

Composites Based on CB/CF/Ag Filled EPDM/NBR Rubber Blends with High Conductivity

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ABSTRACT: For many applications of conductive rubbers, it is desirable to endow the conductive rubber with high conductivity at low conductive filler loading. In this work, composites based on ethylene-propylene-diene monomer (EPDM) rubber and nitrile-butadiene rubber (NBR) were prepared using carbon blacks, carbon fibers, and silver powders as fillers. As the weight fraction of silver powder increased, the hardness of composites increased gradually while the tensile strength and elongation at break decreased. SEM revealed that the EPDM/NBR blends exhibited a relatively co-continuous morphology. The differential scanning calorimetry (DSC) curves reported the EPDM/NBR rubber blends were incompatibility. The thermogravimetry (TG) studies showed that adding a small amount of silver powder could improve the thermal stability of composites. These conductive composites exhibited good electrical property. At room temperature, when the total volume fraction of fillers was 15.20%, the volume resistivity of EPDM/NBR blend was only $0.0058 \Omega \text{ cm}$. © 2014 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* **2015**, *132*, 41357.

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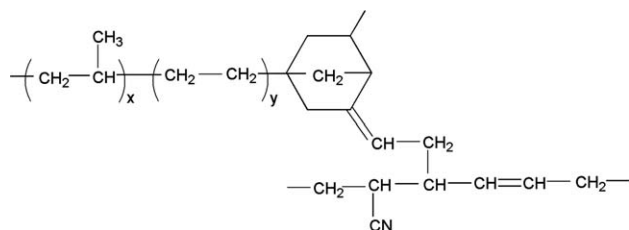
INTRODUCTION

Electrically conductive rubbers have attracted a great deal of scientific and commercial interest. The electrical insulating behavior of rubber materials is well known. The conductivity of rubber is imparted by incorporation of conductive fillers, which possess the ability to conduct electricity. According to the concentrations of fillers, conductive rubber can be formulated over a broad range of conductivity. These materials are distinguished by having good flexibility, processability, and their easily controlled volume resistivity. They are commonly used in electronic applications for thermistors, chemical sensors, and touch control switches.¹ Besides, these conductive composites have also been used as electromagnetic interference (EMI) shielding materials.² All kinds of rubbers have been used for preparation of these composites, such as natural rubber,³ styrene-butadiene rubber,^{4,5} ethylene-propylene-diene rubber,^{6,7} silicone rubber^{8–11}, nitrile rubber,¹² or rubber-rubber blends.^{13–15} The common fillers that used for conductive rubbers were carbon black,^{1,4,5,7} graphite,¹⁶ carbon fiber,^{2,17} carbon nanotube,¹⁸ pure metal,¹⁹ metal-coated inorganic particles,²⁰ and metal powders.^{21–23}

For the past few years, most research interest is focused on the problem of the relationship between the conductivity of the conductive rubber, the weight fraction of the conductive fillers

and percolation threshold in the study of conductive rubber. In reducing the amounts of conductive fillers to improve the mechanical properties of the conductive rubbers, how to maintain or even increase the conductivity of conductive rubbers and electrical stability at the same time is the key point. Many researchers have a number of outstanding works in this respect. Selective distribution of the conductive fillers in a continuous phase of a multiphase polymer, even polymer phase interface and using the conductive fillers having a large aspect ratio are two common methods. In many applications, high conductivity is the primary requirement and the mechanical properties may come to be the secondary consideration. To achieve a higher conductivity at low loading, the selective distribution of several different dimensions of conductive fillers in two different polar rubbers was used to construct the conductive network to achieve collaborative conductive effect.

The blending of immiscible polymers offers attractive opportunity for developing new materials with helpful combinations of properties. Rubber blends are frequently used in the rubber industry to obtain their well-balanced physical and mechanical properties, facile processability with relatively low production cost.²⁴ Blending of two or more types of polymers is a very valuable technique for the preparation and development of materials with properties superior to those of individual constituents.



Scheme 1. Schematic presentation the crosslinking of the polymer chains of EPDM and NBR rubbers.

It has been reported in the literature that rubber blends having differences in polarity are very useful in achieving a high degree of conductivity because of their well-defined interface.^{25,26} The selective localization of conductive fillers in multiphase polymeric materials is a favorable condition for obtaining heterogeneous microdispersion of conductive fillers. Two types of distribution of conductive fillers in the immiscible polymer blend were observed; first, conductive particles were distributed predominantly in one phase of the blend homogeneously. Second, the conductive particles concentrated at the interface of the two phases. The conductivity of these composites was determined by two factors: the preferential concentration of conductive particles in one phase, and the structural continuity of this phase.^{27,28} The distribution of conductive fillers at the interface of two rubbers gives rise to high conductivity. Accumulation of conductive fillers at the interface has the effect of increasing the number of contact points or decreasing the gap width. The heterogeneous microdispersion explains the higher conductivity of blends compared with the single polymer.²⁶

Ethylene-propylene-diene monomer (EPDM) and nitrile-butadiene rubber (NBR) have been chosen in this study because of their distinguished physical properties and their incompatibility. EPDM is a saturated, nonpolar rubber (very low $-C=C-$ content). NBR, however, contains polar CN groups, which improve the cohesive energy density between the chains. Such groups are absent in EPDM. The selective distribution of conductive fillers in the blend of such two polymers was used to prepare high conductive rubber. It is worth mentioning that both EPDM and NBR could be vulcanized by peroxide. Crosslinking between the EPDM and NBR rubber was possible during the co-crosslinking process (Scheme 1).

In recent years, electrically conductive rubbers have drawn considerable interest. Zou prepared the conductive silicone rubber composites filled with nickel-coated graphite (NCG). When the volume fraction of NCG was 33.1%, the volume resistivity of the composites was about $0.01 \Omega \text{ cm}$.²⁰ Mahmoud investigated conductive acrylonitrile butadiene rubber compounds filled with different concentrations of fast extrusion furnace black (FEF). The volume resistivity of composites was $\sim 10^2 \Omega \text{ cm}$ with 80% volume fraction of FEF.²⁹ This article is concerned with detailed studies on a 50/50 blend of nonpolar ethylene-propylene-diene monomer rubber (EPDM) and polar nitrile-butadiene rubber (NBR) that was prepared and loaded together with different amounts of carbon black particles, carbon fibers, and silver powders. Electrical properties have been studied in an attempt to understand the mechanism of conduction in such

systems. Some mechanical properties such as tensile strength, elongation at break, and hardness of the blends were also studied in order to evaluate their potential in industrial applications. When 30 phr CB, 10 phr CF together with 10 phr Ag were added into the rubber blend, high conductive rubber was prepared successfully with a electrical resistivity of $0.0058 \Omega \text{ cm}$. For this composite, the total volume fraction of fillers was 15.20%, lower than that reported in the literature.

EXPERIMENTAL

Materials

Liquid ethylene-propylene-diene monomer (Trilene 65, DCPD-EPDM) with 44.7% ethylene was supplied by Lion Copolymer Company. Liquid nitrile-butadiene rubber (NBR-40) with 30–34% acrylonitrile was provided by Lanzhou Petrochemical Company, China. Carbon black (HK-T120) and carbon fiber (Kajet CF) were obtained from Shanghai Kajet Chemical Technology Co., China. Silver powder (PA301) was obtained from Shenzhen Xinshengfeng Technology Co., China. Carbon black and carbon fiber were treated with silane coupling agent in order to improve their dispersity in the matrixes. Silver powder was treated with malonic acid before use to remove the organic lubricating layer that existed on the silver powder surface. The physical characteristics of carbon black, short carbon fiber, and silver powder was presented in Table I. Dicumyl peroxide (DCP, melting point $41\text{--}42^\circ\text{C}$, purity of 98%) was procured from Sinopharm Chemical Reagent Co., China. Triallyl isocyanurate (TAIC, melting point $26\text{--}28^\circ\text{C}$, purity of 98%) and poly(1,2-dihydro-2,2,4-trimethyl-quinoline) (antioxidant RD, melting point $72\text{--}94^\circ\text{C}$) were supplied by Jiangsu Huada Chemical Group Co., China.

Sample Preparation

Formulation of the composites was given in Table II. The compounds were prepared by solution blending. Styrene was used

Table I. Physical Characteristics of Conductive Carbon Black, Carbon Fiber, and Silver Powder

Carbon black	
DBP absorption number	310–345 mL/100 g
I absorption number	740–840 mg/g
Ash, weight	0.05%
PH value	8–10
Specific gravity	$1.78\text{--}1.82 \text{g/cm}^3$
Electrical resistivity	$\sim 0.8 \Omega \text{ cm}$
Carbon fiber	
Precursor	PAN
Average length	3 mm
Diameter	$7 \mu\text{m}$
Density	1.82g/cm^3
Electrical resistivity	$\leq 0.0014 \Omega \text{ cm}$
Silver powder	
Partical size	$2 \mu\text{m}$
Specific surface area	$0.4\text{--}0.6 \text{m}^2/\text{g}$
Ag/Cu	$\geq 99.9\%$

Table II. Formulation of the Composites Based on EPDM/NBR Rubber Blends

Sample	Compounds(phr ^a)						Ag
	EPDM/NBR	DCP ^b	TAIC ^b	RD ^c	CB	SCF	
1	50/50	4	8	1	30	10	0
2	50/50	4	8	1	30	10	2
3	50/50	4	8	1	30	10	4
4	50/50	4	8	1	30	10	6
5	50/50	4	8	1	30	10	8
6	50/50	4	8	1	30	10	10

^aPhr, parts per hundred.

^bCuring agent (Dicumyl peroxide -DCP; Triallyl isocyanurate-TAIC).

^cAntioxidant (Poly(1,2-dihydro-2,2,4-trimethyl-quinoline)).

as solvent to mix the fillers with the rubbers. After the homogenization of the rubber blend, the other additives were added. Mechanical stirring was also used for better particle dispersion. After 2 h of vigorous mixing, the solvent was evaporated in a vacuum oven at 80°C for 24 h. The compound blends were cured using Rubber Flat Vulcanizing Machine (QLB-D/Q, Nanjing, China) under a pressure of 15 MPa at 170°C to optimum cure times that had been previously determined. These cured sheets were conditioned before testing (24 h maturation at room temperature).

Characterization

Mechanical properties such as tensile strength and elongation at break were measured using a universal testing machine (Model CMT-5105) as per ASTM standard testing method with a cross-head speed of 50 mm/min at room temperature. For the tensile experiment, dumbbell samples were cut from 2 mm thick molded sheet. Five samples from each formulation were tested. The hardness of the composites was measured using a shore A durometer according to the standard ASTM D2240 testing method. For hardness measurement, the sheets having an effective thickness of 6 mm were used. Five measurements were recorded and the average value was reported. The samples of SEM analysis were immersed in liquid nitrogen for more than 10 min. to cool down and then fractured immediately. The SEM observation of fracture surface of the dried samples was made using a scanning electron microscope (Model Hitachi S-4800). Differential scanning calorimetry (DSC) was carried out

with a Netzsch DSC 200F3 calorimeter. The predried samples ranging from 2 to 5 mg were hermetically sealed in an aluminum pans. DSC analysis of the samples was performed under a liquid nitrogen atmosphere using a flow of 50 cm³/min. in temperatures ranging from -100°C to 100°C at a heating rate of 10°C/min. The thermal stability of samples was determined by nonisothermal thermogravimetry (TG) using a NETZSCH STA409 PC instrument. The measurements were conducted at a heating rates of 10°C/min in a nitrogen atmosphere (flow rate was 25 cm³/min.). The examined samples weight about 15 mg. Samples were heated from the room temperature to 700°C. The volume resistivity for all composites was measured by four-point probe method so that it was free from the resistance of the sample/electrode contacts. The volume resistivity data reported here was the average of three samples.

RESULTS AND DISCUSSIONS

Mechanical Properties of Composites

There was a balance problem between mechanical properties and electrical properties of high conductive rubber. The effect of filler content on the performance of high conductive rubber was shown in Table III. It can be seen from the table that conductive filler contents had great effect on the mechanical properties of conductive rubber. In the experimental range, the hardness of composites increased gradually while the tensile strength and elongation at break decreased as the weight fraction of silver powder increased. The bonding force between silver powder and rubber matrix deteriorated, rather than reinforcing the rubber matrix but making the mechanical properties of conductive rubber worse. As the weight fraction of silver powder increased, the average distance between silver powders that were in the dispersed state was shorter and shorter and the conductive channels formed by silver particles contacting each other increased gradually, so that the rubber matrix coated silver powder was markedly thinned. Therefore, the compound was more easily destroyed resulting decrements in the tensile strength and elongation at break of composites. At the same time, with the increase of silver powder content, the motion of molecular chains was restricted due to the interaction between conductive fillers and rubber molecular chains, which also led to the decrease in elongation at break. For the composites loaded together with 30 phr CB, 10 phr CF, and 10 phr silver powder, the electrical resistivity of this blend was 0.0058 Ω cm, but the Shore A hardness was up to 78. The tensile strength and elongation at break were 1.89% and 120%, respectively.

Table III. Effect of Conductive Filler Content on the Properties of Rubber Blends

Sample	Properties			
	Tensile strength (MPa)	Elongation at break (%)	Shore A hardness (HA)	Electrical resistivity (Ω·cm)
1	2.56 ± 0.03	260 ± 4	65 ± 2	0.28
2	2.42 ± 0.02	220 ± 6	67 ± 2	0.19
3	2.29 ± 0.05	190 ± 3	70 ± 3	0.12
4	2.06 ± 0.03	173 ± 5	72 ± 1	0.0552
5	1.94 ± 0.03	165 ± 3	76 ± 2	0.0152
6	1.89 ± 0.02	120 ± 3	78 ± 1	0.0058

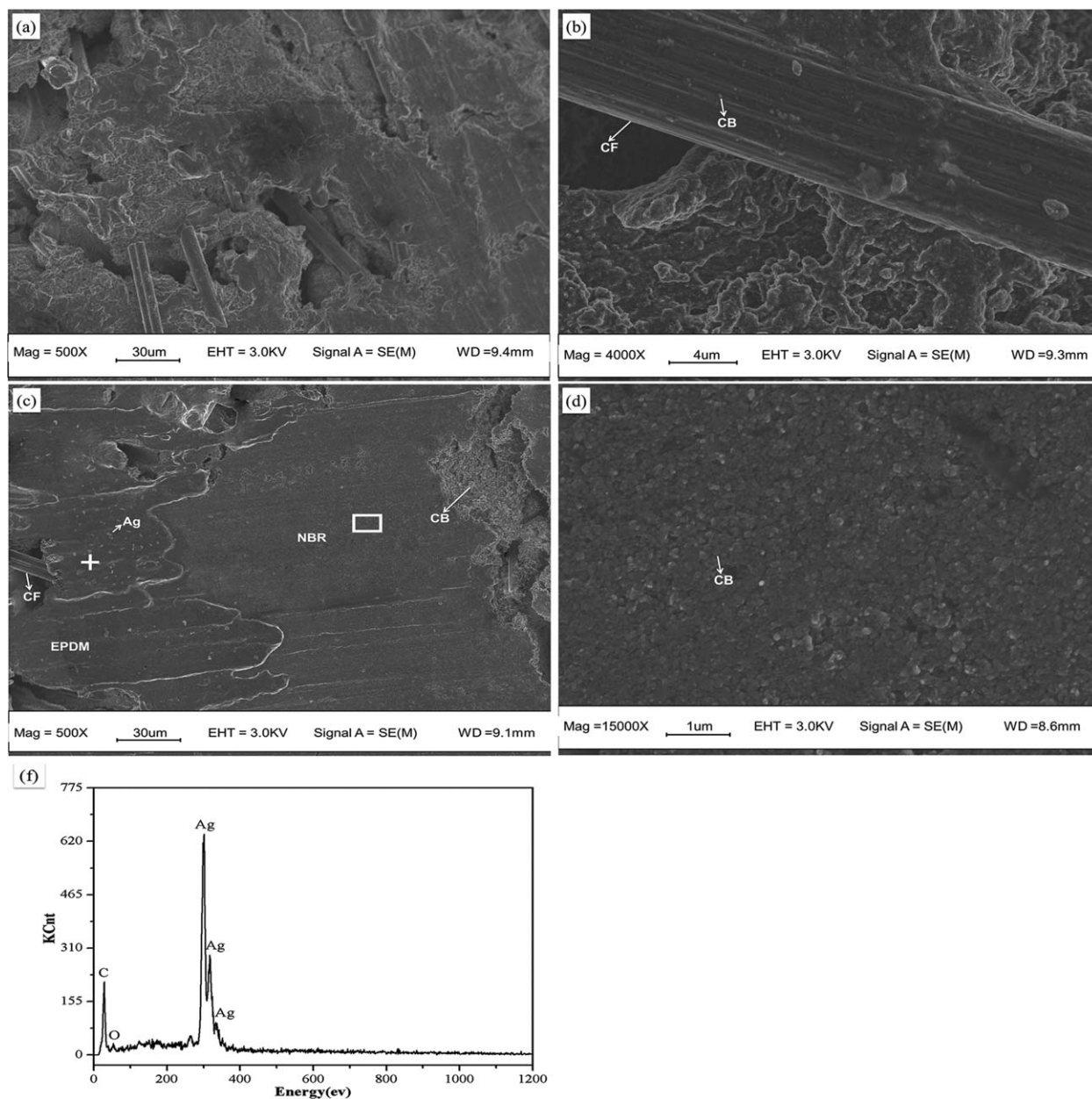


Figure 1. SEM micrographs and EDS analysis of the fractured surface of the EPDM/NBR rubber blends with different content of conductive fillers: (a) 30 phr CB + 10 phr CF; (b) (c) (d) (e) (f) 30 phr CB + 10 phr CF + 10 phr Ag.

Despite increasing the weight fraction of conductive fillers can reduce the volume electrical resistivity of composites, the processability, and mechanical properties of the composites get worse and the costs increase. Therefore, on the premise of meeting the electrical properties, conductive filler contents should be limited as low as possible.

SEM Analysis of Composites

Morphology is a major factor of rubber blends. It is well known that the phase structure of the blend is influenced by several factors, including the blend ratio, viscosity of each component, surface characteristics, and compounding process. The primary factor that determines the final morphology of the blends is their composition. NBR has strong molecular polarity and

surface tension higher than that of EPDM, which results in their incompatibility. SEM was employed to study the fractured composites based on the EPDM/NBR rubber blends and confirmed that certain features of the fracture can be correlated with the electrical properties.

There are lots of functional groups such as hydroxyl, carboxyl, pyrone, lactone, quinone, ketone, and phenol on the carbon black surface. The fact that the carboxylic, phenolic, lactonic on the surface of CB is a factor affecting the selective distribution of CB particles.³⁰ If the interactions between CB particles and one polar polymer component are strong enough, they will influence the dispersion of CB particles. As for CB/EPDM/NBR composite, NBR had a very high surface tension and polarity,

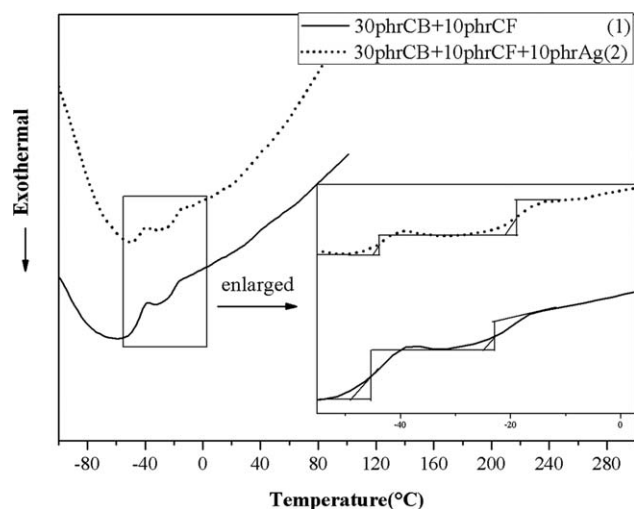


Figure 2. DSC thermograms of the rubber blend loaded with CB/CF and CB/CF/Ag.

whereas the EPDM was a nonpolar polymer. Thus, the interaction between CB particles and NBR phase was stronger than that between CB particles and EPDM phase. In other words, CB particles had a priority to locate in the NBR phase.

On the contrary, Chan and coworkers found that CB particles prefer to selectively locate in the lower viscosity phase in a multiphase blend.³¹ For the rubber blend, the viscosity of NBR and EPDM are 50 and 1900 Pa s, respectively. Therefore, CB particles have stronger tendency to locate in NBR phase than EPDM phase.

Figure 1(a–f) presented SEM micrographs and EDS analysis of the EPDM/NBR = 50/50 rubber composites. As shown in figures, CB particles were selectively distributed in the NBR phase, whereas the Ag particles were mainly located in the EPDM phase and CFs were uniformly distributed in the matrix and through multiple phase zone, which played a collaborative conductive effect. In our experiments, the ratio of EPDM and NBR was 50/50. At this point, both EPDM and NBR formed continuous phases in three-dimensional space; Such structures are known as co-continuous phase structures [see Figure 1(a,c)]. Because of the uneven dispersion of CB, more carbon black dispersed in NBR phase thereby increasing the concentration of CB in NBR under the condition of the overall content of CB unchanged and decreased the electrical resistivity of conductive rubber. From Figure 1(f), the EDS analysis showed that the phase that the Ag particles located was EPDM phase, because nitrogen was not found, which indicated that this phase did not contain -CN groups. So this phase was EPDM phase and another phase was NBR phase. When the selected region in Figure 1(c) was amplified to specify multiple, it got Figure 1(d). It could be seen from Figure 1(d) that CB particles were mainly distributed in this phase. It also proved that this phase was NBR phase. Ag particles had excellent conductivity. Adding a small amount of silver powder simultaneously in the rubber blend can largely reduce the electrical resistivity of the composites. CFs had large dimensions and high aspect ratios which belonged to the two-dimensional conductive filler. For the

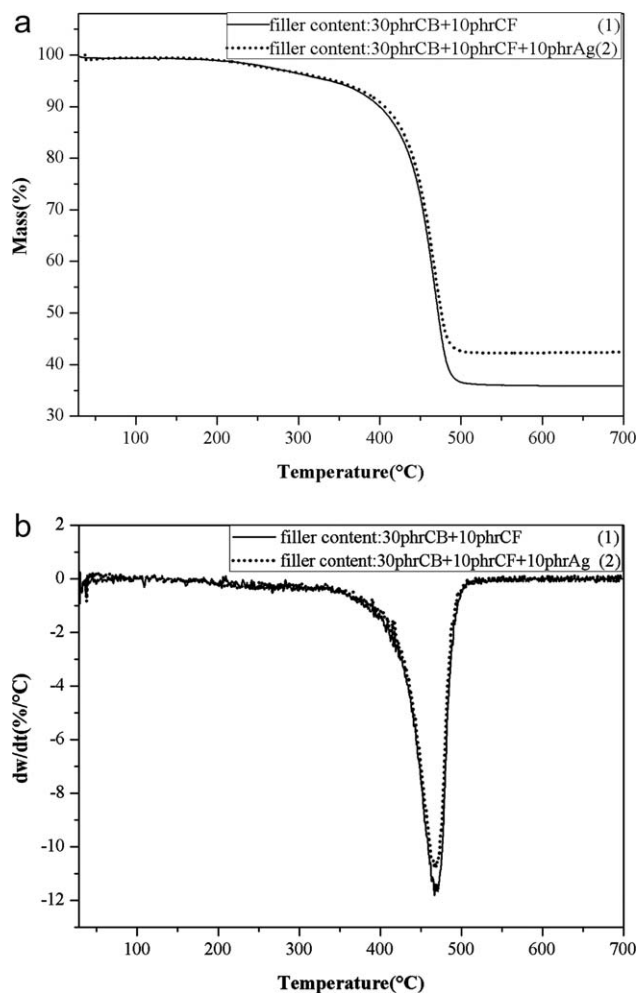


Figure 3. a: TG curves of the CB/CF/EPDM/NBR composites and the CB/CF/Ag/EPDM/NBR composites. b: DTG curves of the CB/CF/EPDM/NBR composites and the CB/CF/Ag/EPDM/NBR composites.

rubber blend, CFs were distributed throughout the entire matrix and might pass through several nonadjacent phases. They transported electrons over long distances, acting as bridges between conducting phase regions. The combined effect of CB, CF, and Ag assisted the formation of three-dimensional conductive networks, thereby improving the conductivity of composites. It could be seen from the Figure 1(b) that many CB particles aggregated on the surface of CF, which promoted the formation of conductive path. The collaborative conductive effect of conductive fillers can effectively reduce the electrical resistivity of rubber blend. Two phases with irregular shape can be observed in Figure 1(c). This meant that EPDM/NBR rubber blends were incompatible. EPDM was a nonpolar rubber, whereas NBR containing polar -CN groups was a polar rubber. The incompatibility of the two kinds of rubber made composites existing obvious interface. The interphase adhesion between EPDM and NBR was very weak.

DSC Studies of Composites

The DSC thermogram of the rubber blend loaded with CB/CF (30/10) and CB/CF/Ag (30/10/10) was given in Figure 2. Two separate T_g values of the EPDM/NBR rubber blend can be

Table IV. TGA Data of EPDM/NBR Blends with Different Content of Conductive Filler

Sample	$T_{d, 5\%}$ (°C)	$T_{d, 10\%}$ (°C)	T_{max} (°C)	W (700°C/%) ^a
CB/CF/EPDM/NBR	339	400	466	35.87
CB/CF/Ag/EPDM/NBR	345	406	468	44.24

^a W 700°C/% was the char residue of composites at 700°C.

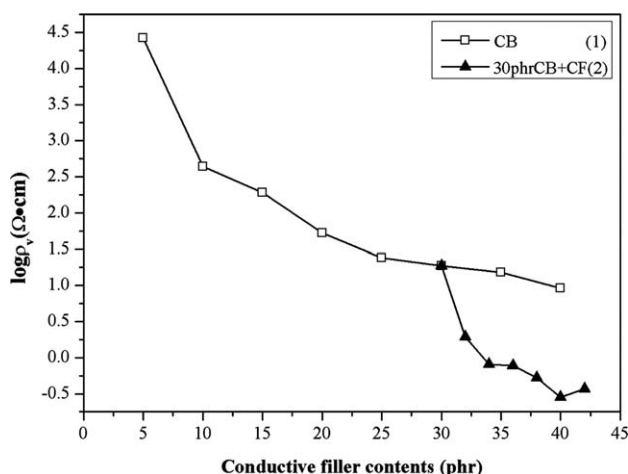
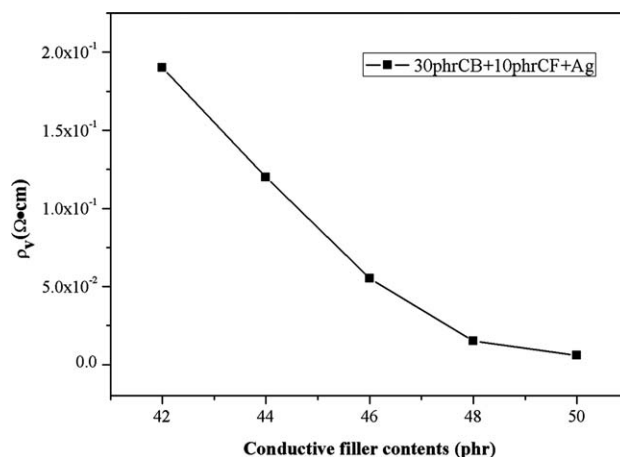
observed. For the CB/CF/EPDM/NBR composites, the T_{g1} was found to be -48.7°C , which resulted from the EPDM rubber and the T_{g2} was -23.3°C which was produced by the NBR rubber. For the CB/CF/Ag/EPDM/NBR composites, the T_{g1} was -46.1°C and the T_{g2} was -20.7°C . The T_g of the blends was seldom changed with the addition of Ag. The addition of Ag restricted the mobility of the polymer chain through crosslinking and polymer-filler networking. Therefore, as indicated by the DSC curve of the EPDM/NBR rubber blend, the EPDM/NBR polymer blends were incompatible.

TG Studies of Composites

Thermal stability of a material is stability against degradation on exposure to elevated temperatures in an inert environment. Rubbers are often exposed to high temperatures during processing or use. Therefore, thermal stability is among the most important properties of rubbers for a wide range of applications. The thermal stability of rubber materials is usually defined as the beginning temperature of decomposition of the sample at a programmed heating rate.³²

The TG and DTG curves of the CB/CF/EPDM/NBR composites and the CB/CF/Ag/EPDM/NBR composites were shown in Figure 3(a,b). The relative thermal stability of the samples was evaluated by the $T_{d,5\%}$, $T_{d,10\%}$, the temperature of maximum rate of weight loss (T_{max}) and the percent char residue at 700°C. These data were listed in Table IV.

It was clearly seen that the decomposition temperature at 5% weight loss ($T_{d,5\%}$) of CB/CF/EPDM/NBR composites was 339°C. Its TGA trace showed only one weight-loss stage. The

**Figure 4.** Volume resistivity against filler loading for EPDM/NBR rubber blend loaded together with CB and CF.**Figure 5.** Volume resistivity against filler loading for EPDM/NBR rubber blend loaded together with CB, CF, and Ag.

stage was in the temperature range of 310–500°C corresponding to a single DTG peak at 466°C (T_{max}) and the weight loss was about 64.14%. When adding a small amount of Ag in the CB/CF/EPDM/NBR composites, the decomposition temperature at 5% weight loss ($T_{d,5\%}$), the decomposition temperature at 10% weight loss ($T_{d,10\%}$) and the temperature of maximum rate of weight loss (T_{max}) rose slightly. This indicated that adding a small amount of Ag had favorable influence on the thermal stability of rubber blends. It could be that Ag itself had a good heat-resistant stability.

Effect of Conductive Filler Loading on Conductivity

The electrical resistivity of CB/CF/EPDM/NBR composites and CB/CF/Ag/EPDM/NBR composites were represented in Figures 4 and 5. It was observed that the electrical resistivity decreased with conductive filler loading. In our early studies, we prepared the conductive rubber whose electrical resistivity was 0.28 Ω cm when 30 phr CB together with 10 phr CF were used to fill the EPDM/NBR (50/50) rubber blend (Figure 4). In order to further reduce the electrical resistivity of composites, a small amount of silver powder was added into the composites. When 30 phr CB, 10 phr CF together with 10 phr Ag were added into EPDM/NBR rubber blend, high conductive rubber was prepared successfully. For this composite, the total volume fraction of fillers was 15.20%, and the electrical resistivity was 0.0058 Ω cm (Figure 5). When 30 phr CB was added into rubber blend, the continuous conductive networks had already been formed. However, the change in conductivity with the carbon black loading was only marginal; hence, this loading would be the optimum choice for getting good conductivity from these composites. At this time, the conductivity of the composites mainly depended on the conductivity of filler particles rather than on their ability to form a continuous conductive network through aggregation. Conductive carbon fiber had good electrical conductivity, large dimensions and high aspect ratios. Using carbon black and carbon fiber simultaneously in the rubber blend, the electrical resistivity of composites can be reduced to some extent. As described earlier, when the CB content reached 30 phr, the CB particles formed continuous conductive networks. The addition of CF increased the conductivity of the

composites. The CFs transported electrons over long distances with few barriers, whereas CB particles provided charge transport over relatively short distances and produced improved interfiber contact as a result of bridge formation.³³ Additionally, CFs were uniformly distributed in the matrix and through multiple phase zone which made CF function as bridges between the nonadjacent conductive phases. On the basis of the aforementioned details, a small amount of silver powder was added into the rubber blend in order to prepare high conductive rubber. Silver powders with laminate structure used in our studies which belong to the two-dimensional conductive filler had excellent conductivity. Adding a small amount of silver powders simultaneously in the rubber blend can largely reduce the electrical resistivity of the composites. In the rubber blend, the carbon fibers also functioned as bridges between CB particles or silver powders. The conductive paths were increased and eventually built three-dimensional conductive networks. The combined effects of CB, CF, and Ag can effectively reduce the electrical resistivity so as to improve the conductivity of composites.

CONCLUSIONS

In this study, the system of EPDM/NBR blends at a 50/50 ratio with different content of conductive fillers was investigated. For all EPDM/NBR rubber blends, the hardness of composites increased while the tensile strength and elongation at break decreased as the weight fraction of silver powder increased. DSC analysis concluded that the EPDM/NBR rubber blends were incompatible. SEM results showed that CB particles were selectively distributed in the NBR phase, whereas the Ag particles were mainly located in the EPDM phase and CFs were uniformly distributed in the matrix and through multiple phase regions, which played a collaborative conductive effect. In addition, the TG studies showed that adding a small amount of silver powder can improve the thermal stability of composites. When 30 phr CB, 10 phr CF together with 10 phr Ag (the total volume fraction of fillers was 15.20%) were added into EPDM/NBR rubber blend, high conductive rubber was prepared successfully whose electrical resistivity was 0.0058 Ω cm.

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REFERENCES

1. Tanrattanakul, V.; Bunchuay, A. *J. Appl. Polym. Sci.* **2007**, *105*, 2036.
2. Das, N. C.; Khastgir, D.; Chaki, T. K.; Chakraborty, A. *Compos. Part A: Appl. S.* **2000**, *31*, 1069.
3. Bokobza, L. *Express Polym. Lett.* **2012**, *6*, 213.
4. Mohanraj, G. T.; Chaki, T. K.; Chakraborty, A.; Khastgir, D. *Polym. Eng. Sci.* **2006**, *46*, 1342.
5. Mohanraj, G. T.; Chaki, T. K.; Chakraborty, A.; Khastgir, D. *J. Appl. Polym. Sci.* **2004**, *92*, 2179.
6. Ghosh, P.; Chakrabarti, A. *Eur. Polym. J.* **2000**, *36*, 1043.
7. Soto-Oviedo, M. A.; Araújo, O. A.; Faez, R.; Rezende, M. C.; De Paoli, M. A. *Synth. Met.* **2006**, *156*, 1249.
8. Petkovic, D.; Issa, M.; Pavlovic, N. D.; Zentner, L. *Sensor Rev.* **2013**, *33*, 114.
9. Wang, L. H.; Ding, T. H.; Wang, P. *Sens. Actuators A: Phys.* **2007**, *135*, 587.
10. Duan, Y. P.; Liu, S. H.; Guan, H. T. *Sci. Technol. Adv. Mater.* **2005**, *6*, 513.
11. Wang, X.; Xia, Z.; Yuan, B.; Zhou, H.; Li, Z.; Chen, N. N. *Mater. Des.* **2013**, *51*, 287.
12. Ma, R.; Kwon, S.; Zheng, Q.; Kwon, H. Y.; Kim, J. I.; Choi, H. R.; Baik, S. *Adv. Mater.* **2012**, *24*, 3344.
13. Das, N. C.; Chaki, T. K.; Khastgir, D. *Carbon* **2002**, *40*, 807.
14. Kueseng, P.; Sae-oui, P.; Sirisinha, C.; Jacob, K. I.; Rattanasom, N. *Polym. Test.* **2013**, *32*, 1229.
15. Kueseng, P.; Sae-oui, P.; Sirisinha, C.; Rattanasom, N. *Polym. Test.* **2013**, *32*, 731.
16. Mansour, S. A.; El-Salam, M. A.; Moharram, A. H.; Hussein, M.; Al-Agel, F. A. *J. Appl. Polym. Sci.* **2012**, *126*, 593.
17. Shen, L.; Wang, F. Q.; Yang, H.; Meng, Q. R. *Polym. Test.* **2011**, *30*, 442.
18. Kim, Y. A.; Hayashi, T.; Endo, M.; Gotoh, Y.; Wada, N.; Seiyama, J. *Scr. Mater.* **2006**, *54*, 31.
19. Lee, H. H.; Chou, K. S.; Shih, Z. W. *Int. J. Adhes. Adhes.* **2005**, *25*, 437.
20. Zou, H.; Zhang, L. Q.; Tian, M.; Wu, S. Z.; Zhao, S. H. *J. Appl. Polym. Sci.* **2010**, *115*, 2710.
21. Jang, J.; Ryu, S. K. *J. Mater. Process. Technol.* **2006**, *180*, 66.
22. Novák, I.; Krupa, I.; Chodák, I. *Synth. Met.* **2004**, *144*, 13.
23. Liang, T. X.; Guo, W. L.; Yan, Y. H.; Tang, C. H. *Int. J. Adhes. Adhes.* **2008**, *28*, 55.
24. Findik, F.; Yilmaz, R.; Köksal, T. *Mater. Des.* **2004**, *25*, 269.
25. Sircar, A. K.; Lamond, T. G. *Rubber Chem. Technol.* **1973**, *46*, 178.
26. Marsh, P. A.; Voet, A.; Mullens, T. J.; Price, L. D. *Rubber Chem. Technol.* **1970**, *43*, 470.
27. Tchoudakov, R.; Breuer, O.; Narkis, M.; Siegmann, A. *Polym. Eng. Sci.* **1996**, *36*, 1336.
28. Breuer, O.; Tchoudakov, R.; Narkis, M.; Siegmann, A. *J. Appl. Polym. Sci.* **1997**, *64*, 1097.
29. Mahmoud, W. E.; El-Lawindy, A. M. Y.; El Eraki, M. H.; Hassan, H. H. *Sens. Actuators A: Phys.* **2007**, *136*, 229.
30. Huang, J. C. *Adv. Polym. Tech.* **2002**, *21*, 299.
31. Feng, J.; Chan, C. M.; Li, J. X. *Polym. Eng. Sci.* **2003**, *43*, 1058.
32. Ajloo, D.; Sharifian, A.; Behniafar, H. *Bull. Korean Chem. Soc.* **2008**, *29*, 2009.
33. Shen, L.; Xu, J. W.; Yi, X. S. *Fuhe Cailiao Xuebao/Acta Mater.* **2001**, *18*, 18.