

Poly(ethylene terephthalate)/Expanded Graphite Conductive Composites: Structure, Properties, and Transport Behavior

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ABSTRACT: Poly(ethylene terephthalate)/expanded graphite conductive composites were prepared by the melt-blending method. The relationships between the preparation methods, microstructures, and conductivity properties of the composites were studied with scanning electron microscopy and conductivity measurements. The results showed that the composites presented a low percolation threshold and strong anisotropic conductivity. The epoxy resin had a strong intercalation effect on the expanded graphite that led to the easy formation of the conductive network. With classical statisti-

cal percolation theory, the conductivity behaviors of the composites were investigated. The results indicated that the nonuniversal critical exponent should be attributed to the anisotropy of conductivity, the tunneling conduction, and the particular structure. In addition, preliminary studies on the crystallization and dynamic mechanical behavior of the composites were performed. © 2008 Wiley Periodicals, Inc. *J Appl Polym Sci* 108: 1482–1489, 2008

Key words: composites; crystallization; polyesters

INTRODUCTION

Adding conducting fillers to insulated polymer materials to enhance their electrical conductivity is a practical, simple, and economical way of manufacturing conducting polymer composites, which have been significantly applied in numerous ways, including antistatic thermal conductors, electromagnetic interference shields, self-control temperature materials, and other functional applications.^{1–5} However, this approach has the effect of reducing the mechanical performance and processability. To obtain electrically conductive systems with high conductivity and acceptable processing properties, reducing the concentration of conductive fillers is required. The shape and size of the filler are very important to the conductive network.^{6,7} Expanded graphite (EG), which is composed of many nanosheets with a thickness of only 30–80 nm inside, has a loose and multipore structure.⁸ EG can be exfoliated into a graphite nanosheet with a large diameter-to-thickness ratio under strong shear deformation, and this makes it easier to form a conducting network in the polymer matrix in

comparison with spherical fillers, thus resulting in a very low percolation value.

Conducting polymer composites have attracted much attention over the past 3 decades. Celzard et al.⁹ first reported epoxy resin/EG conducting composites with a percolation threshold of only 1.3 vol %. Subsequently, several researchers prepared various conducting composites, such as poly(methyl methacrylate)/EG,¹⁰ polystyrene/EG,¹¹ and polyamide 6/EG¹² by *in situ* polymerization and PE/EG¹³ and polypropylene/EG¹⁴ by solution intercalation. Because the EG particles were distributed homogeneously in the polymer matrix in the form of nanosheets varying in thickness from 10 to 50 nm on the basis of transmission electron microscopy results, a very low volume fraction of EG was needed to reach the percolation threshold. Although the intercalation effect of polymerization and solution methods is very good and the composites have excellent conductive properties, the techniques are very complex and time-consuming. Moreover, the first method is restricted by the sort of polymer, and the latter needs a great deal of solvent, which is hard to reclaim. As a result, the application of these two methods is limited, and mass production is difficult to put into practice. Melt-blending intercalation does not need lots of solvent and uses the general processing equipment for plastics. Hence, melt intercalation is a more efficient and environmentally benign processing method for polymer/EG conductive composites.

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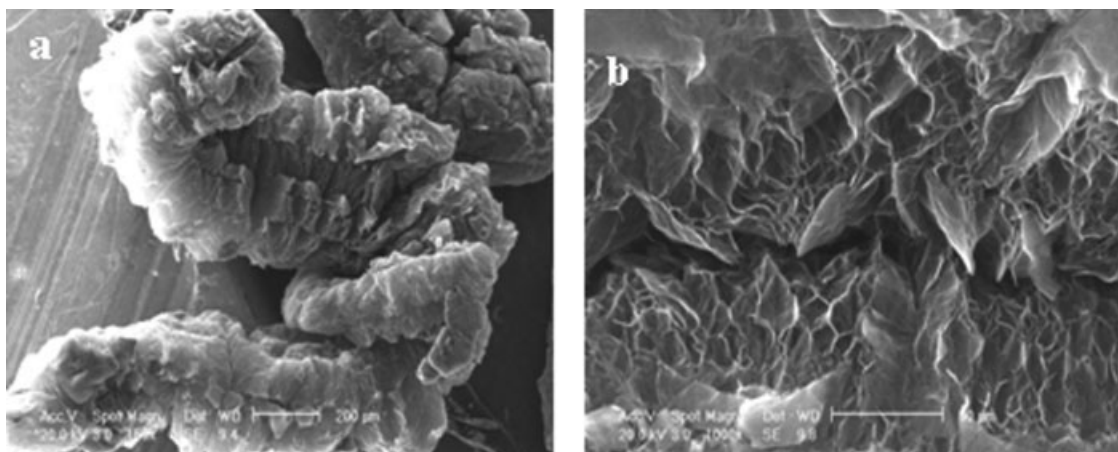


Figure 1 SEM micrographs for EG.

In this article, we report the preparation of poly (ethylene terephthalate) (PET)/EG conductive composites by melt processing. The influence of the EG content and preparation method on the conductivities of the as-prepared composites is studied first. Then, we investigate the electrical conduction mechanism, using the percolation theory, and provide some interpretation about the nonuniversal critical exponent. The dynamic mechanical properties and crystallization behaviors are also researched in detail.

EXPERIMENTAL

Materials

Expandable graphite (Kp32) was supplied by Qingdao Tianhe Graphite Co., Ltd. (Qingdao, China), with an expansion ratio of about 200–400. PET was produced by Yizheng Chemical Fiber Co., Ltd. (Yangzhou, China), with an intrinsic viscosity of 0.65 dL/g.

Sample preparation

The expandable graphite was placed in an oven at 900°C for rapid expansion and exfoliation to obtain the EG. The EG was modified by the epoxy resin, and the yield was named EP-EG. EG and EP-EG were mixed with PET in a PolyDrive Rheocord (Thermo Electron GmbH, Karlsruhe, Germany) at 265°C for 8 min at a rotation speed of 50 rpm. The composites were compression-molded into thin sheets with dimensions of 100 × 100 × 1 mm³ at 270°C for characterization.

Scanning electron microscopy (SEM) observation

SEM (XL-30 SEM, Philips, Eindhoven, Holland) was employed to observe the microscopic structure of EG and the composites. The samples were freeze-fractured, and the fractured surfaces were gold-sputtered *in vacuo*.

Electrical conductivity measurements

The volume conductivity (σ) of the PET/EG composites was measured at room temperature with a ZC36 high-resistance meter (Shanghai No. 6. Precise Instrument Factory, Shanghai, China) when σ was less than 10⁻⁸ S/cm or with a UT33 numeric multimeter (Shenzhen Youlide Co., China) when σ was greater than 10⁻⁸ S/cm. In the latter case, pieces of copper foil fastened with a conducting paste were used as the electrodes. Data were recorded 10 s after application of the voltage.

Dynamic mechanical thermal analysis

The dynamic mechanical thermal properties were measured with a Netzsch (Selb, Germany) DMA242C in the dual-cantilever bending mold at a fixed frequency of 1 Hz. The sample dimensions were 60 × 10 × 1 mm³. The sample was tested with temperatures ranging from 25 to 180°C at a heating rate of 5°C/min.

Differential scanning calorimetry analysis

A differential scanning calorimeter (DSC-204F1, Netzsch) was employed to examine the nonisothermal crystallization characteristics of the composites. The samples were first heated to 290°C and held for 10 min and then were cooled to room temperature at a rate of 10°C/min in an N₂ atmosphere.

RESULTS AND DISCUSSION

Microstructure of EG

EG with a wormlike shape [Fig. 1(a)], about 2000–20,000 μm long, is a loose and porous vermicular material that shows a basic structure of parallel boards that collapse and deform desultorily¹⁵ [Fig. 1(b)]. The thickness of the graphite sheets in the

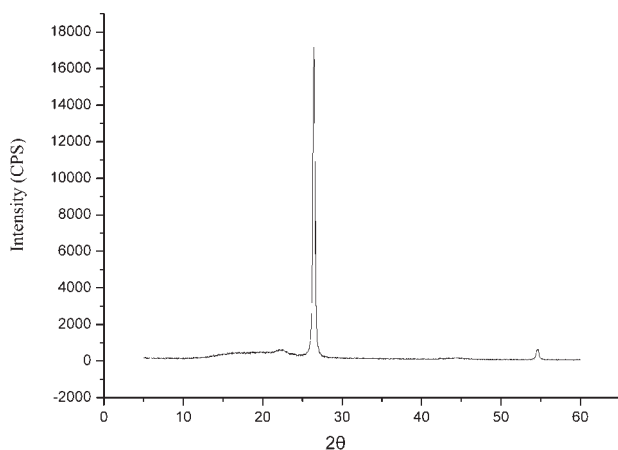


Figure 2 X-ray diffraction of EG.

exfoliated graphite observed with SEM is about 100–400 nm.⁸ When graphite intercalation compounds (GICs) are heated to a high temperature, molecules inside the carbon layer of the graphite decompose sharply, and this leads to expansion of the carbon layer along the *c* axis. However, because the formation of GIC is nonhomogeneous, EG can be expanded only partly. However, just this structure of EG affords an advantage in preparing polymer/EG composites.

Figure 2 shows X-ray diffraction of EG. There are two peaks in the graph, which is similar to X-ray diffraction of natural graphite. A 2θ peak at 26.46° is assigned to the 002 crystal face, the space between crystal face can be calculated with the Prague equation, and hence the *d*-spacing is 0.3366 nm. The *d*-spacing of EG increases a little in comparison with d_{002} of natural graphite (0.3354 nm).¹⁶ This indicates that the least constituent cell of EG is still the graphite crystallite, although EG behaves as a multiple-layer structure.

Relationship between the conductivity and EG content

Figure 3 shows the relationship between the volume electrical conductivity (σ) and EG content of the conductive composites prepared by different methods. The S-shaped curves indicate that the composites exhibit a typical percolation transition. σ changes tremendously (ca. 10 orders) in the region of the percolation transition but changes slowly in other regions. The critical conductive filler content of the percolation transition is named the percolation threshold value (ϕ_c). Obviously, ϕ_c of PET/EG and PET/EP-EG is far less than that of spherical particles. The functional groups of $-\text{OH}$, $-\text{COOH}$, and $\text{C}-\text{O}-\text{C}$ on the EG surface produced by the oxidation during the preparation of the precursor GICs¹⁷ improve the compatibility between EG and the polymer matrix.

As a result, the PET molecules diffuse into the interlayer of EG and make EG exfoliate into nanosheets during the melting process. This is propitious for the formation of the conductive network and decline of ϕ_c accordingly. Furthermore, it can be distinctly seen that ϕ_c of PET/EP-EG is lower than that of PET/EG composites. Concerning the intercalation action of epoxy to EG, Celzard et al.⁹ studied epoxy resin/EG conducting composites with a ϕ_c value of only 1.3 vol %. Because of the strong mutual effect between the epoxy group and functional groups on the EG surface, the epoxy resin can intercalate the network of EG easily with the help of a solvent. When the master batch of epoxy resin/EG is mixed with PET resin, PET molecules exfoliate on EG again. In addition, the selective localization of graphite flakes in multiphase polymeric blends is also one reason that PET/EP-EG composites have a very low percolation value.

Anisotropy of conductivity

The graphite nanosheets are flaky and possess a very large aspect ratio. However, this particular morphology imparts to the graphite nanosheets prominent anisotropy. During the melting process, EG is dispersed homogeneously in the polymer matrix in the form of nanosheets. However, the graphite sheets can be oriented in the matrix along the way of the melt flow in the step of compression molding. It makes the composites have strong anisotropy of conductivity. Figure 5 gives the conductivities measured in the two orthogonal radial directions [σ_x is the conductivities for parallel orientation, σ_z is the conductivities for perpendicular orientation; shown in Figure 4] and the anisotropy extent (σ_x/σ_z). σ_x/σ_z increases with the addition of EG, and the value is about 70 at an EG content of 9 vol %. This indicates that the probability of graphite flakes

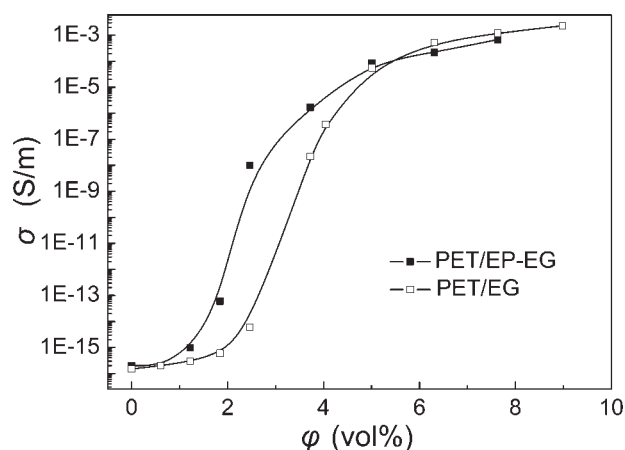


Figure 3 Electrical conductivity (σ) of the PET/EG composites as a function of the EG content (ϕ).

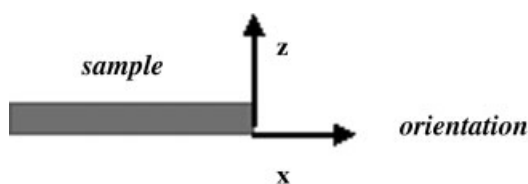


Figure 4 Orientation sketch map for the material.

contacting one another in the x direction is very large, and this leads to a strong three-dimensional conductive network, whereas the probability of graphite flake contact is very low in the z direction, and the conductive network is faulty. The conductivity difference between these two orientations proves the existence of the sequence of conductive networks and the anisotropic idiosyncrasy of PET/EG conductive composites, and this offers a way to prepare conductive composites with a sequencible conductive network and strong anisotropy.

Microstructure of PET/EG composites

Figure 6 shows the SEM photographs of the PET/EG and PET/EP-EG composites. The white strip is the graphite flake, and the fuscous part is the PET matrix. When the EG content is 2.46%, as shown in Figure 6(a), the space between graphite particles is far; as a result, electrons cannot be transported effectively, so the resistance of the composites is high. When the EG content reaches 7.64%, lots of EG particles can be seen in Figure 6(b). Those graphite flakes come into contact with one another, and this is propitious for electron transport. Therefore, the composites transform from an insulator to a conductor. This agrees with the conductivity/EG content curve in Figure 3. Figure 6(c) is a partially magnified photograph of Figure 6(b). Obviously, the graphite presents a multilayer structure, and its interspaces are filled with PET resin. This indicates that polymer molecules can diffuse into the network of EG. To study the shape of graphite in the matrix, the composites were dissolved in a phenol/tetrachloroethane solution. The solution was black and did not subside for 1 week. This means that the graphite flake is very small, even nanometer-grade. Figure 6(d) shows an SEM photograph of the graphite flakes. In the photograph, many graphite nanosheets can be observed. This indicates that EG particles have been exfoliated by the shear effect during the melting process. As for the photographs of the PET/EP-EG composites [Fig. 6(e,f)], there are many EG particles distributed in the matrix, and the extent of denseness of the EG particles is far higher than that in PET/EG with the same EG content. This proves that the extent of exfoliation of EG is greater in this system than that in PET/EG. Figure 6(f) shows the

much more microcosmic structure of PET/EP-EG composites. The original network structure of EG is essentially retained, and this indicates that the graphite flakes have constructed a micrometer composite network with polymers. However, the structural regularity of the network decreases because of the resin inserted into the interspace of the graphite flakes.

Statistical percolation theory

To study the relation between the formation process of the percolation network and the macroscopic electrical conducting properties of the conductive composites, Kirkpatrick¹⁸ applied the statistical percolation theory to the duality complex system of conductive fillers and insulators and pointed that the composite conductivity could be related to the conducting filler concentration by a simple power law:

$$\sigma = \sigma_0(\varphi - \varphi_c)^t \quad (1)$$

where σ is the composite conductivity, σ_0 is the conductivity of the filler, φ is the volume fraction of the filler, φ_c is the critical volume fraction, and t is the critical exponent of conductivity. t is considered to have universal applicability, and it depends only on the space dimension and is independent of the lattice structure.

According to the percolation theory, the experimental σ - φ data obtained from Figure 3 have been fitted with eq. (1), and the fitted results are shown in Figure 7. The peak value of the differential curve is defined as the percolation value (φ_c), and the t value can be determined from the slope of the straight line. Thus, where R is linearity correlation coefficient, the calculated values are $\varphi_c = 3.39\%$, $t = 4.25$, and $R = 0.9921$ for PET/EG [Fig. 7(a)] and $\varphi_c = 1.86\%$, $t = 2.71$, and $R = 0.9984$ for PET/EP-EG [Fig. 7(b)]. It is obvious that the percolation equa-

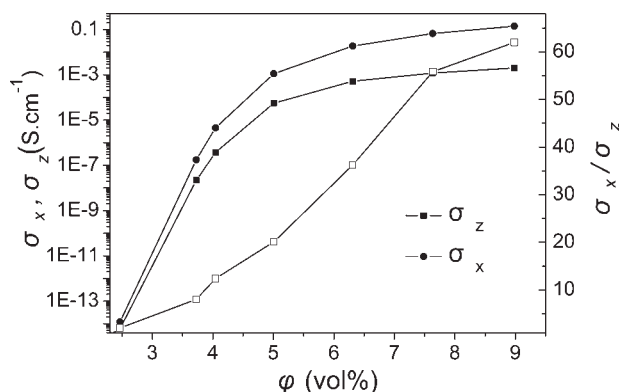


Figure 5 Conductivities parallel to the orientation (σ_x) and conductivities for perpendicular to the orientation (σ_z) and anisotropy extent (σ_x/σ_z) of the PET/EG composites.

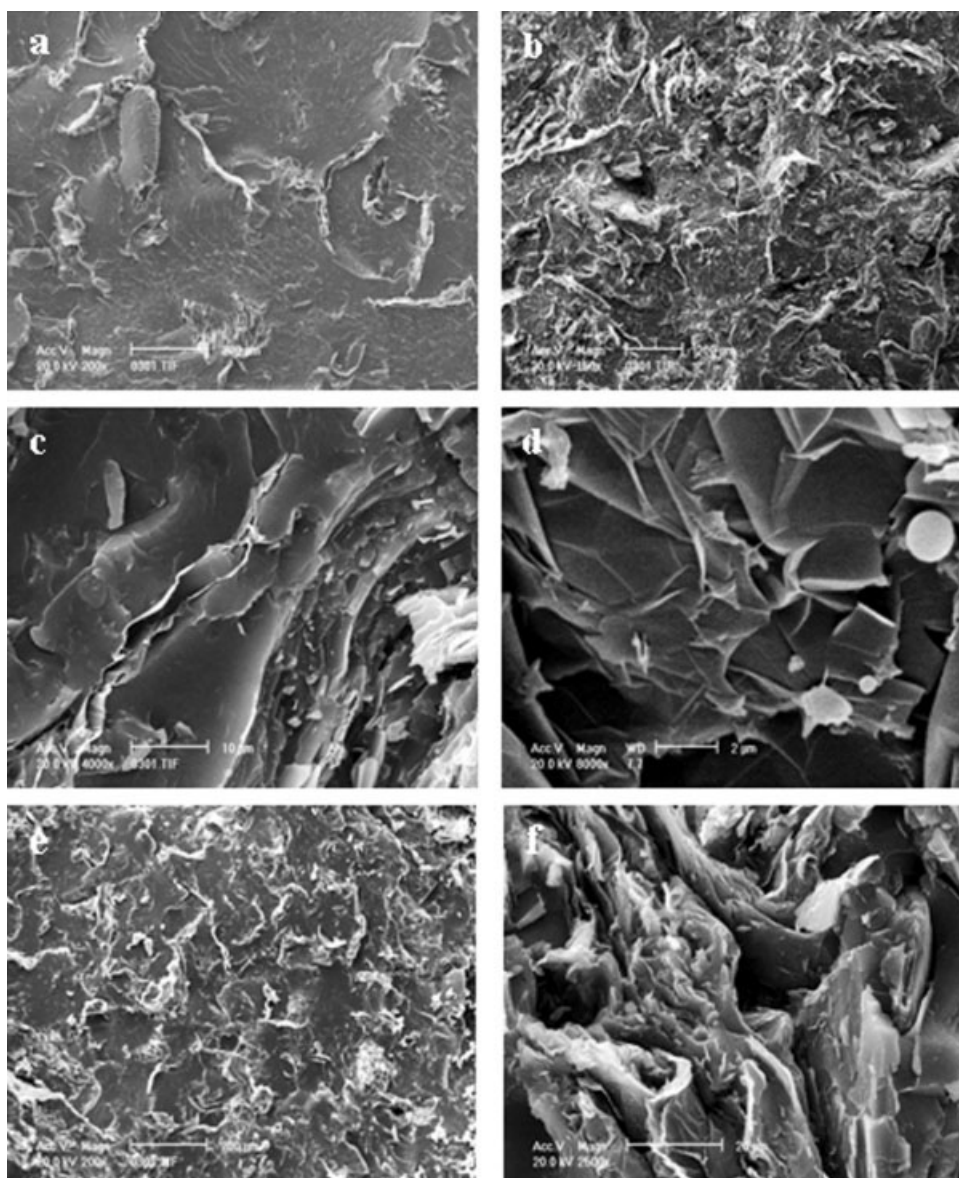


Figure 6 SEM micrographs of PET/EG composites: (a) PET/EG (2.46 vol %), (b,c) PET/EG (7.64 vol %), (d) after dissolution, and (e,f) PET/EP-EG (2.46 vol %).

tion [eq. (1)] is well obeyed for the conductivity data above the percolation threshold, and this indicates the applicability of the percolation theory for describing this system. However, the t value of PET/EG is obviously greater than the accepted universality value of the percolation theory ($t \approx 2$ in three-dimensional system)¹⁹ and mean-field theory ($t \approx 3$),²⁰ and the t value of PET/EP-EG approaches that of the mean-field theory, indicating some differences between the real system and theory mold.

Nonuniversal conductivity critical exponent

The nonuniversal critical exponent range of 2 to about 6 has been observed in many materials.^{21–25} Many researchers have widely studied this phenom-

enon, and several theories have been presented to explain the high t values. However, the conductivity percolation system is very complicated, so no theory can explain all real materials. Hence, reasonable interpretations are necessary to explore the nonuniversality behavior of the PET/EG system.

One reason for nonuniversality of the critical exponents is possibly the anisotropy of the conductivity. Celzard and Maréché²⁶ and Smith and Lobb²⁷ researched the critical exponent in anisotropic percolating systems adequately and pointed out that if σ_x/σ_z is constant in a given critical region, this will lead to isotropic exponents, whereas a nonconstant value obviously yields nonuniversal critical exponents. As for PET/EG composites, they have strong anisotropy, especially in the case of a high EG

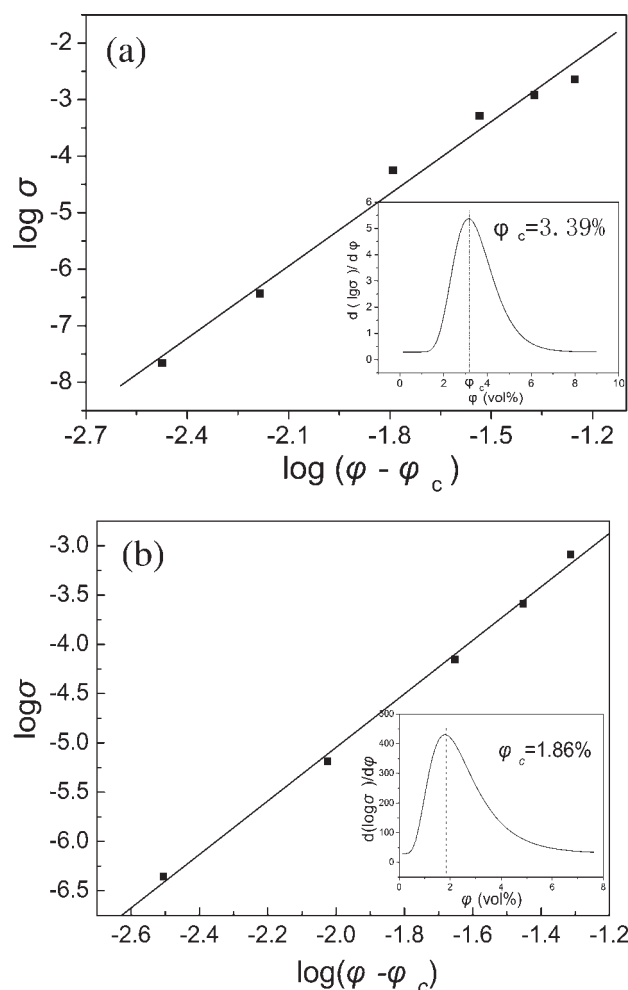
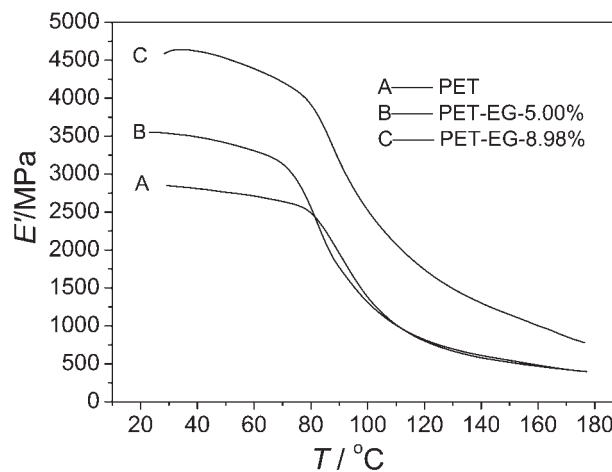


Figure 7 Plots of $\log \sigma$ versus $\log(\phi - \phi_c)$ for (a) PET/EG and (b) PET/EP-EG composites.

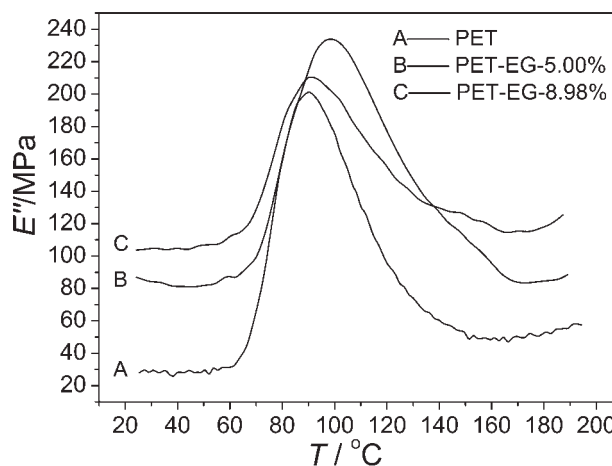
content. From Figure 4, it is found that the value of σ_x/σ_z is not a constant, and it increases with increasing EG contents. Obviously, the behavior of the critical exponents, deviating from the accepted value in the PET/EG system, agrees with the previously mentioned point of Smith and Lobb and Celzard and Marêché. Therefore, the anisotropic behavior of conductivity can be applied to explain the critical exponent's deviation when the anisotropic extent is unfixed.

Another interpretation may be the influence of the tunneling conduction.²⁸ Because of the existence of some functional groups on the EG surface, the physical and chemical interactions could take place between EG and the polymer matrix;¹² this would make polymer molecules permeate the pores and interlayers of EG easily with the help of the shear effect, so wrapping of the EG sheet by polymer molecules would become possible.²⁹ However, such a structure of the composites leads to the following result: the electron-transport mechanism cannot

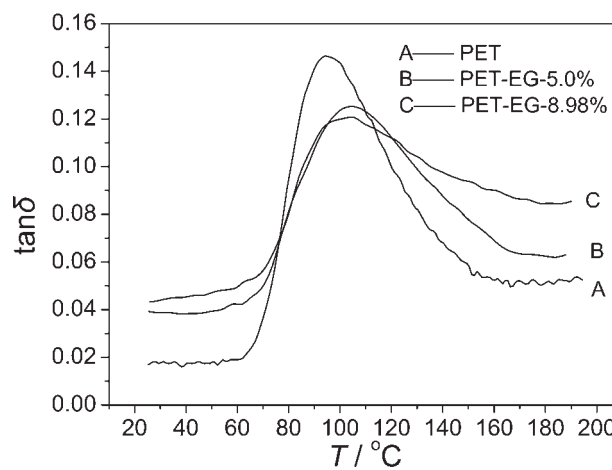
absolutely transform from tunneling conduction to ohm contact conduction at high EG contents. According to Nakamura et al.,³⁰ tunneling conduc-



(a)



(b)



(c)

Figure 8 (a) E' , (b) E'' , and (c) $\tan \delta$ of PET/EG composites with different filler contents as a function of temperature.

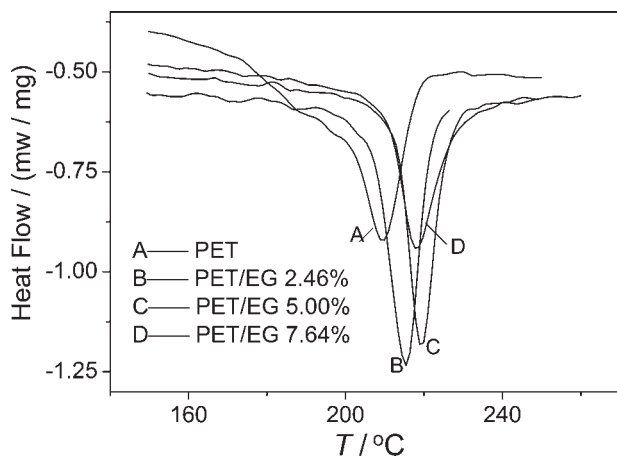


Figure 9 Differential scanning calorimetry cooling curves of pure PET and PET/EG composites.

tion still exists: above the percolation value, it will make the percolation region become broader, and the conductivity behavior will deviate from the classical statistical percolation theory. Hence, the nonuniversal behavior of the PET/EG system can be explained by the tunneling effect.

Besides the presented explanations, the particular structure of EG is possibly another reason for the increase in t . Because polymer molecules cannot fully permeate all pores of EG during the blending process, there are many air bubbles in the materials. Therefore, the filler volume fraction becomes uncertain, and the filler itself has an uncertain structure, which leads to an unacceptable critical exponent value.

From the analysis, we conclude that the nonuniversal conductive behavior reflects the difference between the classical statistical percolation model and real percolation system and shows some localization in the lattice model to describe the conductive behavior of the complex system.

Dynamic mechanical properties

The storage modulus (E') and loss modulus (E'') of PET/EG composites as a function of temperature at

different filler contents are shown in Figure 8(a,b), respectively. Obviously, E' of the composites increases with increasing EG contents, and this is due to the reinforcing effect of EG particles.³¹ In addition, E'' of the composites is higher than that of pure PET, and this is due to the interfacial interaction between EG and the matrix. With increasing EG contents, the interface binding effect and the mutual friction effect between EG and the matrix molecules increase, and this heightens the energy of molecule movement.

Figure 8(c) shows $\tan \delta$ of PET/EG composites as a function of temperature. The glass-transition temperature of the composites shifts to a high temperature compared with that of the pure PET, the intensity of the peak declines, and the peak shape broadens; this indicates a strong filler-polymer interaction. This is possibly due to the higher surface area of EG and the existence of some functional groups, which can promote the adsorption of molecular chains onto the EG surface and restrict the mobility of the adsorbed polymer segments. Therefore, the glass-transition temperature of the composites increases.

Crystallization behavior

Figure 9 shows curves of the heat flow as a function of the temperature during the nonisothermal crystallization of PET and PET/EG composites at the cooling rate of 10°C/min. The different crystallization temperatures (T_c 's) obtained from the curves are listed in Table I. At the same cooling rate, T_c of the PET/EG composites is obviously higher than that of pure PET, the temperature range of crystallization of the composites becomes smaller compared with that of PET, and the supercooling degree ($T_m - T_c$, where T_m is the melting temperature) as a thermodynamic driver of crystallization declines with increasing EG loadings; all this indicates that the addition of EG leads to increasing crystallizability of the PET matrix. Because graphite sheets have an adsorption effect on PET molecules, the movement of macromolecules is restricted, and PET molecules are easily

TABLE I
Crystallization Parameters of Pure PET and PET/EG Composites Obtained from Differential Scanning Calorimetry at a Cooling Rate of 10°C/min

Sample	T_0 (°C)	T_c (°C)	T_e (°C)	D (°C)	$T_m - T_c$ (°C)	Crystallinity (%)
PET	218.6	209.5	201.2	17.4	51.1	41.0
PET/EG (2.46%)	221.7	215.4	207.1	14.6	42.5	27.9
PET/EG (5.00%)	225.8	219.3	212.9	12.9	38.3	26.3
PET/EG (7.64%)	227.6	218.0	210.9	16.7	39.0	25.5

The enthalpy of crystallization of 100% crystallized PET was 140 J/g.³² T_0 = onset crystallization temperature; T_c = peak crystallization temperature; T_e = end crystallization temperature; D = temperature range of crystallization.

heterogeneously nucleated; as a result, PET crystallizes at a higher temperature during the cooling process. However, as the EG content reaches 7.64%, the temperature range of crystallization, T_c , and $T_m - T_c$ change to the opposite aspect in comparison with PET/EG (2.46 and 5.0%), and this indicates that the PET crystallizability decreases. Therefore, the addition of EG can improve crystallization when the EG content is low but block the crystal process when the EG content is high. Although EG can accelerate the formation of the crystal nucleus with the appropriate EG content, the crystallinity of the PET resin decreases as the EG content increases. These phenomena imply that EG might behave as a heterogeneous nucleating agent for PET, but at the same time, the particles also hinder the transport of molecular chains, leading to a reduction in the rate of crystallite growth.

CONCLUSIONS

PET/EG electrical conducting composites were prepared via the melt-blending process. With the assistance of the shear effect on EG and the interaction between EG and PET, graphite sheets were effectively dispersed in the PET matrix, and this was propitious for the formation of a conductive network. The strong mutual effect between the epoxy group and the functional groups on the EG surface meant that the epoxy resin had a greater exfoliation effect on EG, and this led to a much lower percolation threshold value. On the basis of the classical statistical percolation theory, the conductive behaviors of the composites were analyzed. The results indicated that the critical exponents were higher than the accepted value. To find the origins of the deviant result, three factors were taken into account. The microstructure of the composites, the existence of the tunneling conduction mechanism above the percolation value, and the anisotropic conductive behavior all should be responsible for the nonuniversality of the critical exponent. Moreover, EG showed a significant reinforcement effect on the PET matrix, leading to an increase in the dynamic modulus of the PET/EG composites. The addition of EG changed the crystal mechanism, accelerated the crystal velocity, but decreased the crystallinity of the PET resin.

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