

ELECTRICAL PROPERTIES OF CONDUCTIVE POLYMER COMPOSITES PREPARED BY AN INNOVATIONAL METHOD*

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Abstract An innovational method that poly(styrene-co-maleic anhydride) (SMA), a compatibilizer of immiscible nylon6/polystyrene (PA6/PS) blends, was first reacted with carbon black (CB) and then blended with PA6/PS, has been employed to prepare the PA6/PS/(SMA-CB) composites of which CB localized at the interface. In PA6/PS/CB blends, CB was found to preferentially localize in the PA6 phase. However, in the PA6/PS/(SMA-CB) blends, it was found that CB particles can be induced by SMA to localize at the interface. The electrical properties of PA6/PS/(SMA-CB) composites were investigated. The results showed that the composites exhibited distinct triple percolation behavior, *i.e.* the percolation is governed by the percolation of CB in SMA phase, the continuity of SMA-CB at the interface and the continuity of PA6/PS interface. The percolation threshold of PA6/PS/(SMA-CB) was only 0.15 wt%, which is much lower than that of PA6/PS/CB. Moreover, the PTC (positive temperature coefficient) intensity of PA6/PS/(SMA-CB) composites was stronger than that of PA6/PS/CB and the negative temperature coefficient (NTC) effect was eliminated. The electrical properties of PA6/PS/(SMA-CB) were explained in terms of its special interface morphology: SMA and CB localize at interphase to form the conductive pathways.

Keywords: Polymer composites; Carbon black; Morphology; Electrical properties.

INTRODUCTION

It is well-known that conductive polymer composites (CPCs) can be produced by adding of a critical amount of carbon black (CB), which is referred to as the percolation threshold, in insulating polymers. CPCs have many applications, including antistatic materials, positive temperature coefficient (PTC) materials, electromagnetic shielding and chemical vapor detection^[1–3]. In recent years CPCs with low percolation threshold have received increasing attention because the reduction of the percolation threshold is of significance in lowering cost, easier processing and better mechanical properties. Among all of the processes investigated to reduce the percolation threshold, CB-filled immiscible polymers is the most promising method, and the decrease of percolation threshold can be attributed to the double percolation resulting from the selective distribution of CB in one of the two phases or at the interface of the polymers^[4–13]. Obviously the selective localization of the CB at the interface can provide a composite with the lowest percolation threshold. The selective distribution of CB in polymer blends is mainly due to the different affinity of CB for each component of polymer blends^[14]. For most polymer blends studied, CB showed affinity for the one component of polymer blends and did not show affinity

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for the interface^[15–20].

Up to now, very few investigations about controlling CB localization at the interface were reported. Gubbels *et al.*^[21] employed a kinetic strategy to localize CB at the interface of PS/PE blends. CB was first blended with PS for which it has the lowest affinity, and then PE for which CB has the higher affinity was added. The CB particles would transfer from the PS phase to the PE phase through the interface. At a certain mixing time, CB particles should localize at the interface. As the result of localization of CB at the interface, the percolation threshold of composites is only 0.4 wt%, being one of the lowest percolation threshold reported in the open literature. Li *et al.*^[22] also employed this method to prepared the PET/PE/CB conductive composites of CB localization at the interface. The difficulty for this strategy is to determine exactly the mixing time when CB particles are located just at the interface. Feng *et al.*^[23] found that for the blends of PP/PMMA the dispersion of CB was strongly influenced by the viscosity of PMMA. When the viscosities of PP and PMMA were comparable, the CB particles were found to be localized at the interface. This method worked in PP/PMMA blends but it may not be a universal method. Al-Saleh^[24] introduced SBS which localized at the interface of immiscible PS/PP blends and have the highest affinity for CB to induce CB localization at the interface. Obviously this method could be worked only when the affinity of CB for compatibilizers is higher than that for the immiscible polymers.

It is clear that the key factor for controlling CB localization at the interface is how to make the affinity of CB for interface more than for polymer matrix. Maleated polymers, as a widest known families of the functionalized polymers, have been employed as compatibilizers to improve the compatibility of immiscible polymer blends and polymer/inorganic filler composites in industry^[25, 26]. Considering that compatibilizer will selectively localize at the interface of immiscible polymer blends as well as the reaction between CB and maleated polymers will result in their strong affinity, an innovational method that maleated polymers are first reacted with CB, and then blended with immiscible polymer blends would be effective to control CB localization at the interface.

Based on this, in this paper, in order to prepare the CPCs of which CB is localized at the interface, a method that poly(styrene-*co*-maleic anhydride) (SMA) was first reacted with CB, and then blended with PA6/PS was introduced to prepare the PA6/PS/(SMA-CB) composites. The morphology and the percolation threshold, percolation phenomenon and positive temperature coefficient (PTC) effect were also investigated.

EXPERIMENTAL

Materials

The materials used in this study were polystyrene (PG-383M, MI = 8.5 g/10min, $d = 1.05 \text{ g/cm}^3$) supplied by Zhenjiang Chi Mei Chemicals Co., Ltd and polyamide-6 (PA-6) (33500, relative viscosity is 3.50, $d = 1.14 \text{ g/cm}^3$), supplied by Xinhui Meida-DSM Nylon Chips Co., Ltd. The compatibilizer employed in this study was SMA (MPC1501, Shanghai SUNNY New Technology Development Co., Ltd.). The amount of maleic anhydride in SMA was 18 wt%. An electrically conductive grade carbon black supplied by Shandong Ling Zhi Chemicals Co., Ltd (HG-1P, average diameter is 30 nm) was used as conductive filler.

Preparation of CPCs

Based on the condition of the reaction between hydroxyl group at CB surface and maleic anhydride in the SMA^[27], the SMA/CB composites were prepared as follows: SMA and CB with desired proportions were dissolved in dimethylbenzene and then reacted at 120°C for 24 h. The reactant was deposited with acetone at room temperature and then dried in a vacuum oven at 90°C for 24 h to remove the solvent. The content of CB in SMA-CB was 10%, 15%, 20% and 30%.

The conductive polymer composites were prepared using a Haake mixer at 240°C and 60 r/min for 10 min. The materials obtained were further compressed into 40 mm × 40 mm × 1 mm sheets by a hot press at 240°C for electricity tests. In order to reduce contact resistivity, the two sides of the samples were bonded with aluminum foil.

Measurements and Characterization

In order to investigate the amount of SMA reacted with CB, SMA in SMA-CB (80/20) was extracted for 48 h by acetone using Soxhlet extractor. The ratio of SMA reacted with CB was calculated according to Eq. (1).

$$\text{Ratio (\%)} = \left(1 - \frac{(\text{SMA} - \text{CB})_i - (\text{SMA} - \text{CB})_f}{(\text{SMA} - \text{CB})_i \times 0.8} \right) \times 100\% \quad (1)$$

where $(\text{SMA} - \text{CB})_i$ and $(\text{SMA} - \text{CB})_f$ are the initial and final mass of SMA-CB, respectively.

FTIR spectra were recorded on a Bruker Vector 33 spectrometer. The remainder of SMA-CB extracted by acetone and CB were pressed into disks with KBr. SMA was prepared into film.

The electrical resistivity of conductive composites was measured by using a multimeter when the resistivity was lower than $2 \times 10^7 \Omega$. The measurements were done by using ZC-36 High Resistance Electrometer when the resistivity exceeded $2 \times 10^7 \Omega$. The temperature dependence of composites resistivity was measured by heating the samples at a rate of 1 K/min in an oven.

Transmission electron microscope (TEM) micrographs were taken using an FEI TECNAI-G20 microscope at an acceleration voltage of 300 kV. Ultra thin sections of 70 nm in thickness were cryogenically cut with a diamond knife at -100°C .

Optical microscope (OM) was taken to study the morphology of composites. Thin sections were prepared by a cryomicrotome at -100°C .

RESULTS AND DISCUSSION

SMA-CB Composites

Figure 1 shows the FTIR spectra of remainder of SMA-CB (80/20) extracted by acetone, SMA and CB. The obvious absorption peak of CB can not be observed due to the strong light-absorption of CB. Although the FTIR spectrum of remainder of SMA-CB extracted by acetone is similar to that of the CB due to the strong light-absorption of CB, the absorption bands at 2923 cm^{-1} and 2855 cm^{-1} assigned to the $-\text{CH}_2$ stretching vibrations can be observed, indicating the residue of SMA in the extracted SMA-CB. The result of extracting experiment can identify that the remainder of SMA-CB (80/20) extracted by acetone contains 46% SMA. The residue of SMA in the extracted SMA-CB indicated that SMA had reacted with CB and could not be extracted by acetone.

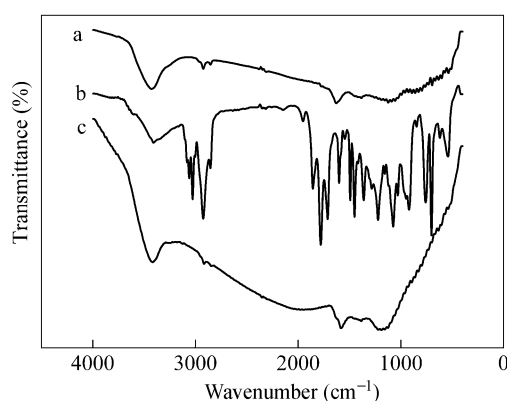


Fig. 1 FTIR spectra of (a) remainder of SMA-CB extracted by acetone, (b) SMA and (c) CB

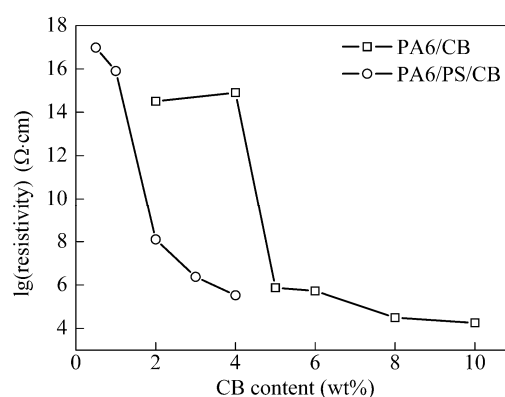


Fig. 2 Effect of CB content on the room-temperature resistivity of PA6/CB and CB-filled PA6/PS (60/40) composites

Percolation Phenomena of PA6/CB and PS6/PS/CB

Figure 2 shows the influence of CB content on the room-temperature resistivity of the composites. At lower CB contents (2 wt%–4 wt%), the composites of PA6/CB is insulating. When CB content increases to 5 wt%, the resistivity of the composites falls down sharply from $10^{15} \Omega \cdot \text{cm}$ to $10^6 \Omega \cdot \text{cm}$, indicating that the percolation

threshold of PA6/CB composites is 5 wt%. From Fig. 2 it can be observed that the percolation threshold of CB-filled PA6/PS (60/40) composites is 2 wt%, which is lower than that of PA6/CB composites (5 wt%), due to the double percolation. The OM result shows that 2 wt% CB-filled PA6/PS (60/40) exhibits co-continuous morphology and the CB is selectivity localized in one phase (Fig. 3). In order to investigate the localization of CB, the dimethylbenzene or formic acid was used to extract the PS phase (Fig. 4a) or PA6 phase (Fig. 4b) of PA6/PS/CB composites. It can be seen that the solution of the sample extracted by dimethylbenzene is transparent, while the solution of the sample extracted by formic acid is dark, indicating the localization of CB in PA6 phase.

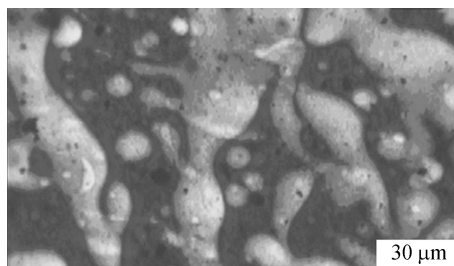


Fig. 3 OM image of 2 wt% CB-filled PA6/PS (60/40) blend

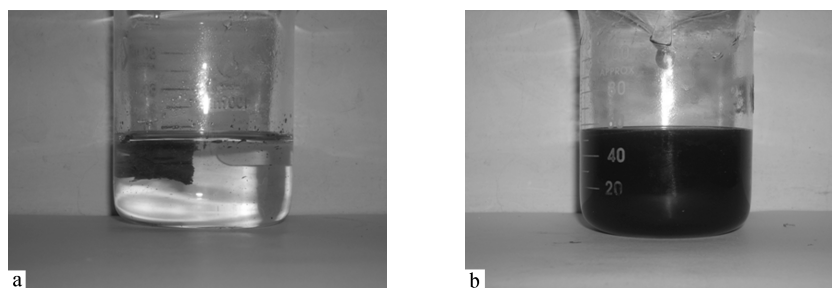


Fig. 4 Solutions obtained by etching 2 wt% CB-filled PA6/PS (60/40) blends with different solvents: (a) dimethylbenzene and (b) formic acid

TEM of PA6/PS/(SMA-CB) Composites

In order to investigate the distribution of CB in the composites of PA6/PS/(SMA-CB), TEM was applied. As shown in Fig. 5, the interface of PA6/PS can be observed clearly. The PS phase is white, the PA6 phase is gray, and the interface between PA6 and PS is dark, indicating that the CB particles are selectively localized at the interface. It is well-known that the compatibilizer will selectively localize at the interface of immiscible polymer blends to reduce the interfacial tension. SMA copolymer, as the compatibilizer of PS/PA6 blends, should congregate at the interface of PA6/PS blends. Furthermore, as SMA had reacted with CB before being introduced into the PA6/PS blends, so the affinity of CB for SMA was stronger than that for PA6 and PS. Therefore, the CB particles can be localized at the interface by SMA spontaneously during the processing of PA6/PS/(SMA-CB) blends.

The localization of CB at the interface induced by SMA indicates that the interface of PA6/PS/(SMA-CB) contains SMA and CB, thus resulting in interface morphology more complicated than that of CPCs of which the interface contains CB only^[21–24]. The complicated interface morphology of PA6/PS/(SMA-CB) blends may imply special electrical properties. Based on this, the percolation behaviors, percolation threshold and PTC effect of PA6/PS/(SMA-CB) composites were investigated.

Percolation Phenomena of PA6/PS/(SMA-CB) Composites

The effect of adding 6 wt% SMA-CB (70/30) composites on the room-temperature resistivity of PA6/PS blends

as a function of PA6 concentration is shown in Fig. 6. The results demonstrate that PA6 concentration has a significant effect on the volume resistivity of PA6/PS/(SMA-CB) blends. The blends are insulating when the concentration of PA6 in PA6/PS blends is greater than 70% or less than 40%. The conductive composites can be prepared when the PA6/PS ratios are between 50/50 and 60/40. The lowest volume resistivity appears at the ratio of 60/40. So in the following research, the ratio of PA6/PS was fixed at 60/40.

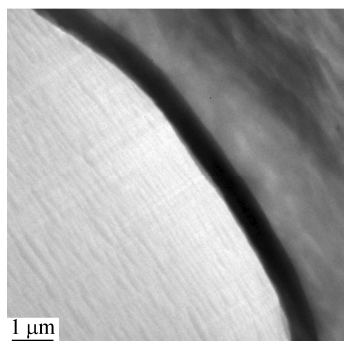


Fig. 5 TEM photograph of 2 wt% SMA-CB (70/30) filled PA6/PS (60/40) blend

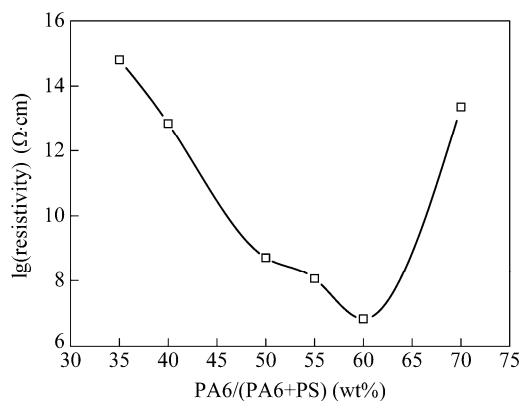


Fig. 6 Effect of the PA6 concentration on the room-temperature resistivity of 6% SMA-CB filled PA6/PS

The morphologies of PA6/PS/(SMA-CB) are investigated by OM in order to explain the drastic change of composite conductivity, as shown in Fig. 7. For the composites that the mass ratios of PA6/PS are 70/30 and 35/65, the sea-islands morphologies are observed by OM, indicating that the PA6/PS interface is discontinuous and CB can not establish continuous conductive pathways. On the other hand, PA6/PS co-continuous morphology is observed by OM at the PA6/PS mass ratio of 60/40, indicating that the PA6/PS interface is continuous and the continuous conductive pathway at the interface can be established by CB. So it can be conclusion that the percolation of PA6/PS/(SMA-CB) blends can be governed by the interface continuity between PS and PA6 phases.

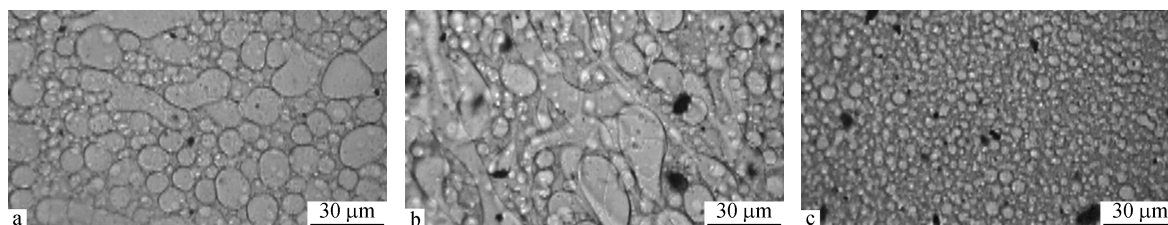


Fig. 7 OM photographs of 6% SMA-CB filled PA6/PS composites at different ratio of PA6/PS: (a) 35/65, (b) 60/40 and (c) 70/30

The effects of loading SMA-CB on the room-temperature resistivity of PA6/PS (60/40) as a function of CB/(SMA-CB) mass ratios are studied, as shown in Fig. 8. The room-temperature resistivity of SMA-CB composites at various SMA/CB ratios is also shown in Fig. 8. The results demonstrate that the ratio of SMA/CB has great effect on the electrical resistivity of PA6/PS/(SMA-CB) composites. Surprisingly the percolation phenomena are still observed at the ratios of SMA/CB between 85/15 and 80/20 even SMA-CB composites show good conductivity. Those percolation phenomena should be attributed to the different aggregation states of SMA molecular chains before or after being introduced into the PA6/PS blends. The introduction of SMA-CB composites to the PA6/PS blends should induce the SMA molecular chain from the curled state to uncurled state as a result from the compatibilization of SMA. The extension of SMA molecules at the interface should broaden the distance between CB particles due to the fact that SMA chains are grafted on the CB surface. So for the low

ratio of CB/(SMA-CB) the extension of SMA molecules will lead to the separation of CB particles, resulting in interruption of conductive network. On the contrary, for the high ratio of CB/(SMA-CB), the content of CB particles in SMA/CB composites is so high that the CB particles can still contact each other to form the conductive pathway even the SMA chains extend at the interface. Therefore the percolation phenomenon described above can be attributed to the percolation of CB in SMA-CB.

Influence of SMA-CB (70/30) composites loading on the room-temperature resistivity of PA6/PS (60/40) blends is studied, as shown in Fig. 9. The composites are observed to have a dramatic decrease in resistivity at a critical SMA-CB loading of 0.5 wt%. In order to confirm the percolation threshold of PA6/PS/(SMA-CB) composites, it should identify whether all CB particles are reacted with SMA or not. So the reactant of SMA and CB was still standed for 48 h to observe the deposition of CB. If some of CB particles are not reacted with SMA, they should deposit. The results showed that deposition was not observed. So it can be concluded that almost all CB particles can react with SMA and disperse at the interface. Considering that the mass ratio of SMA/CB is 70:30, it can be calculated that the percolation threshold of PA6/PS/(SMA-CB) composites is only 0.15 wt% of CB, which is much lower than that of PA6/PS/CB composites and is the lowest percolation threshold reported in the open literature. From Fig. 9 it can also be known that the percolation of PA6/PS/(SMA-CB) composites is governed by the content of SMA-CB and the percolation phenomenon appears at the loading of SMA-CB between 0.3 wt% and 0.5 wt%.

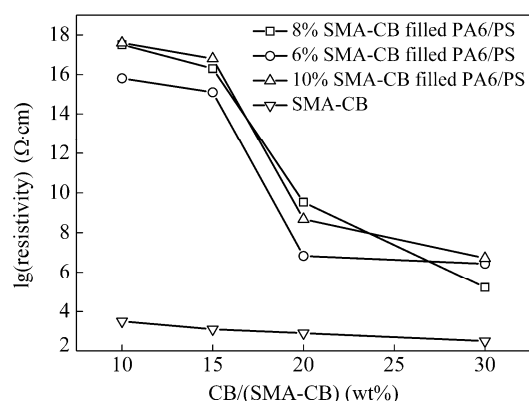


Fig. 8 Effects of loading SMA-CB on the room-temperature resistivity of PA6/PS and the resistivity of SMA-CB composites as a function of CB/(SMA-CB) mass ratios

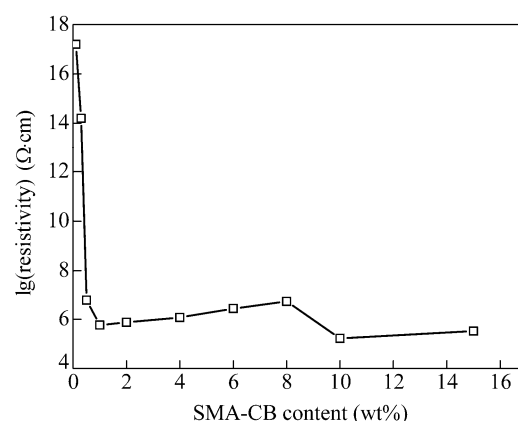


Fig. 9 Effect of SMA-CB content on the room-temperature resistivity of PA6/PS (60/40) composites

In order to explain the percolation governed by the content of SMA-CB, OM is employed to investigate the morphology of PA6/PS/(SMA-CB) composites, as shown in Fig. 10. From the OM images it can be observed that the morphologies of composites at the SMA-CB loadings of 0.3 wt% and 0.5 wt% are similar: the co-continuous phase is observed. Therefore it is confirmed that the percolation phenomenon has nothing to do with the morphology of composites. So the percolation phenomenon should be attributed to the continuity of SMA-CB at the interface. At the SMA-CB loading of 0.3%, the distribution of SMA-CB at the interface is incontinuous due to the very low loading, resulting in the incontinuity of conductive pathway. As the loading of SMA-CB is increased to 0.5%, SMA-CB can fill the interface to form a continuous conductive pathway.

From above investigated it can be concluded that the percolation of PA6/PS/(SMA-CB) composites is governed by the continuity of SMA/CB at the interface, the percolation of CB in SMA phase and the interface continuity of PS and PA6, which can be defined as triple percolation, a notable difference from double percolation and seldom reported in literature. The localization of CB and SMA at the interface is the main reason for the very low percolation threshold, as well as the percolation of CB in SMA-CB and the percolation of SMA-CB at the interface are also beneficial to the decrease of percolation threshold, namely the very low percolation threshold can be owed to the triple percolation arising from the special interface morphology.

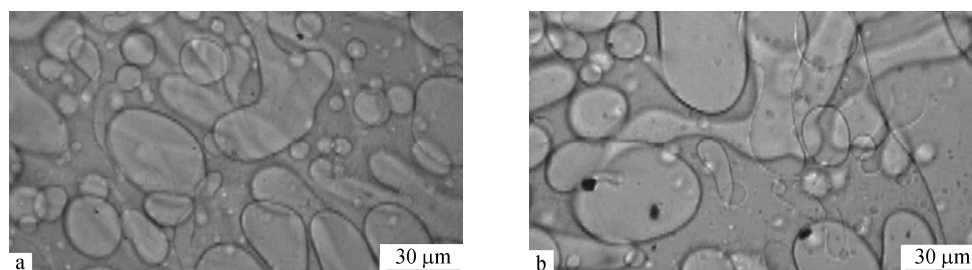


Fig. 10 OM photographs of SMA-CB filled PA6/PS (60/40) composites at different SMA-CB loading
a) 0.3 wt%; b) 0.5 wt%

PTC Effect of PA6/PS/(SMA-CB) Composites

In this paper, the temperature dependence of the resistivity of 6% SMA-CB (80/20) filled PA6/PS (60/40) and PA6/PS/CB (60/40/2) is also studied. From Fig. 11 it can be known that the PTC effect and NTC (negative temperature coefficient) effect of PA6/PS/(SMA-CB) are different from that of PA6/PS/CB. For PA6/PS/CB blend the resistivity shows a weak jump at about 170°C followed by the NTC effect. It was identified the localization of CB in PA phase. So the PTC effect of PA6/PS/CB blend can be attributed to a significant volume expansion of PA phase near its melting temperature (T_m), leading to the disconnection of CB in PA6. The appearance of NTC effect can arise from the CB reagglomeration to form the conductive pathways again. For the composites of PA6/PS/(SMA-CB), the PTC effect occurs at 90°C and its PTC intensity is higher than that of PA6/PS/CB. Moreover the volume resistivity shows a slow increase or decrease within the temperature range of 100°C and 175°C and the rapid decrease of resistivity occurs when the temperature is beyond 175°C, indicating the elimination of NTC effect between 100°C and 175°C.

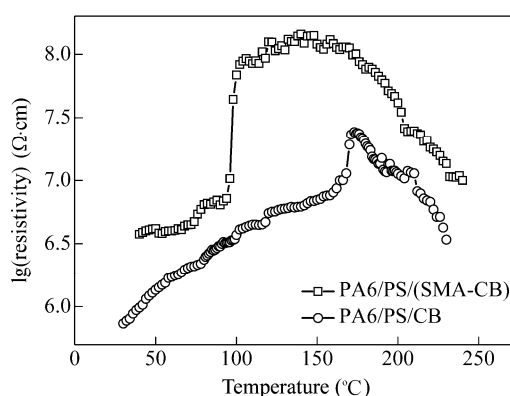


Fig. 11 Temperature-resistivity curves of 6% SMA-CB filled PA6/PS (60/40) and PA6/PS/CB (60/40/2)

The special PTC performance of PA6/PS/(SMA-CB) should be attributed to its interface morphology, *i.e.* SMA and the CB particles localize at interface to form the conductive pathways as well as SMA molecular chains are grafted onto the CB surface, causing that the interruption of conductive pathways should be influenced by volume expansion of PS, PA6 and SMA, according to the thermal expansion theory^[28]. Therefore the rapid increase in resistivity near 90°C, corresponding to the glass transition temperature (T_g) of PS phase, can be owed to the volume expansion of PS phase near its T_g , resulting in the interruption of conductive pathways at the interface.

It was well-known that the NTC effect is owed to the reagglomeration of CB particles^[29]. For the PA6/PS/(SMA-CB) composites the SMA molecular chains are grafted on the CB particles and the maleic anhydride in the SMA is reacted with amine group of PA6 to form the copolymer and dispersed at the interface. Therefore when the temperature is lower than the T_m of PA6, the CB particles are fixed by PA molecular chains, prohibiting the reagglomeration of CB particles at the interface, leading to the elimination of NTC effect

between 100°C and 175°C. When the temperature is over the T_m of PA6, the appearance of NTC effect arise from the melting of PA phase, resulting in the reagglomeration of CB particles.

CONCLUSIONS

The PA6/PS/(SMA-CB) composites were prepared by the method that SMA was first reacted with CB, and then blended with PA6/PS and their electrical properties were studied. In PA6/PS/CB blends, CB is selectively localized in the PA6 phase and the percolation threshold is 2 wt%, which is lower than that of PA6/CB due to the double percolation. As the introduction of SMA-CB composites to the PA6/PS blends, CB is induced to localize at interface of PA6/PS by SMA, resulting in the percolation threshold of only 0.15 wt% for PA6/PS/(SMA-CB) composites, which is much lower than that of PA6/PS/CB composites. The results show that the percolation is governed by the percolation of the CB in SMA phase, the continuity of SMA-CB at the interface and the continuity of interface between PS and PA6, which can be defined as triple percolation. Compared with PA6/PS/CB, the PTC intensity of PA6/PS/(SMA-CB) composites is higher and the NTC effect is eliminated within the temperature range of 100°C and 175°C. Its excellent electrical properties arise from the special interface morphology: SMA as well as the CB particles grafted on SMA molecular chains both localize at interphase between the PA6 and PS phases to form the conductive pathways.

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