

# Effect of Morphology on the Electric Conductivity of Binary Polymer Blends Filled with Carbon Black

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Received 10 November 2006; accepted 16 May 2007

DOI 10.1002/app.26827

Published online 20 July 2007 in Wiley InterScience (www.interscience.wiley.com).

**ABSTRACT:** Several carbon black (CB)-filled binary polymer blends were prepared in Haake rheometer. Distribution states of CB and effect of morphology on the electric conductivity of different ternary composites were investigated. Under our experimental condition CB particles located preferentially at the interface between polymethyl methacrylate (PMMA) and polypropylene (PP) in PMMA/PP/CB composites, in high-density polyethylene (HDPE) phase in PP/HDPE/CB composites, and in Nylon6 (PA6) phase in polystyrene (PS)/PA6/CB, PP/PA6/CB, PMMA/PA6/CB, and polyacrylonitrile (PAN)/PA6/CB composites; the ternary composites in which CB particles locate at the interface of two polymer components have the highest electric conductivity when the mass ratio of the two polymers is near to 1 : 1. The ternary composites in which CB particles located preferentially in one

polymer have the highest electric conductivity usually when the amount of the polymer component having CB particles is comparatively less than the amount of the polymer component not having CB particles; if the formulations of PS/PA6/CB, PP/PA6/CB, and PMMA/PA6/CB composites equaled and PA6/CB in them is in dispersed phase, PS/PA6/CB composites have the highest electric conductivity and PP/PA6/CB composites have the lowest electric conductivity; suitable amount of PS or PAN in PA6/CB composites increase the electric conductivity due in the formation of a parallel electrocircuit for electrons to transmit. © 2007 Wiley Periodicals, Inc. *J Appl Polym Sci* 106: 2008–2017, 2007

**Key words:** polymer blend; morphology; carbon black; electric conductivity; conducting mechanism

## INTRODUCTION

Conductive filler, such as metal powder,<sup>1,2</sup> carbon black (CB),<sup>3–5</sup> and carbon fiber,<sup>6</sup> have long been used to decrease the electrical resistivity of insulating polymers and fabricate conductive polymer composites (CPCs). CPCs can be used as EMI shielding materials, electronic packaging materials etc. The critical amount of conductive filler required to form continuous conducting paths and to impart electrical conductivity to the polymer matrix is called percolation threshold. CB is the most widely used conductive filler because of its abundant source, low density, permanent conductivity, and low cost. For CB-filled CPCs, it is desirable to decrease the percolation threshold as low as possible. High CB concentration increases the melt viscosity and decreases the impact resistance, thus the processability and mechanical properties of the composites were lost.<sup>7</sup> In highly filled CB compounds, CB tends to slough and cause contamination in clean room environment.

Increasing CB concentration also increases the final product cost.

Comparing to CB-filled single polymer, CB-filled immiscible polymer blends can obtain lower percolation threshold.<sup>8–15</sup> And there are several factors affecting the percolation threshold in CB-filled immiscible polymer blends, which include use of additives, the optimization of processing conditions, the size distribution, and porosity of CB particles.<sup>16,17</sup> But the most important factor is the control of the morphology of ternary composites, which includes the distribution state of CB particles in immiscible polymer blends and the phase morphology of each individual polymer.

Gubbels<sup>18</sup> focused on the effect of the selective localization of CB particles on the electric conductivity of multiphase polymeric materials. His result showed that the PS/PE/CB composites had the highest electric conductivity when CB distributed at the interface of two polymer components of the cocontinuous 45/55 PE/PS blend. Huang<sup>14</sup> pointed out that when binary blends are considered, there are two types of distribution that are beneficial to electric conduction. One is distributed predominately in one continuous phase; the other case is located preferentially at the interface. When CB preferentially locates in

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**TABLE I**  
**Samples of Blending Experiment 1**

Sample code	Components	Composition (g)
1a	PS/PA6/CB	0/10/3
1b	PS/PA6/CB	5/10/3
1c	PS/PA6/CB	10/10/3
1d	PS/PA6/CB	20/10/3
1e	PS/PA6/CB	30/10/3
1f	PS/PA6/CB	40/10/3
1g	PS/PA6/CB	50/10/3
1h	PAN/PA6/CB	0/30/5
1i	PAN/PA6/CB	10/30/5

one phase, the phase changes from a dropwise domain into a cocontinuous structure and the mixture of the two polymers becomes conductive. Feng and Chan<sup>19</sup> investigated the electric conductivity and morphology of CB-filled immiscible blends of poly(vinylidene fluoride) (PVDF) and high-density polyethylene (HDPE). CB selectively located in the HDPE phase in HDPE/PVDF/CB composites and they found out the CB content and PVDF/HDPE volume ratio were the two main factors influencing the electric conductivity. At a fixed PVDF/HDPE volume ratio of 1/1, a percolation threshold of 0.037 volume fraction of CB was observed, and that value was much lower than that for conventional CB-filled polymer composites. At a fixed CB content (10 wt % CB), a maximum electric conductivity was observed at a PVDF/HDPE volume ratio of 2.75. Tchoudakov<sup>20</sup> studied the electric resistivity and morphology of high impact polystyrene (HIPS)/styrene-isoprene-styrene copolymer (SIS)/CB composites. Conductive CB particles locate preferentially within the HIPS phase of the HIPS/SIS blends. The blends studied remain conductive as long as HIPS maintains a continuous phase and the effective CB concentration within HIPS surmounts its percolation threshold.

Although there are many studies on the effect of morphology on the electrical conductivity of CB-filled immiscible polymer blends. There is a long

**TABLE II**  
**Samples of Blending Experiment 2**

Sample code	Components	Composition (g)
2a	PS/PA6/CB	20/30/3
2b	PS/PA6/CB	30/20/3
2c	PS/PA6/CB	35/15/3
2d	PMMA/PP/CB	15/35/3
2e	PMMA/PP/CB	20/30/3
2f	PMMA/PP/CB	25/25/3
2g	PMMA/PP/CB	30/20/3
2h	PMMA/PP/CB	35/15/3
2i	PP/HDPE/CB	15/35/3
2l	PP/HDPE/CB	25/25/3
2m	PP/HDPE/CB	35/15/3

way before knowing the exactly relationship between morphology and electric conductivity of CB-filled immiscible polymer blends. This article discussed some aspects of the effect of morphology on the conductivity of ternary composites.

## EXPERIMENTAL

### Materials

Nylon6 (PA6), 1013B, supplied by Ube Industries, Japan; Polystyrene (PS), GPPS 158K, supplied by Basell, Germany; Polymethyl methacrylate (PMMA), MH/LG2, supplied by Sumitomochemical, Japan; Polypropylene (PP), F1001, supplied by China Petroleum and Chemical Corp.; Polyacrylonitrile (PAN), supplied by China Petroleum and Chemical Corp.; HDPE, 5000s, supplied by Basell, Germany; CB, HCB-A, supplied by Agrofert, Czechoslovakia; Tetrahydrofuran (THF) and formic acid.

### Preparation of ternary composites

Prior to blending, PA6 was dried at 100°C in vacuum for about 12 h. Unless otherwise specified, the ternary composites were prepared by melt-mixing of the dry-blended components in a Haake HBI System90 Rheometer (Germany) simultaneously at 230°C and 100 rpm for 10 min. The compositions of three series of the composites are listed in Tables I–III.

### Characterization

The composites were compression-molded at 200°C for 3 min to form round slice (thickness = 3 mm, diameter = 2 cm), then covered these slices with copper net on both sides, and repeated the compression molding process in the same condition to get the samples, which were used to measure electric conductivity. The electric conductivity of the composites samples were measured with EDM-1341 multimeter (Taiwan).

The distribution state of CB particles and the morphology of ternary composites were determined by using a JEM- 200EX TEM (Japan) at an acceleration voltage of 120 KV and a SIRION-100 FSEM (Holland) at an acceleration voltage of 10 KV.

For quantificationally counting the statistical average critical distance between PA6/CB dispersed

**TABLE III**  
**Samples of Blending Experiment 3**

Sample code	Components	Composition (g)
3a	PS/PA6/CB	40/10/3
3b	PMMA/PA6/CB	40/10/3
3c	PP/PA6/CB	40/10/3

phase particles of PS/PA6/CB composites, 500 diameters of PA6/CB dispersed phase particles and 500 distances between PA6/CB dispersed phase particles on the TEM pictures of composites made in blending experiment 3 were counted with Image Tool software and were averaged to get the statistical average distance between PA6/CB dispersed phase particles.

## RESULTS AND DISCUSSION

### Distribution state of CB particles in PS/PA6/CB ternary composites

Four different methods were used to judge the distribution state of CB particles in CB-filled PS/PA6 ternary composites.

### Combination of TEM and SEM characterization

TEM micrographs of PS/PA6 (50/50) blends filled with different amount of CB are shown in Figure 1. It can be seen that CB particles exist in only one phase even when the CB content is as high as 16 phr.

SEM micrographs of PS/PA6 (50/50) blends filled with 6 phr CB are shown in Figure 2. The fracture

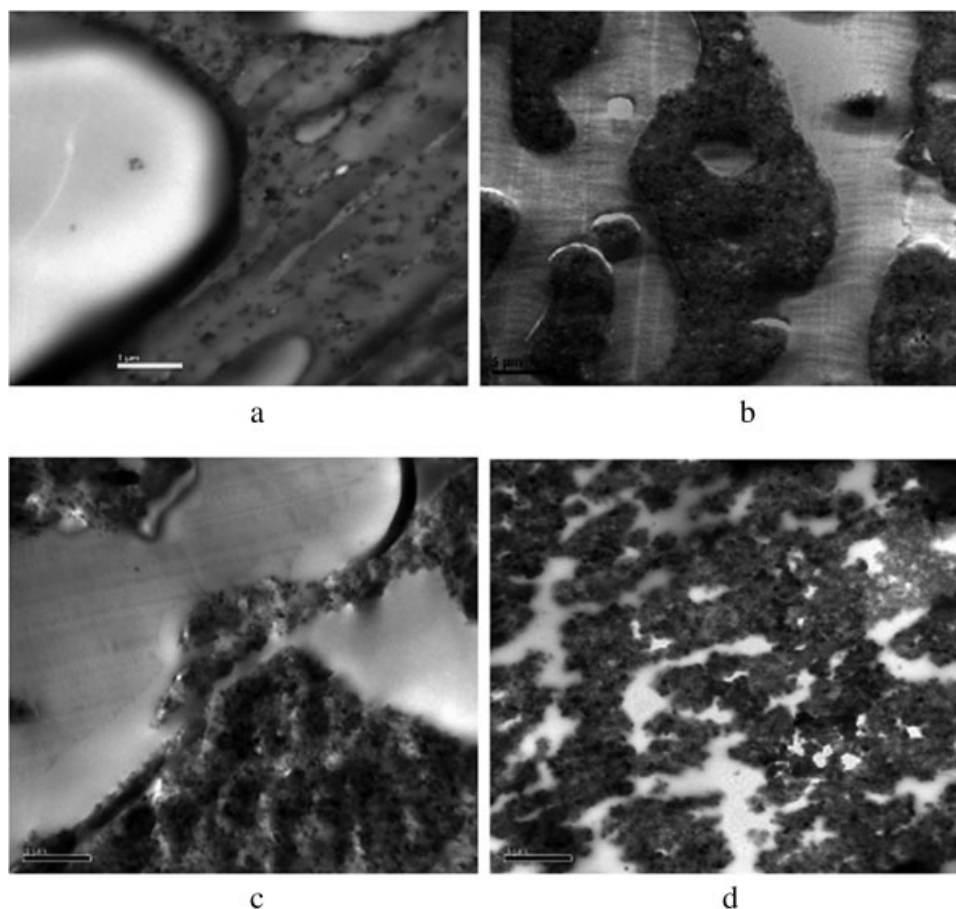
surfaces were etched with formic acid and THF, respectively. It can be seen from Figure 2(a) that CB particles do not exist on the fracture of PS when the PA6 phase was dissolved by formic acid. Contrarily, it can be seen from Figure 2(b) that CB particles exist on the fracture of PA6 when the PS phase was dissolved by THF.

From the TEM and SEM micrographs of CB-filled PS/PA6 (50/50) composites, we can get the conclusion that CB particles distribute in PA6 phase only in PS/PA6 blends.

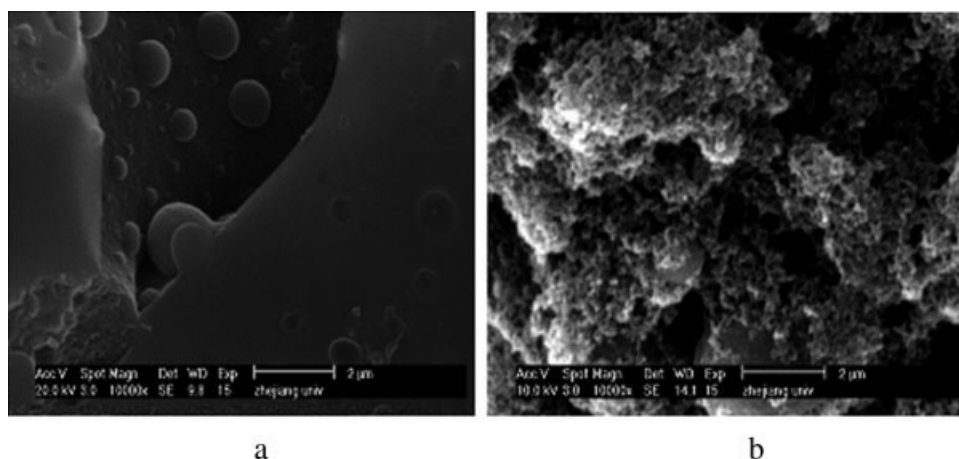
### TEM micrograph of CB-filled PS/PA6 blends with different mass ratio of PS and PA6

TEM micrographs of CB-filled PS/PA6 blends with different mass ratio of PS and PA6 are shown in Figure 3.

First, it can be seen from Figure 3(a,b) that CB particles distribute in the dispersed phase and do not distribute in the continuous phase. Second, from the mass ratio of PS and PA6 (40 g/8 g and 40 g/10 g) we know that PA6 is dispersed phase and PS is the continuous phase. It can be seen from Figure 3(c) that CB particles distribute in the continuous phase



**Figure 1** TEM micrograph of PS/PA6 (50/50) blends filled with CB. (a) 2 phr CB, (b) 6 phr CB, (c) 10 phr CB, (d) 16 phr CB.

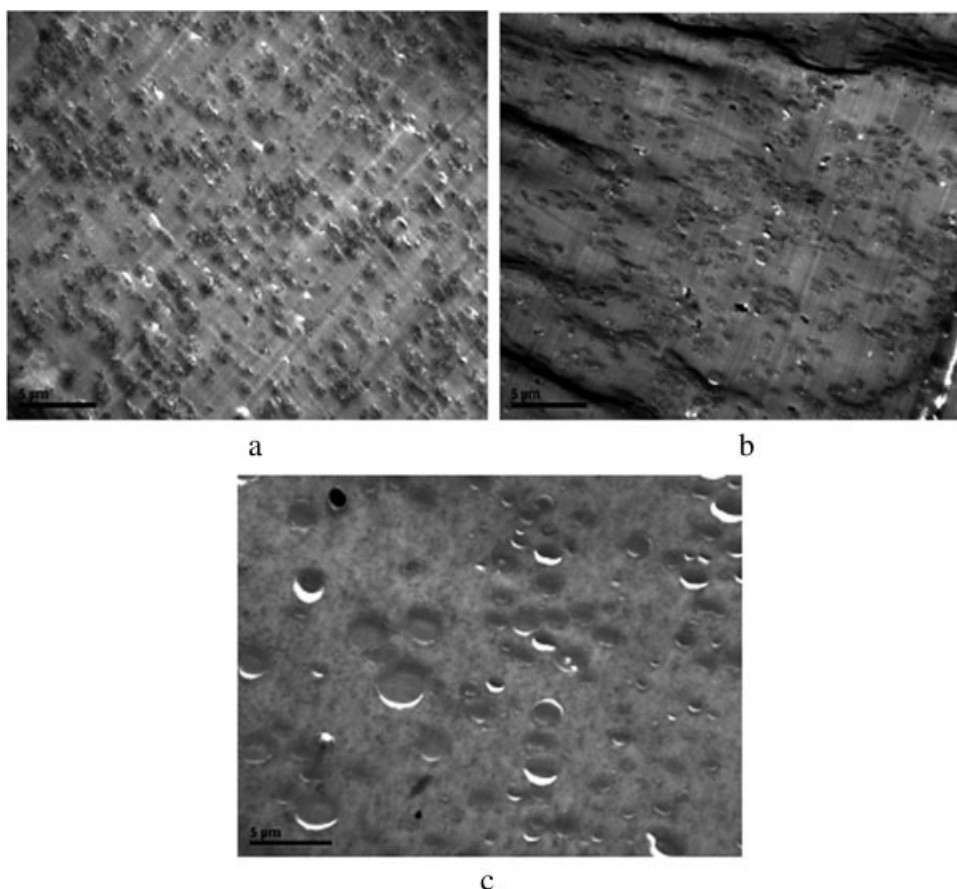


**Figure 2** SEM micrographs of fractured surfaces of a PS/PA6 (50/50) blend filled with 6 phr CB. (a) PA6 is dissolved by formic acid, (b) PS is dissolved by THF.

and do not distribute in the dispersed phase. And from the mass ratio of PS and PA6 (10 g/40 g) we know that PS is dispersed phase and PA6 is the continuous phase. From the earlier discussion we can conclude that CB particles distribute in PA6 phase only in PS/PA6 blend.

#### Selectively dissolving by solvent

THF and formic acid solvent were used to dissolve the PS and PA6 of PS/PA6/CB composites, respectively. Five days later, formic acid (the solvent for PA6) turned to being black and THF (the solvent for PS) kept being achromaticity. This phenomenon proved that CB



**Figure 3** TEM micrograph of PS/PA6/CB composites with different mass ratio of PS and PA6 (in g/g/g). (a) 40/8/3, (b) 40/10/3, (c) 10/40/3.

particles distributed in PA6 phase in PS/PA6/CB composites only: PA6 was dissolved by formic acid solvent, and CB particles entered into formic acid solvent along with PA6 at the same time, which made the formic acid solvent turn black. PS was dissolved by THF solvent, but there is no CB particles in PS phase, so there are no CB particles entering into THF solvent, which make the THF solvent keep being achromaticity.

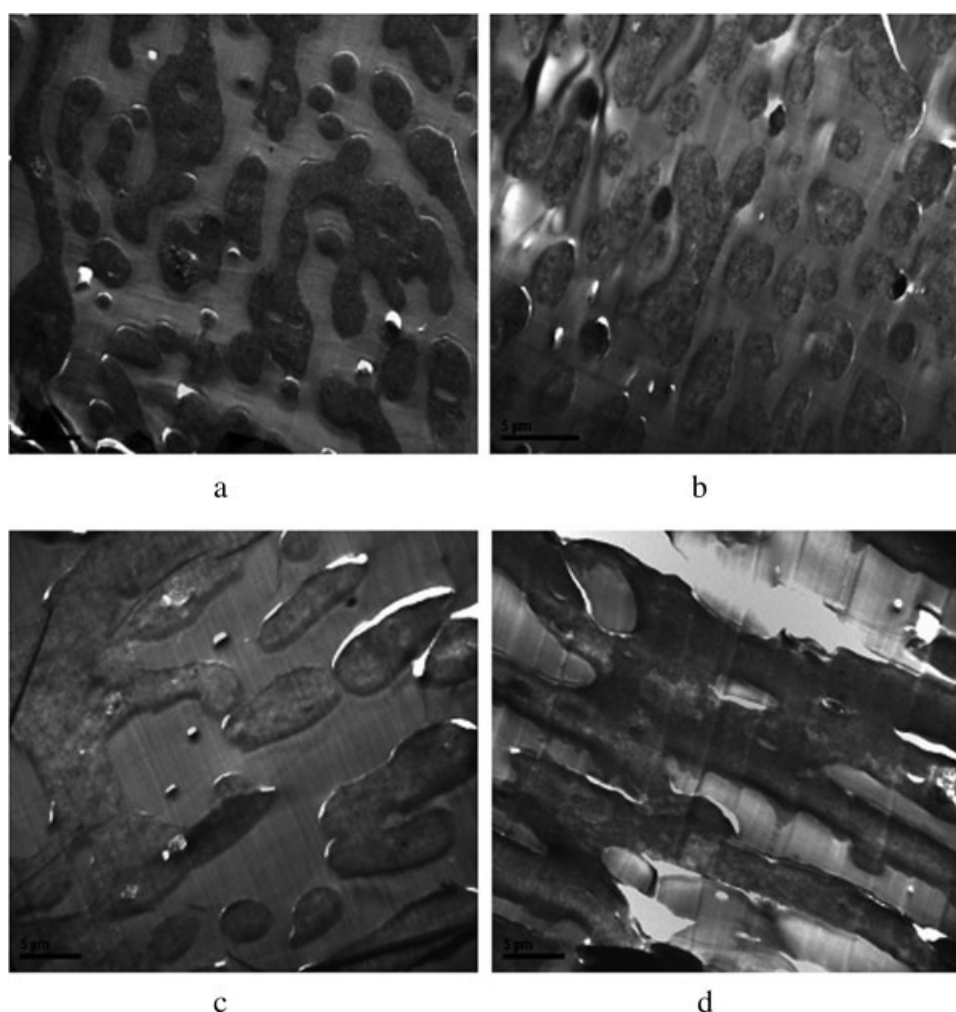
#### Transplant phenomenon between PS and PA6 phase of CB particles

First, CB particles were mixed with one polymer of PS and PA6 for 10 min, respectively. Then, another polymer of PS and PA6 was added and the mixing was continued. According to the remixing time, we can get different samples, the TEM micrographs were shown in Figures 4 and 5.

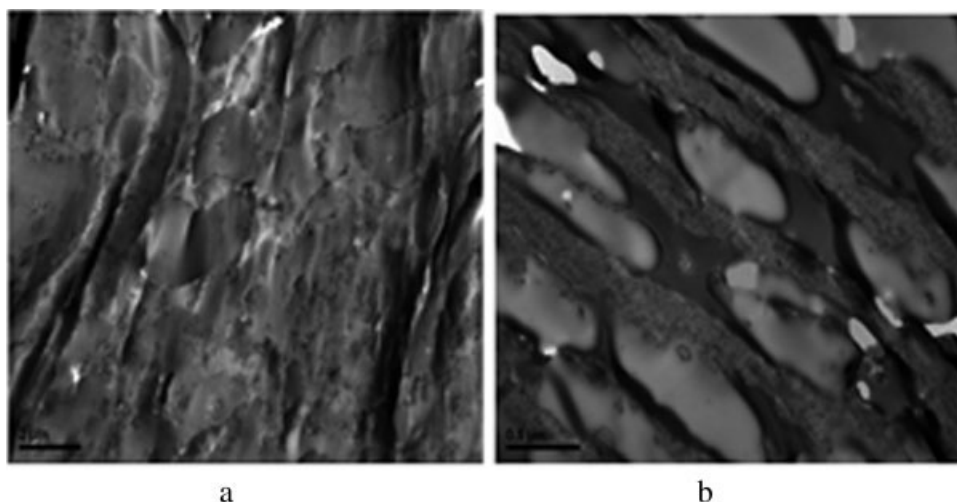
It can be seen from Figure 4 that if CB (3 g) particles were mixed with PA6 (25 g) first for 10 min and PS (25 g) was filled later, no matter how long the remixing time was, there was no transplant phe-

nomenon of CB particles between PS and PA6 phase, CB particles existed in one phase only. However, if CB (3 g) particles were mixed with PS (25 g) first for 10 min and PA6 (25 g) was filled later, the phenomenon is quite different. When the remixing time of PA6 is 0.5 min, we can see from Figure 5(a) that CB particles located in both phases, inferring that part of CB particles transferred from PS phase to PA6 phase. And when the remixing time is 2 min, we can see from Figure 5(b) that CB particles existed in one phase only again, indicating that all CB particles has migrated from PS phase to PA6 phase.

In a word, four different methods were used to judge the distribution state of CB particles in PS/PA6 blends, and we get the same conclusion that CB particles distribute in PA6 phase only in the PS/PA6/CB composites. By using these four methods we can also judge the distribution state of CB particles in other different binary polymer blends, such as PMMA/PP, PP/HDPE, PP/PA6, PMMA/PA6, and PAN/PA6 blends.



**Figure 4** TEM micrograph of PA6/CB (25 g/3 g) and PS (25 g) blends with different blending time of PS. (a) 0.5 min, (b) 2 min, (c) 3 min, (d) 5 min.



**Figure 5** TEM micrograph of PS/CB (25 g/3 g) and PA6 (25 g) blends with different blending time of PA6. (a) 0.5 min, (b) 2 min.

#### Distribution states of CB in different binary polymer blends

Different distribution states of CB in different binary polymer blends are shown in Figure 6. It can be clearly seen that CB particles located preferentially at the interface between PMMA and PP in PMMA/PP/CB or in the HDPE in PP/HDPE/CB composites, respectively. CB particles all located in the PA6 in PS/PA6/CB, PP/PA6/CB, PMMA/PA6/CB, and PA6/PAN/CB composites.

Different distribution states of CB in different composites are the result of difference in polarity and melt viscosity of polymers. Generally, if the difference between the viscosities of two polymers is not obvious, CB particles readily disperse into polymer having stronger polarity and if none of the two components has strong polarity the CB particles locate preferably at the interface of two polymers. In comparison, if the difference between the polarities of two polymers is not obvious, the melt viscosity of polymers will play an important role on the distribution state of CB particles in binary polymer blends. As an example, for ternary composites of PMMA/PP/CB, Feng et al.<sup>21</sup> discovered that the dispersion of CB was strongly influenced by the viscosity of PMMA. The CB was dispersed in the PMMA phase when the viscosities of PMMA and PP are comparable. As the viscosity of PMMA increases, the CB was found to be located at the interface between the PMMA and PP phases. On further increase in the viscosity of PMMA, the CB was found to be dispersed in the PP phase.

#### Increase of the electric conductivity of PA6/CB composites due to existence of PS or PAN

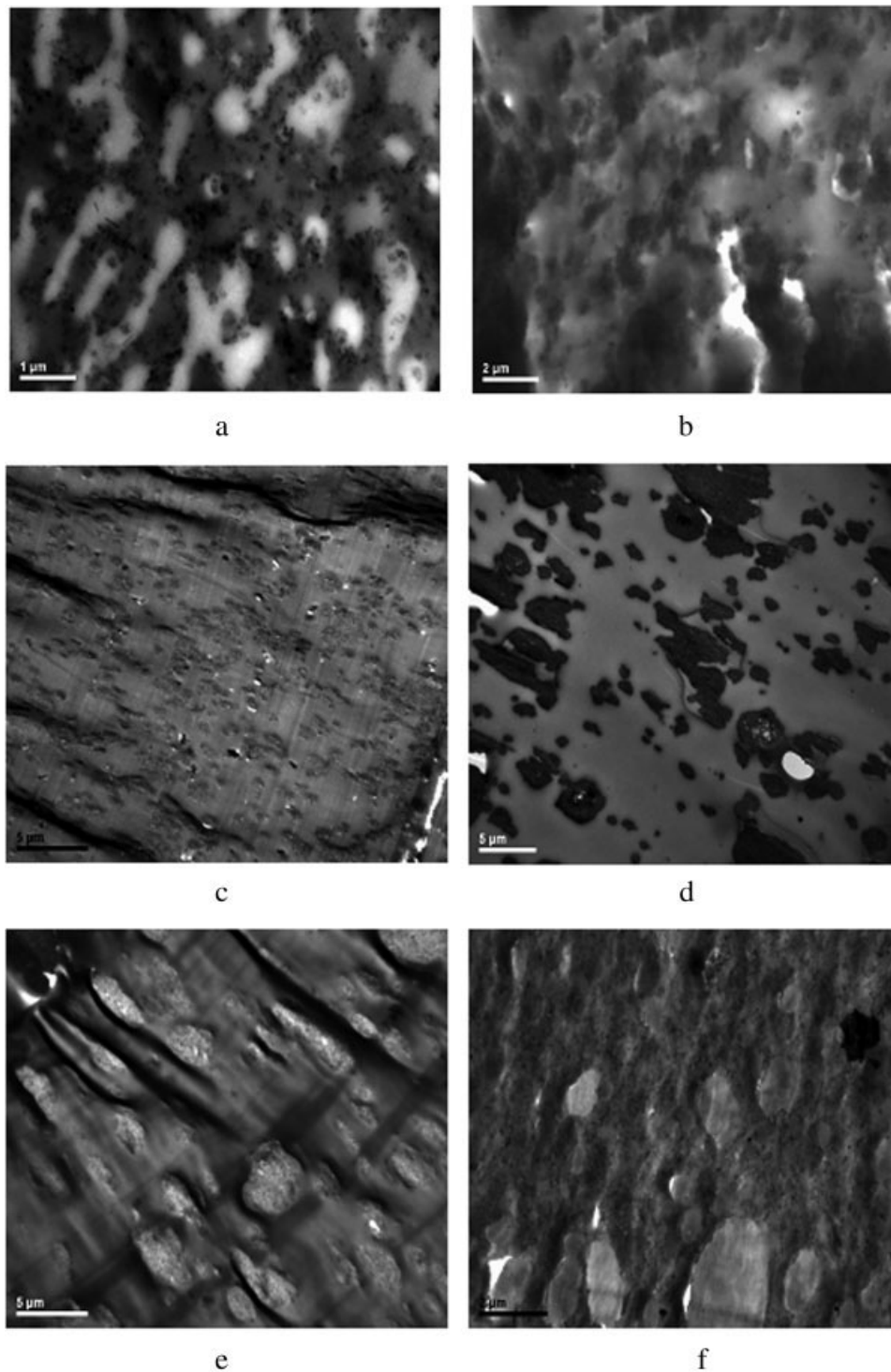
It is known from Figure 6 that CB particles sometimes can only distribute in one polymer phase in immisci-

ble binary polymer blend. Then, how does the electric conductivity change if a second polymer is mixed into CB-filled single polymer and the CB particles still stay in the original polymer only? To find the answer we design the CB-filled PS/PA6 and PAN/PA6 immiscible polymer blends, in which CB particles both selectively distribute in PA6. That is, if a certain amount of PS or PAN polymers are mixed into PA6/CB composites, the fact that CB particles exist in PA6 only and the content of CB in PA6 phase are unchanged, the only thing changed is the morphology of PA6/CB phase. And the effect of it on the electric conductivity of CPCs is shown in Figures 7 and 8.

It can be clearly seen that existence of PS or PAN makes the electric conductivity of PA6/CB composites higher, but if the PS content is too much the electric conductivity decreases, which has been discussed.<sup>22</sup> The reason of the increase of the electric conductivity of PA6/CB composites due to existence of a little PS or PAN is the parallel electrocircuit made by PS and PAN phase. As it has been discussed, CB particles only distribute in PA6 phase in PS/PA6/CB and PAN/PA6/CB composites. And existence of PS and PAN phase where has no CB particles creates a parallel electrocircuit for electrons to transmit, which is shown in Figures 9 and 10. Furthermore, it is well known that parallel electrocircuit can decrease resistivity. Hence, addition of PS or PAN makes the electric conductivity of PA6/CB composites increase.

#### Effect of the morphology of PA6/CB dispersed phase on the electric conductivity of three different ternary composites

PS/PA6/CB, PMMA/PA6/CB, and PP/PA6/CB composites in which CB particles all distribute in

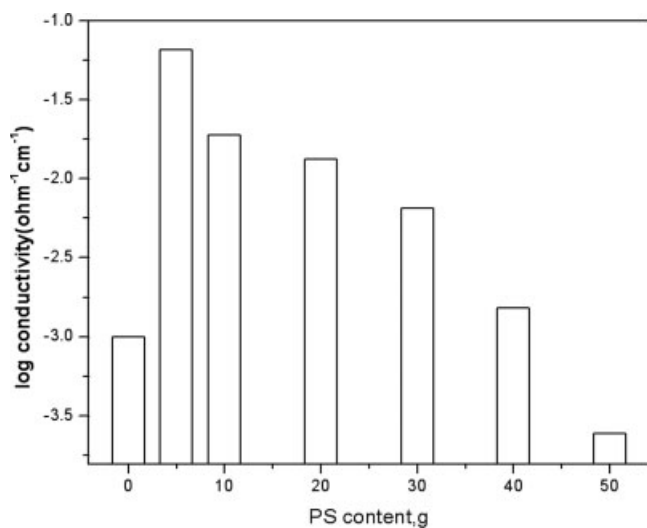


**Figure 6** TEM micrographs of different ternary composites. (a) PMMA/PP/CB (25/25/3); (b) PP/HDPE/CB (35/15/3); (c) PS/PA6/CB (40/10/3); (d) PMMA/PA6/CB (40/10/3); (e) PP/PA6/CB (40/10/3); (f) PA6/PAN/CB (30/10/5).

PA6 phase shown in Figure 6 are chosen, and the PA6/CB phase is designed to be dispersed phase to study the effect of the morphology of PA6/CB dispersed phase on the electric conductivity of three different ternary composites. It can be seen from Figure 11 that the PS/PA6/CB composites have the highest electric conductivity and the PP/PA6/CB

composites have the lowest electric conductivity as the mass ratio of PS/PA6, PP/PA6, and PMMA/PA6 are the same and PA6/CB is dispersed phase.

It can be clearly seen from Figure 6 that CB particles distribute in PA6 and PA6/CB phase is dispersed phase in all the three composites. The difference between them is the distance between PA6/CB

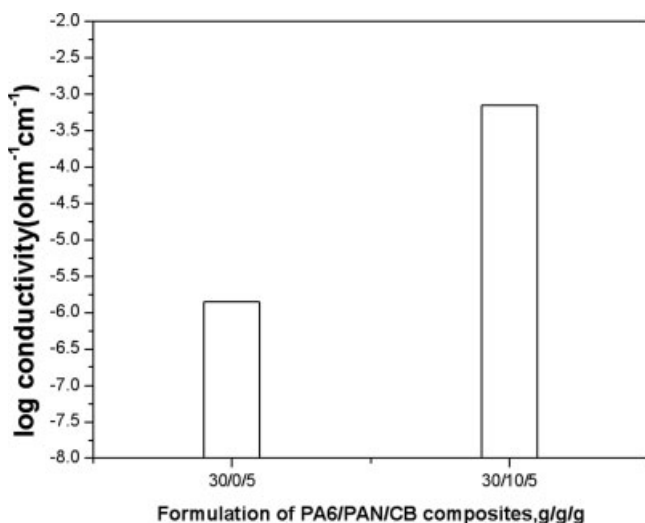


**Figure 7** Influence of PS content on the electric conductivity of PA6/CB (10/3) composites.

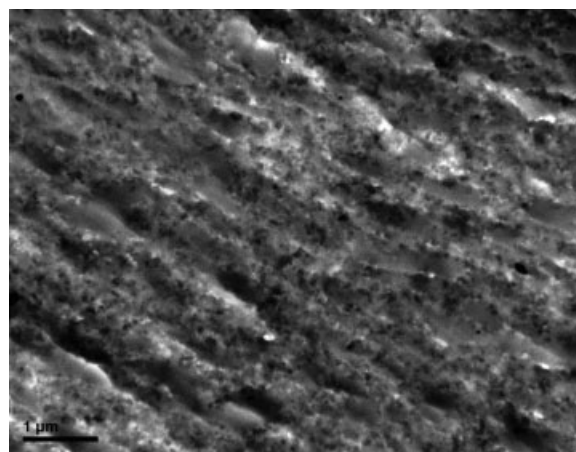
particles, which may be a reason why these three composites have different electric conductivity.

The average distance between PA6/CB particles of three ternary composites are shown in Figure 12. It can be seen from Figure 12 that under the condition CB content equals, the distance between PA6/CB particles in PS/PA6/CB composites is the smallest and the distance between PA6/CB particles in PP/PA6/CB composites is the largest among the three composites.

The difference of the distance between PA6/CB particles leads to the different electric conductivity of the three composites. The bigger the distance between PA6/CB particles is the harder for the electron to traverse the second polymer phase between



**Figure 8** Comparison of the electric conductivity of PA6/CB (CB content in PA6 is 17 phr) blend and PA6/PAN/CB composites.



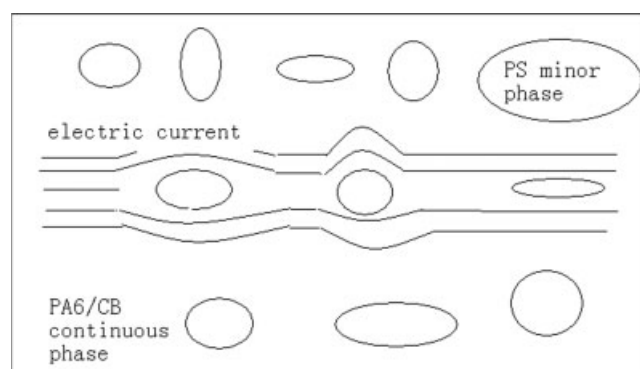
**Figure 9** TEM micrographs of PS/PA6/CB (5/10/3) composite.

PA6/CB particles and the lower the electric conductivity the ternary composites have. Hence, PS/PA6/CB composites have the highest electric conductivity and PP/PA6/CB composites have the lowest electric conductivity among the three composites.

#### Effect of distribution state of CB on the optimization phase construction of two components which makes ternary composites have the highest electric conductivity

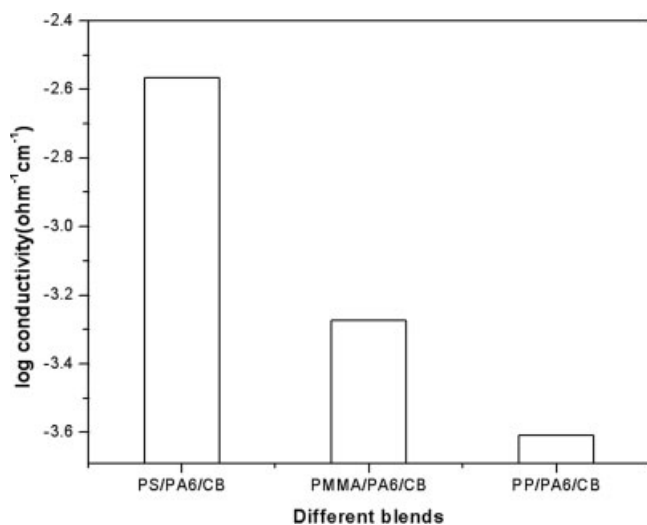
Because of different CB particles distribution state, CB-filled PMMA/PP blend in which CB locate preferentially at the interface of the two polymers and CB-filled PS/PA6 and PP/HDPE blends in which CB locate in PA6 and HDPE especially are chosen to study the effect of distribution state of CB on the optimization phase construction of two components, which makes ternary composites have the highest electric conductivity.

The effect of the mass ratio of two polymer components on the electric conductivity of different composites was shown in Figure 13. It can be clearly



**Figure 10** The parallel electrocircuit model of PS/PA6/CB composites.

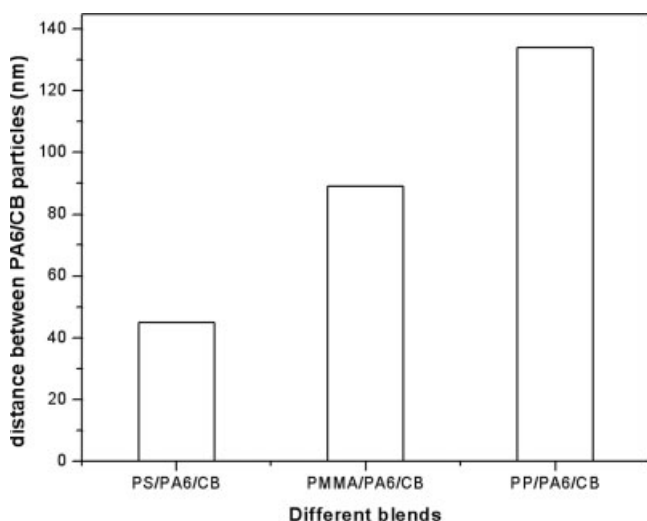




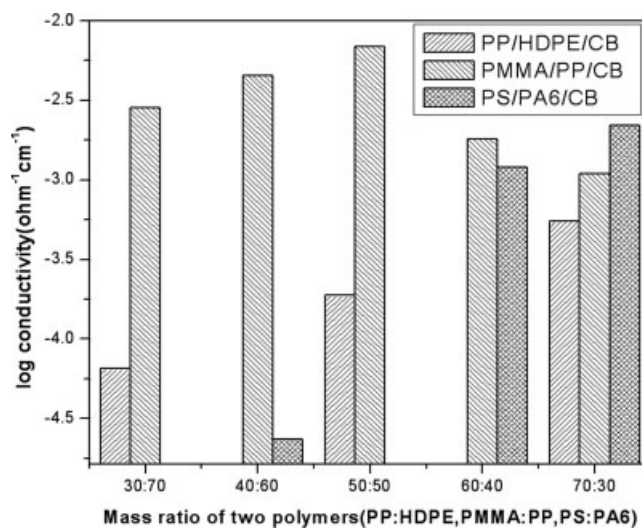
**Figure 11** Electric conductivity of different composites (40/10/3).

seen that under the condition the mass of CB particles and the total mass of two polymer components in ternary composites keeping constant and the mass ratio of two polymer components changing, PMMA/PP/CB composites have the highest electric conductivity when the mass ratio of PMMA and PP is 50 : 50 (sample 2f), in which PMMA and PP are cocontinuous as shown in Figure 1. But both PS/PA6/CB and PP/HDPE/CB composites have higher electric conductivity when the PA6 and HDPE are dispersed phase than continuous phase.

When the CB particles locate preferentially in PA6 in PS/PA6/CB composites, both effective amount of CB inside unit volume of PA6 and the effective distance between conductive domains (PA6/CB) affecting the conductivity of the composites. Under the



**Figure 12** Distance between PA6/CB particles of different composites (40/10/3).



**Figure 13** Effect of the mass ratio of two components on the electric conductivity of different composites (CB content in composites is 6phr).

condition CB content in binary polymer blends keeping constant, making effective amount of CB inside unit volume of highest in PA6 and making the continuous degree highest of PA6/CB by changing the mass ratio of PS/PA6 are ambivalent. The result of the competition between them is an optimization mass ratio of PS/PA6, at which PA6 is less than PS, making the effective amount of CB inside unit volume of PA6 higher and making the distance between particles of PA6/CB not too big at the same time, which makes the PS/PA6/CB composites have the highest electric conductivity.

For the PP/HDPE/CB composites we can get the same conclusion as PS/PA6/CB composites, and the reason is the same: There is an optimization mass ratio of PP/HDPE, at which HDPE is less than PP, making the effective amount of CB inside unit volume of HDPE higher and making the distance between particles of HDPE/CB not too big at the same time, which makes the PP/HDPE/CB composites have the highest electric conductivity.

When the CB particles locate preferentially at the interface of the two polymers in PMMA/PP/CB composites, because of the small volume of the interface, the CB density at the interface readily keeps a high level, which makes its effect on the electric conductivity of ternary composites less distinct. At this time the continuous degree of the interface itself decides the electric conductivity of ternary composites. The cocontinuous state of the two polymers in ternary composites makes the continuous degree of the interface highest. Hence, when the CB particles locate preferentially at the interface of the two polymers the ternary composites have the highest electric conductivity when the two polymers are cocontinuous.

In a word, under the condition the mass of CB particles and the total mass of two polymer components in ternary composites keeping constant and the mass ratio of two polymer components changing, the CB distribution state decides the optimization phase construction of two polymer components, which makes ternary composites have the highest electric conductivity. For the composites where CB particles locate at the interface of two polymer components, the ternary composites have the highest electric conductivity when two polymer components are cocontinuous, e.g., the mass ratio of the two polymers is near to 1 : 1. For the composites where CB particles located preferentially in one polymer, the ternary composites have the highest electric conductivity usually when the amount of the polymer component in which the CB particles distribute is comparatively less than the polymer component in which the CB particles do not distribute.

### CONCLUSIONS

1. Under our experimental condition, CB particles located preferentially at the interface between PMMA and PP in PMMA/PP/CB composites and in the HDPE phase in PP/HDPE/CB composites, respectively, and in the PA6 phase in PS/PA6/CB, PP/PA6/CB, PMMA/PA6/CB, and PA6/PAN/CB composites.
2. Existence of PS or PAN makes a parallel electrocircuit for electrons to transmit, which makes the electric conductivity of PA6/CB composites increase.
3. Under the condition that the formulations of PS/PA6/CB, PP/PA6/CB, and PMMA/PA6/CB composites equal in which PA6/CB is dispersed phase, the difference of the distance between PA6/CB particles leads to the different electric conductivity of three composites. The distance between PA6/CB particles in PS/PA6/CB composites is the smallest among the three composites, and PS/PA6/CB composites have the highest electric conductivity. The distance between PA6/CB particles in PP/PA6/CB composites is the largest among the three compo-

sites and consequently PP/PA6/CB composites have the lowest electric conductivity.

4. For the composites where CB particles locate at the interface of two polymer components, the ternary composites have the highest electric conductivity when two polymer components are cocontinuous, e.g., the mass ratio of the two polymers is near to 1 : 1. For the composites where CB particles located preferentially in one polymer, the ternary composites have the highest electric conductivity usually when the amount of the polymer component in which the CB particles distribute is comparatively less than the polymer component in which the CB particles do not distribute.

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