

Morphology and Electrical Properties of Carbon Black/Poly(ethylene Terephthalate)/Polypropylene Composite

H. Ebrahimi Farimani, N. Golshan Ebrahimi

Polymer Engineering Department, Chemical Engineering Faculty, Tarbiat Modares University, Tehran, Iran

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ABSTRACT: This work attempts to develop a carbon black (CB) filled conductive polymer composite based on poly(ethylene terephthalate) (PET) and polypropylene (PP). The process follows by localizing the CB particles in the minor phase (PET), and then the conductive masterbatch was elongated to form conductive microfibrils in PP matrix during melt extrusion process. After compression molding, a fine conductive three-dimensional microfibrillar network is constructed. For comparison purpose, CB, PET, and PP are mixed using different pattern. The morphology and the volume resistivity of the obtained composites are evaluated. Electrical conductivity investigation

shows that the percolation threshold and resistivity values are dependent on the CB concentration. The best morphological observation shows that the PET phases forms well-defined microfibrils, and CB particles overwhelmingly localize in the surfaces of the PET microfibrils, which led to a very low percolation threshold, i.e., 4.5 phr, and a reasonable conductivity. © 2011 Wiley Periodicals, Inc. *J Appl Polym Sci* 124: 4598–4605, 2012

Key words: poly(ethylene terephthalate); polypropylene; conductive polymer composite; electrical properties; carbon black

INTRODUCTION

One of the methods to increase the electrical conductivity of polymeric materials is the addition of conductive fillers such as carbon black (CB), carbon nanotubes, graphite, metal powders, etc. CB-filled polymer composites show electrical conductivities ranging from insulators to semiconductors. Percolation takes place at a critical CB loading, called percolation threshold, where the first three-dimensional continuous CB network is built throughout the polymer matrix. The percolation threshold of such mixtures depends on the CB structure (porosity, aggregate shape and structure, surface chemistry and particle size), polymer characteristics (chemical structure and crystallinity) and processing methods and conditions. Generally, higher conductivities of the polymer composites can be obtained by using CBs of smaller particle size (larger surface area), higher structure (better aggregation), lower particle density (higher particle porosity) and low volatility (fewer chemisorbed oxygen groups).^{1–3}

Concerning how to reduce the percolation concentration and how to control the desired conductivity

of the CB-filled polymer composites, most studies focus on optimizing the degree of mixing and morphology of the matrix apart from CB structure and polymer characteristics. The conduction in carbon black-filled polymers may result from the strong electric field effect between the conductive particles or just their direct physical contact. In the first case processes such as tunneling, field emission and space charge limited transport should be considered. In the second case, when carbon black particles are in direct contact, a continuous conducting network forms and the dependence between the current and voltage is of the ohmic type.^{4–9}

Reduction of the percolation threshold of conductive polymer composites (CPCs) is of significance in lowering cost, easier processing and better mechanical properties. Therefore, it is a long-term major topic to investigate how to control the percolation threshold efficiently. Up to now, there have been many investigations on controlling the percolation threshold by manipulating the distribution of CB particles, such as multipercolation, segregated distribution, and electrical field-induced method. Multipercolation can be achieved by selectively locating CB particles in one phase of a continuous multiphase blend. Then the percolation threshold of CPC can be reduced greatly. Nevertheless, every approach on reducing the percolation threshold of CPC has its more or less limitations. For example, the CPCs obtained by a multipercolation method

Correspondence to: N. G. Ebrahimi (ebrahimn@modares.ac.ir).

TABLE I
Characteristics of Polymer Used

Material	Intrinsic viscosity (dL/g)	Melt flow rate (g/10 min)	Melting point (°C)
PP	–	16	165
PET	0.6–0.66	–	260

usually have poor mechanical properties because of weak interfacial adhesion between immiscible polymer components. Therefore, it is still a tough challenge to reduce percolation threshold of CPC with less sacrifice of the combination of their properties.^{2,9–13}

A significant progress has been made in the past few decades for further reduction in the percolation threshold by means of mixing CB with immiscible polymer blends. The CB particles are found to be selectively located in one of two polymer matrices or at their interfaces. Double percolation is the most adequate for describing the conductivity of the heterogeneous distributed systems, where both the conductive network of CB in filler-rich phase and the continuity of this phase in the composites are basic requirements for maintaining a conductive network throughout the composites. Much effort has been devoted toward controlling the location of CB. It has been suggested that the difference of the interfacial energy between CB and two polymers could be as a thermodynamic criterion to predict the selective distributions of CB in a polymer blend.^{14–17}

It has been established that the filler geometry has influences on selective location and phase continuity in polymer blend matrices. Usually, a composite filled with a fiber or flake has a lower percolation threshold than one filled with spherical conductive particle. Fillers with high aspect ratio may also increase the tendency to form the co-continuous phases in polymer blend matrices.^{1,18} These morphology-property relationships imply that, if the conductive filler is preferentially, even totally, localized in the minor phase (a-polymer) of a polymer blend, and the conductive filler/a-polymer blend is elongated or oriented to form conductive microfibers in the polymer matrix (b-polymer), the composite obtained may have high conductivity (construction of 3D conductive in situ microfiber network), good mechanical properties (reinforcement of the microfibers) and low cost (simple processing operation and low conductive filler content).^{19–22}

TABLE II
Characteristics of Carbon Black Used

Grade name	Reference	CTAB-surface area (m ² /g)	DBP adsorption (mL/100 g)	Primary particle diameter (nm)
Printex XE2	XE2	600	380	30
Printex V	PV	100	–	90

TABLE III
Composition of Samples

Sample	Components	Composition (Phr)
I	XE2/PET/PP	2/20/80
	XE2/PET/PP	5/20/80
	XE2/PET/PP	8/20/80
	XE2/PET/PP	10/20/80
II	(PV/PET)/PP	2/20/80
	(PV/PET)/PP	5/20/80
	(PV/PET)/PP	8/20/80
III	(PV/PP)/PET	2/80/20
	(PV/PP)/PET	5/80/20
	(PV/PP)/PET	8/80/20
IV	PV/PET/PP	2/20/80
	PV/PET/PP	5/20/80
	PV/PET/PP	8/20/80

In our previous works,^{23–25} properties of polypropylene (PP)/ poly(ethylene terephthalate) (PET) blends from various angles have been studied. In this study, we attempt to develop a CB filled conductive polymer composite based on PET/PP (20/80) by an extrusion-hot stretching–quenching process. The morphology and the volume resistivity of the composite obtained were evaluated.

EXPERIMENTAL

Materials and sample preparation

The main materials used in this study included PET, PP, and CB. The PET is a commercial grade of textile polyester and is supplied by Shahid Tondgooyan Petrochemical, Iran. PP is V30S, a commercial product of Arak Petrochemical, Iran. Two types of CBs used in this study with High (Printex XE2) and Medium (Printex V) electrical conductivity, were obtained from Degussa Co. Some physical characteristics of polymer and CBs used in this study are given in Tables I and II, respectively.

Sample preparation

The CB was dried at 120°C for 10 h and PET was dried in a vacuum oven at 100°C for at least 12 h prior to processing. We prepared four types of samples (Table III). In Types I, IV, two different kinds of CB were mixed directly with PP and PET. In Sample II, the CB particles were mixed initially with PET and then PP was added. In Sample III, the process is reverse, the CB particles were added to PP primarily

and then obtained composite blended respectively with PET; we have tried to show the possibility of finding better electrical conductivities than the status lacking of microfibrils by making conductive network with the PET microfibrils containing CB particles. Normal carbon black (Printex V) has been used in Samples II–IV. Sample II, with buildup microfibrillar conductive network, gained closer electrical conductivity to Sample I. Sample I contained high conductive carbon black (XE2), consequently achieved conductivity of this sample became our source of data for comparison with other samples.

The composites were obtained using the following steps.

- I. XE2/PET/PP: First, (conductive CB)/PET compound was prepared through mixing CB and PET at a processing temperature (260°C) in an internal mixer. The PET and CB were added into the preheated rotating internal mixer and allowed to mix for 5 min. Then, after the addition of PP, the compound was allowed to mix for an additional 5 min.
- II. (PV/PET)/PP: The hot stretched composite with PET microfibrils referred hereafter to (PV/PET)/PP was obtained by an extrusion-hot stretching-quenching process at the processing temperature of the PET. PET was first dry-blended with PV and then melt compounded using a twin-screw extruder (with a diameter of 40 mm) with a screw speed of 200 rpm. The PV/PET masterbatch was granulated and dry mixed with PP. The extrusion of the mixture of the binary PV/PET blend and the PP was performed in the same extruder. The temperature profile used for the extruder was 190, 250, 270, and 260°C from hopper to die. The extrudate was hot stretched at a line speed of ca. 1.1 m/min by a take-up device with two pinching rolls to form the microfibrils.²³ After hot stretching, the extrudate was immediately quenched in a cold water (20°C) bath.
- III. (PV/PP)/PET: PP was first dry-blended with PV and then melt compounded using the twin-screw extruder at a setting temperature of 170°C. The PV/PP masterbatch was granulated and dry mixed with PET. Finally the obtained mixture was extruded.
- IV. PV/PET/PP: The mixtures of PV, PET, and PP without elongation were directly extruded.

The temperature profile for Samples III and IV were the same as indicated for Sample II.

To measure the electrical resistivity, composites were compression molded at a pressure of 10 MPa for 10 min at 170°C (the processing temperature of PP)

TABLE IV
Surface Free Energy

Material	γ (mJ/m ²)	γ^d (mJ/m ²)	γ^p (mJ/m ²)
CB	42.5	41.9	0.6
PP	16.08	16.06	0.02
PET	28.3	28.079	0.221

$\gamma = \gamma^d + \gamma^p$, γ , surface free energy; γ^d , dispersion component; γ^p , polar component.

into thick plaques. Because of the high melting point of PET (about 260°C), the PET microfibrils can be successfully reserved in the composite during molding.

Electrical resistivity measurements

The electrical resistivity was measured by a multimeter (TR6845, Advantest) in the perpendicular direction of the molded sheets. In this work, DC resistivity was measured using two-terminal technique. The sample dimensions were 5 × 50 × 50 mm³. For each composite, measurements from three samples were made and the average value was reported. The measurements were carried out at room temperature after 2 min in order to obtain a stationary value. Silver paste was used to ensure good contact of the sample surface with the copper electrodes.

Electron microscopy observation

The morphology and CB dispersion were observed by means of a field emission type scanning electron microscope. Samples for SEM analysis were taken from the central portion of the specimens. The specimen's were impact fractured. The surfaces were covered with a layer of gold to make them conductive prior to examination. In order to examine the morphology of the dispersed phase, some specimens were immersed in hot xylene for 60 min to etch away the PP matrix.

RESULTS AND DISCUSSION

Prediction for selective distribution of CB particles

The fact that there are many functional polar groups like carboxylic, phenolic, lactonic on the surface of CB is a factor affecting the selective distribution of CB particles.¹⁶ If the interactions between one polar polymer component and CB particles are strong enough, the interactions will influence the dispersion of CB particles.^{18,26} In CB/PET/PP composite, PET has a high surface tension and polarity, while the PP is a nonpolar polymer. The surface free energies of the materials at the mixing temperature employed in this work are listed in Table IV.^{27,28} The interaction between PET phase and CB particles seems to be

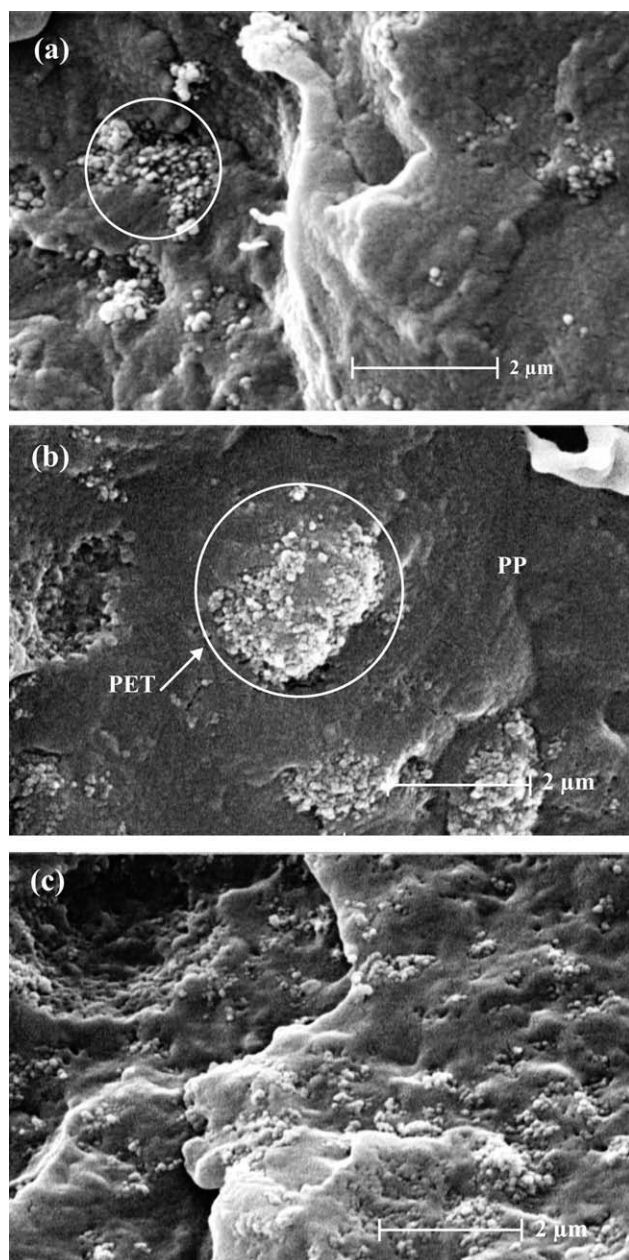


Figure 1 SEM micrographs of XE2/PET/PP composites with different XE2 loading. The volume ratio of PET and PP is 1 : 4, and the XE2 loading is: (a) 5 phr; (b) 8 phr, and (c) 10 phr.

stronger than that of PP phase and CB particles. In other words, CB particles have a priority to locate on the surface of PET phase. It is still widely accepted to use Young's equation to predict the selective distribution of CB in a polymer composite, which was first suggested by Sumita et al.,¹³ The Young's equation is:

$$\gamma_{c-\alpha} + \gamma_{\alpha-\beta} \cos \theta = \gamma_{c-\beta} \quad (1)$$

$$W_a = (\gamma_{c-\beta} - \gamma_{c-\alpha}) / \gamma_{\alpha-\beta} \quad (2)$$

Where θ is the contact angle of the polymer on the CB, w_a is the wetting coefficient, $\gamma_{c-\alpha}$, $\gamma_{c-\beta}$, and $\gamma_{\alpha-\beta}$ are the

interfacial tension between CB and α -polymer, CB and β -polymer, and α and β -polymers, respectively. If $w_a > 0$, that is, $\gamma_{c-\beta} > \gamma_{c-\alpha}$ CB particles will distribute overwhelmingly in α -polymer or at the interface, and if $w_a < 0$, that is, $\gamma_{c-\beta} < \gamma_{c-\alpha}$ CB particles will distribute in β -polymer or at the interfaces.

The calculated interfacial tensions between CB and PET and between CB and PP are 14.2 mJ/m² and 26.8 mJ/m², respectively. Hence, there is a very high probability that CB is better adhered to PET than to PP, or the larger affinity characterizes the CB/PET system due to the lower interfacial tension between CB and PET. In this study, the special mixing procedures were designed to locate CB particles on the surfaces of PET phases.

Morphology of CB particles

In order to probe how the distribution of XE2 particles in polymer composite takes place, the morphology of XE2/PET/PP composites mixed with different XE2 concentration were observed (see Fig. 1). Taking into account the composition of the ternary composite, PP serves as the continuous phase, while the indicated domains seem to belong to PET phase. Figure 1(b) shows the SEM micrograph of XE2/PET/PP composite, with a higher XE2 content, 8 Phr, with comparison to Figure 1(a). It seems that some XE2 particles, even aggregates, were embedded on the surfaces of the PET particles. Hence, the conductive network was built by the contact of the XE2 covered PET particles. The SEM micrograph of XE2/PET/PP composite with 10 Phr XE2 is shown in Figure 1(c). One can see that in the PP matrix, the CB aggregates increase with the CB loading compared with Figure 1(a,b). With the increasing CB loading, the morphology of the matrix changed (see Fig. 1c). When the surfaces of the PET particles were covered, Figure 1(b), CB particles still stay in the matrix (Fig. 1c). These CB particles seem to be almost in the form of aggregates. Nevertheless, the CB particles in the matrix can also help to build electrical networks together with the CB-covered PET particles. In this case, the morphology of the matrix becomes rather complicated.

To elucidate the structure inside the composite domains, a fractured PET particle was particularly observed at a high magnification as shown in Figure 2. It displays a typical incompatible morphology where the interfaces are clear and there is no evidence of any adhesion, Figure 2(a). Taking into account the composition of the ternary composite, it is believed that the continuous phase is PP. The larger domains in the PP matrix are PET, based on the fact that they contain very fine CB particles. Evidently, the PV particles are nonuniformly distributed in the whole system, and preferentially localized in the PET phase, Figure 2(b,c).

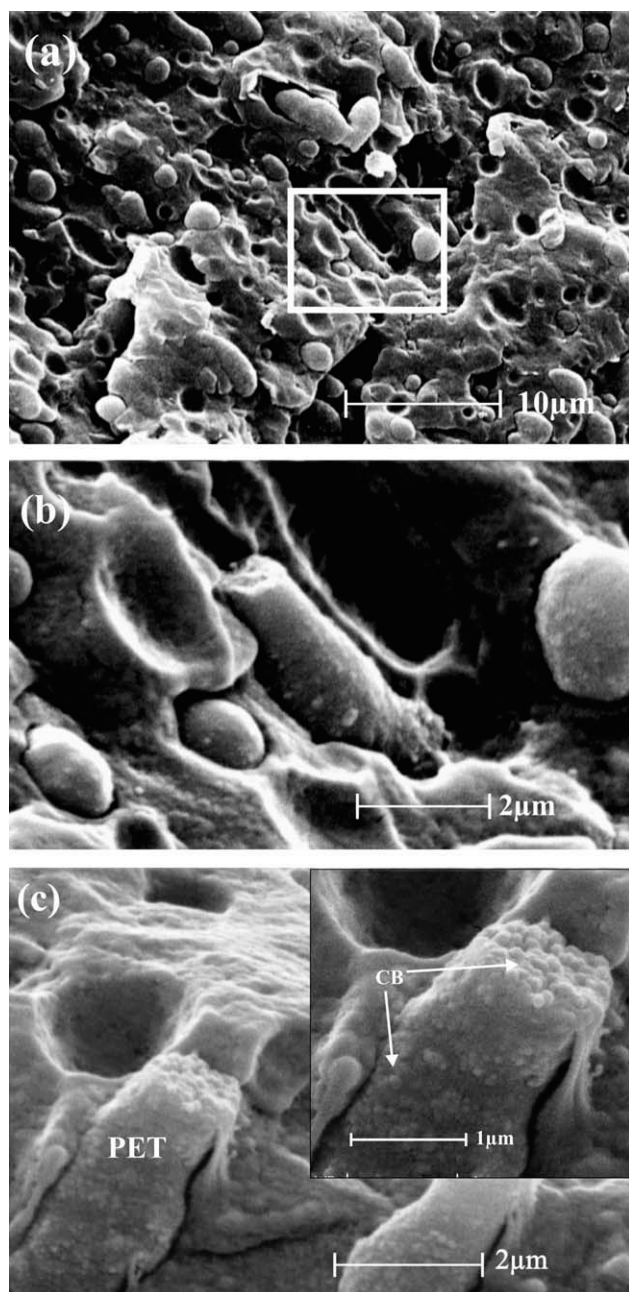


Figure 2 SEM micrographs of (PV/PET)/PP composite with low and high magnifications. The volume ratio of PET and PP is 1 : 4, and the PV loading is 5 phr. (a) \times 2000; (b) \times 10,000; (c) \times 10,000.

Figure 3 shows the SEM micrographs of directly mixed PV/PET/PP composites. It seems that the PET phases are distributed like separate droplets into the PP matrix, Figure 3(a). It is interesting that the PV particles in Figure 3(b) appear as tiny spheres rather than aggregates. The PV spheres in the PET phase are ca. 100 nm in diameter, which should be the primary particles. It seems breakup and cleavage of the PV aggregates occurred to produce the original PV particles during their localization in PET phase. Presumably, there are the following possible

reasons: (1) during drastic shear and elongational flow, some original particles were flaked off from their host aggregates; (2) these flaked primary particles had higher possibility to enter the PET phase due to their smaller size and higher affinity between CB and PET than between CB and PP.^{15,17}

Figure 4 shows the SEM micrographs of (PV/PP)/PET composites. This sample also displays a typical incompatible morphology, Figure 4(a); a large amount of the PV particles are seen in the PP matrix, Figure 4(b). It seems that the PV particles in the PP matrix, Figure 4(c), tend to form agglomerates. On the other hand, an evident gap between the PV/PET phase and the PP matrix exists as observed in the SEM micrograph [Fig. 2(c)] with higher magnification, indicating that the two phases have weak interfacial adhesion.

Therefore this weak interfacial adhesion and finely suitable dispersion may be valuable for the formation of microfibrils during hot stretching, but seemingly it goes against the mechanical properties.

During hot stretching through two pinching rolls, the melted PET droplets were elongated into liquid microfibrils, and subsequently, the extrudate was

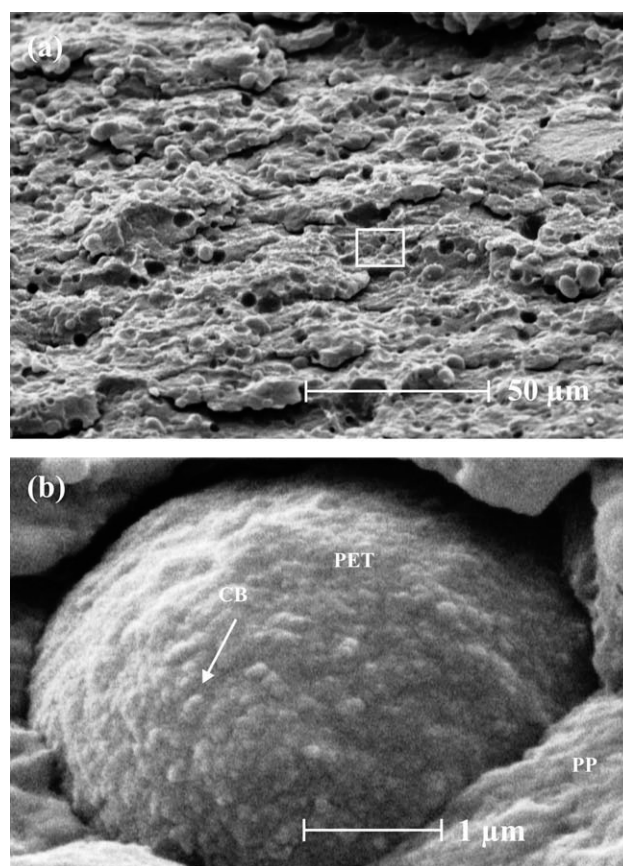


Figure 3 SEM micrographs of PV/PET/PP composite with low and high magnifications. The volume ratio of PET and PP is 1 : 4, and the PV loading is 5 phr. (a) \times 500; (b) \times 20,000.

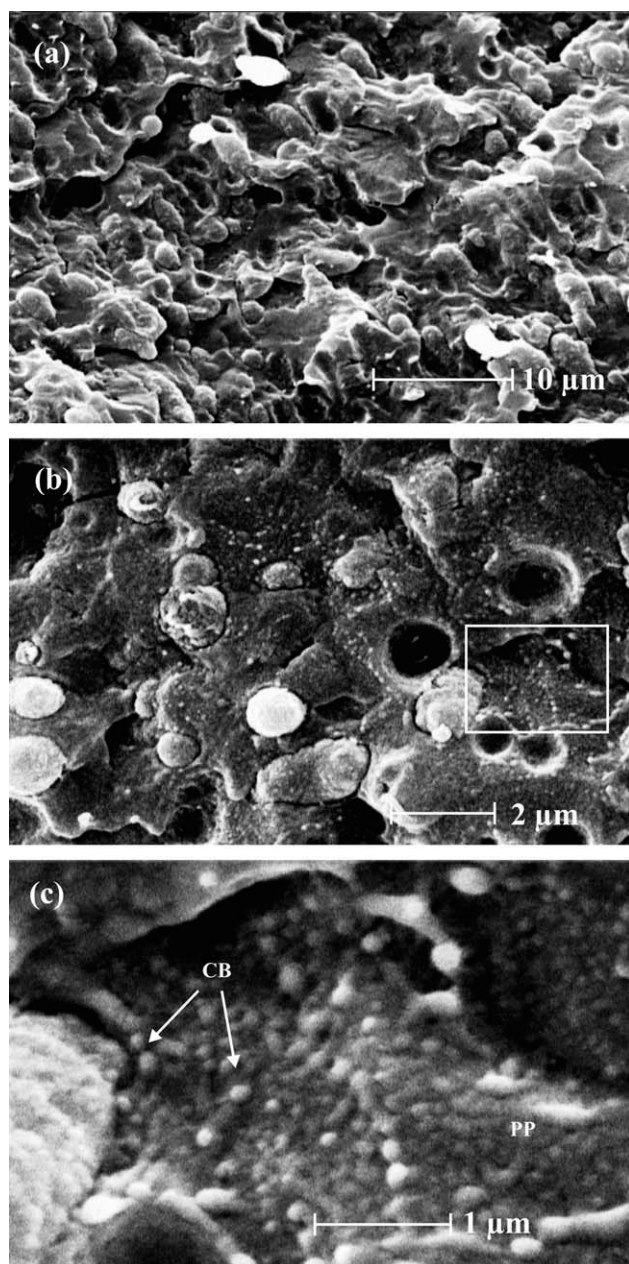


Figure 4 SEM micrographs of (PV/PP)/PET composite. The volume ratio of PET and PP is 1 : 4, and the PV loading is 5 phr. (a) $\times 2000$; (b) $\times 10,000$; (c) $\times 20,000$.

quenched by cold water to freeze the microfibrils. For clear observation, the PP matrix was dissolved away by hot xylene.

Figure 5 shows the representative morphology of the remaining microfibrillar composite. This figure indicates that the well-defined PET microfibrils were generated by this extrusion-hot stretching-quenching process. The diameter of the microfibrils is about 1–10 μm . The particles on the surfaces of the microfibrils are PV.

It is found that some PV particles, even aggregates, were embedded in the surfaces of the PET microfibrils. In addition, it can be found that the

surfaces of the microfibrils are coarse. Fibrillation of the dispersed phase in an incompatible polymer blend has long been studied with the aim of rheologically and morphologically examining the nature of deformation of a blend during the process. Many studies have showed that an elongational flow field and a small viscosity ratio can facilitate fibrillation of the dispersed phase in a polymer blend. The importance of the elongational flow is more prominent. It has been found that the dispersed phase was deformed into fibers in the blend with a viscosity ratio much higher than the unity.²⁶ The viscosity of the PET is lower than PP. When the CB was added into PET, the viscosity of the CB/PET compound was greatly increased since the CB cannot flow. Therefore, the fibrillation capability of the CB/PET compound in the composite depends not only on the elongational flow field, but also on the CB loading. Therefore, it is reasonable that the well-defined microfibrils cannot be achieved in the composite with the higher CB loading.

Composite electrical conductivity

The effect of CB on the conductivities of the composites was further studied by varying the

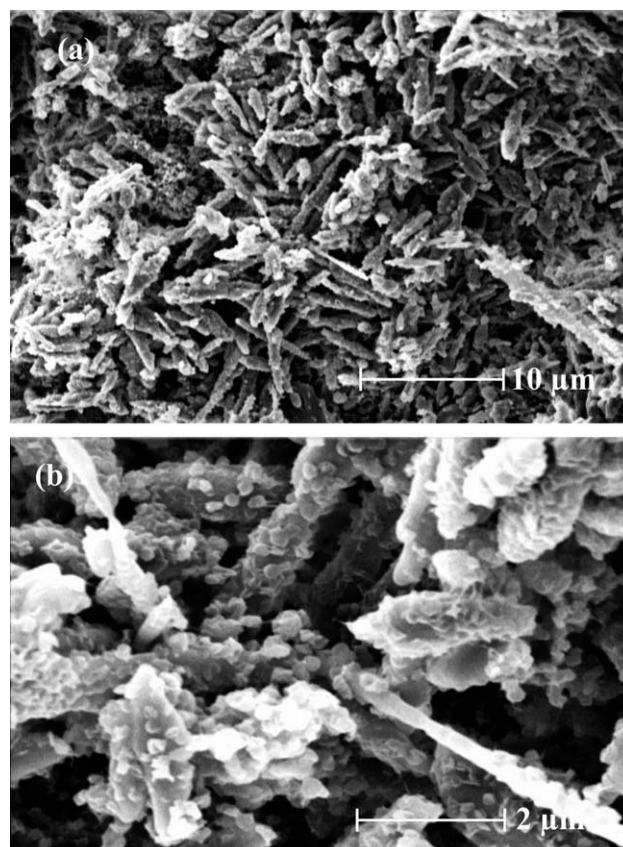


Figure 5 SEM micrographs of (PV/PET)/PP composite. The volume ratio of PET and PP is 1 : 4, and the PV loading is 8 phr. (a) $\times 2000$; (b) $\times 5000$.

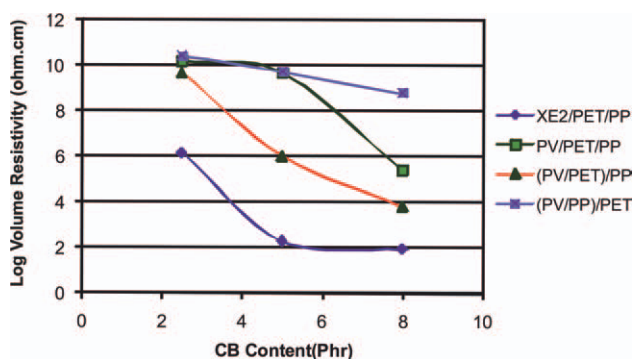


Figure 6 DC Volume resistivity of CB/PET/PP composites with high conductive XE2/PET/PP, PV/PET/PP, conductive microfiber network and (PV/PP)/PET as a function of CB concentration. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

concentration of CB from 2.5 to 8 phr. Figure 6 shows the DC volume resistivity for the XE2/PET/PP, PV/PET/PP, (PV/PET)/PP and (PV/PP)/PET composites as a function of the CB concentration.

The increase of conductivity is noticeable in all composites containing CB at all contents. From these curves, the percolation can be estimated, which is ca. 3.5 phr for XE2/PET/PP, whereas ca. 4.5 phr for microfibrillar conductive composite and ca. 8 phr for the common PV/PET/PP composites. A considerable reduction in percolation threshold has been successfully achieved using the present approach. At the PV percolation level, the resistivity of the microfibrillar composite is approximately reduced by five orders of magnitude. This assuredly demonstrates the superiority of the (PV/PET)/PP composite to the common conductive PV/PET/PP composite and (PV/PP)/PET.

The low percolation threshold of (PV/PET)/PP is attributed to the fine contact of PET microfibrils with PV particles being distributed in their surfaces, see Figure 5, but for the PV/PET/PP, PET phases were dispersed as separate droplets, see Figure 3(a), though PV particles were distributed on the surfaces of PET phases, a perfect conductive network was hard to be constructed, and a high percolation threshold was obtained. In general, (PV/PET)/PP blends had a better conductivity than both (PV/PP)/PET and PV/PET/PP.

Because of the formation of the conductive network in the (PV/PET)/PP composite, during compression molding, it seems that the local flow and diffusion of the melted PP made the PV particles-coated microfibrils contact with each other and thus built a fine electrical network. Furthermore, the remaining CB particles in the matrix would quite possibly contribute to high conductivity and transferring the electrons, especially at a higher CB loading. When the CB loading was relatively low, CB

particles were mostly located on the surfaces of microfibrils, and the conductive paths were, hence, formed mainly by the contacting of microfibrils, but their amount was limited due to the low CB content. If the CB particles scattered in the matrix and the CB coated microfibrils were not close enough, the tunneling conduction could also happen.¹⁰ Although the two conduction modes coexisted, the resistivity of the microfibrillar (PV/PET)/PP composite was still high because of the few conductive paths. In contrast, at a higher CB loading, the CB particles in the matrix increased the amount of conductive paths also increased by more conductive contact between CB particles and PET microfibrils so the resistivity of the blend was reduced. Therefore, by locating CB particles on the surfaces of the microfibrils, a more perfect conductive network can be built by the contact of a few CB particles. So a high volume of CB particles is not required to fill the PET microfibrils to protrude some of them on the surfaces of the microfibrils to build a conductive network.^{15,29,30}

CONCLUSION

We have established that the electrically conductive microfibrillar (PV/PET)/PP composite with 3D electrically conductive network and selective distribution of CB particles in the surfaces of the microfibrils can be successfully produced. It is more advantageous for conductivity that the CB particles are localized in the body of the PET phases which act as the conductive network. The composite with the well-defined PET microfibrils were generated in situ and a lot of CB particles were accumulated on the surfaces of the PET microfibrils. The percolation threshold of the microfibrillar CPC was only ca. 4.5 phr, showing a considerable decrease of percolation compared with that of the PV/PET/PP composite, ca. 8 phr.

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