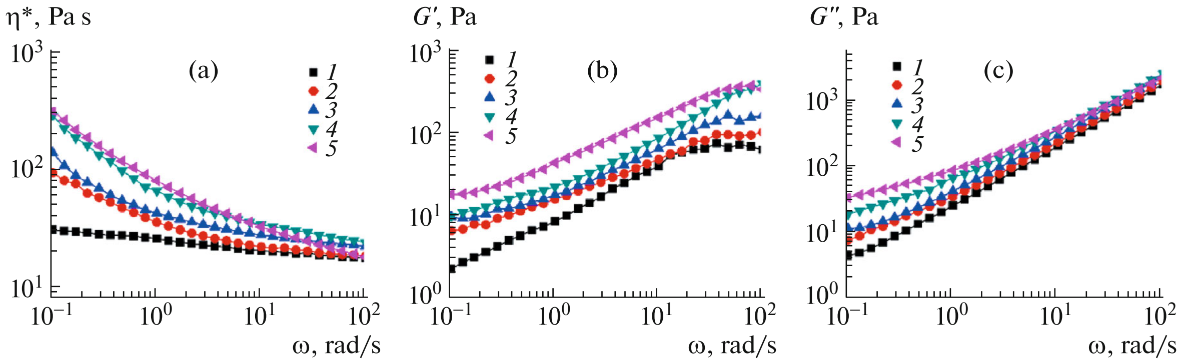


**Fig. 5.** (Color online) Rheological properties (a) $\eta^*$, (b) $G'$, (c) $G''$ vs. frequency for PET samples with different GS contents: (*1*) PET-0, (*2*) PET-GS-1, (*3*) PET-GS-2, (*4*) PET-GS-3, (*5*) PET-GS-4.

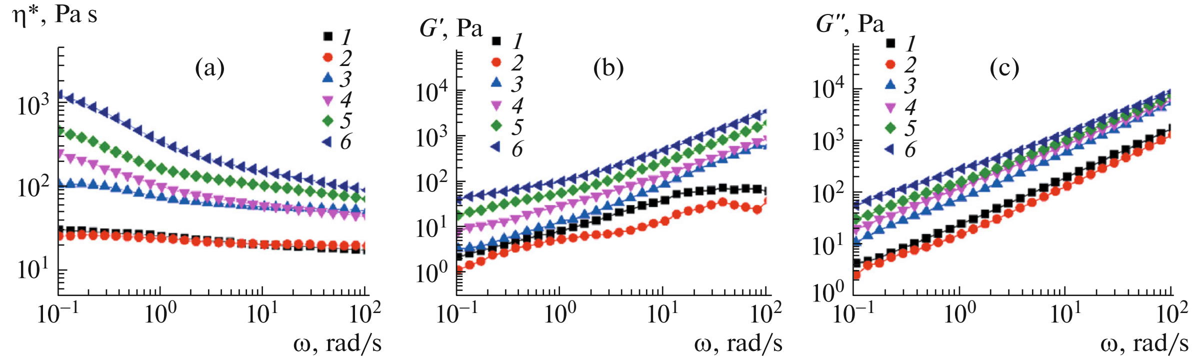


**Fig. 6.** (Color online) Rheological properties (a) η*, (b) *G*', (c) *G*" vs. frequency for PET samples with different PBT-GS contents: (*1*) PET-0, (*2*) PET-PBT, (*3*) PET-(PBT-GS)-1, (*4*) PET-(PBT-GS)-2, (*5*) PET-(PBT-GS)-3, (*6*) PET-(PBT-GS)-4.

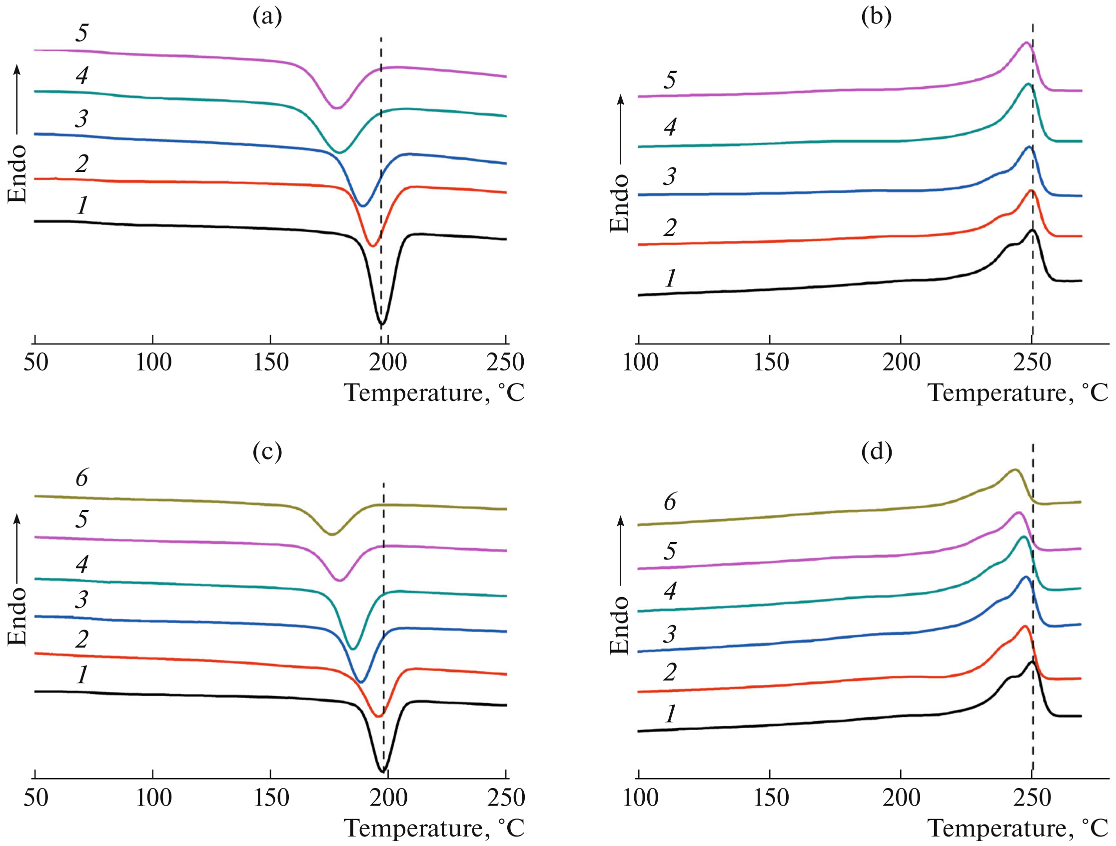


**Fig. 7.** (Color online) DSC plots of different chain extender contents during (a), (c) cooling and (b), (d) second heating scan. (a) and (b) PET with GS: (*1*) PET-0, (*2*) PET-GS-1, (*3*) PET-GS-2, (*4*) PET-GS-3, (*5*) PET-GS-4; (c) and (d) PET with PBT-GS: (*1*) PET-PBT, (*2*) PET-0, (*3*) PET-(PBT-GS)-1, (*4*) PET-(PBT-GS)-2, (*5*) PET-(PBT-GS)-3, (*6*) PET-(PBT-GS)-4.KC

which was better than unmodified PET. Inspiringly, PET-(PBT-GS)-4 had uniform cell structures with average cell diameter 61 μm and high cell density 1.8 × $10^8$ cells/cm$^3$ as shown in Fig. 8c, indicating that the excellent foaming properties. The foaming behaviors of modified PET further demonstrated PBT-GS are more beneficial for the bonding reaction of PET with epoxy groups to improve the branching of PET.

## CONCLUSIONS

Two chain extenders (GS and PBT-GS) containing epoxy group were employed to modify PET. It was

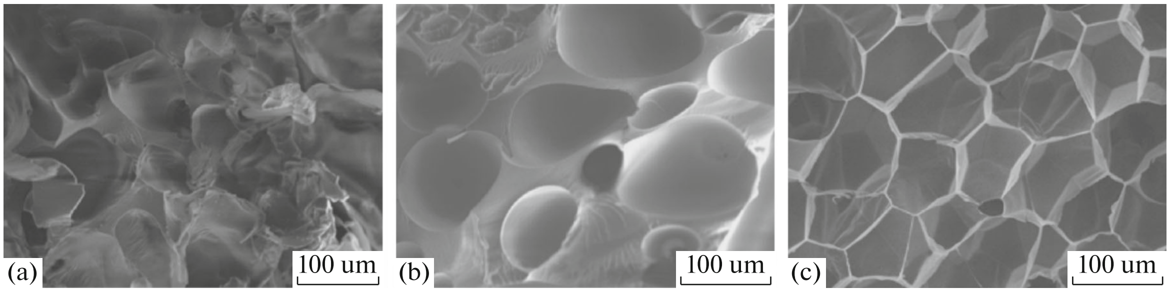


**Fig. 8.** SEM micrographs of (a) PET-virgin, (b) PET-GS-4, and (c) PET-(PBT-GS)-4 at a saturation pressure 15 MPa and foaming temperature 230°C.

found that the intrinsic viscosity of modified PET increased with increasing of the amount of chain extenders. The change of molecular structure resulted in significantly different shear flow properties of melt before and after modification. *G*' of modified PET remarkably increased at the low frequency region and a pronounced shear thinning behavior was found with increasing chain extender content. The results indicated that both GS and PBT-GS had been introduced to the backbone of PET macromolecules and formed branched structures, some even mildly cross-linked, which would not affect the melt processability. Furthermore, $T_c$, $T_m$ and $\chi$ of the modified PETs decreased due to the forming of branched and cross-linked structures. The branched and crosslinked PET with higher melt viscosity and melt strength will expand the application of linear PET in blow-molding, foaming and modification of recycled PET.

Especially, PBT-GS was more effective than GS in branching, which indicated PBT molecular chains chemically bonded on GS were beneficial for the bonding reaction of PET with epoxy groups due to the good compatibility with PET. Therefore, PBT molecular chains bonded with GS may play a role of macromolecular compatibilizer and will have potential applications in the high performance modification of PET. In the application of foaming materials, foamed PET with uniform cell structures, average cell diameter 61 μm and cell density 1.8 × $10^8$ cells/cm$^3$ was obtained using modified PET by PBT-GS.

**Table 3.** Crystalline and melting characteristics of PET modified by different methods

| PET samples | $T_c$, °C | $\Delta H_c$, J/g | $T_m$, °C | $\Delta H_m$, J/g | $\chi$, % |
|---|---|---|---|---|---|
| PET-0 | 197 | –38 | 251 | 34 | 25 |
| PET-GS-1 | 193 | –37 | 250 | 34 | 24 |
| PET-GS-2 | 188 | –35 | 249 | 32 | 23 |
| PET-GS-3 | 179 | –34 | 249 | 32 | 23 |
| PET-GS-4 | 178 | –33 | 248 | 31 | 22 |
| PET-PBT | 196 | –36 | 248 | 34 | 24 |
| PET-(PBT-GS)-1 | 189 | –35 | 248 | 32 | 23 |
| PET-(PBT-GS)-2 | 185 | –35 | 247 | 31 | 23 |
| PET-(PBT-GS)-3 | 178 | –30 | 245 | 29 | 21 |
| PET-(PBT-GS)-4 | 176 | –29 | 244 | 27 | 20 |

## ACKNOWLEDGMENTS

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