Effect of Some Service Conditions on the Electrical Resistivity of Conductive Styrene–Butadiene Rubber–Carbon Black Composites

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ABSTRACT: Conductive polymer composites were prepared using vulcanized styrene–butadiene rubber as a matrix and conductive carbon black as a filler. The filler loading was varied from 10 to 60 phr. The volume resistivity was measured against the loading of the carbon black to verify the percolation limit. The electrical conductivity of filled polymer composites is attributed to the formation of some continuous conductive networks in the polymer matrix. These conductive networks involve specific arrangements of conductive elements (carbon black aggregates) so that the electrical paths are formed for free movement of electrons. The effects of temperature and pressure on the volume resistivity of the composites were studied. The volume resistivity of all the composites increased with increase in temperature, and the rate of increase in the resistivity against temperature depended on the loading of carbon black. The change in volume resistivity during the heating and cooling cycle did not follow the same route, leading to the phenomena of electrical hysteresis and electrical set. It was found that the composites with 40 and 60 phr carbon black become more conductive after undergoing the heat treatment. Generally, all the composites showed a positive temperature coefficient of resistivity. The volume resistivity of all the composites decreased with increase in pressure. The relaxation characteristic of the volume resistivity of the composites was studied with respect to time under a constant load. It was found that the volume resistivity of the compressed specimen of the composites decreased exponentially with time. It was observed that initially a faster relaxation process and later a slower relaxation process occurred in these composites. Some mechanical properties of these composites were also measured to confirm the efficacy of these composites for practical applications. © 2004 Wiley Periodicals, Inc. J Appl Polym Sci 92: 2179–2188, 2004

Key words: conducting composites; conductivity; mechanical properties; relaxation; resistivity

INTRODUCTION

Carbon black and carbon fiber–filled conductive polymeric composites have found use in numerous high-technology applications in aerospace industries and in modern electrical components and devices.[1–10] They consist of a polymer matrix and fine conductive elements like steel, carbon black, aluminum fiber, and carbon fiber as conductive filler. The main problem associated with the production of a conductive polymer composite is obtaining reproducible conductivity, given that electrical conductivity and mechanical properties are strongly affected by the type of conductive additive, its state of dispersion, and various processing and service conditions. During processing and service conditions, objects made from polymers are simultaneously subjected to mainly mechanical and thermal stresses, occasionally radiation, and other types of influences. Some

investigators[11–15] have reported the effect of mechano-chemical processes on electrical resistivity during manufacturing of polymers into objects. Extensive research has also been done to elucidate the effect of processing parameters on the electrical properties of polymers incorporated with conductive fillers.[16–18] The effect of temperature on the resistivity of the polymer composites has also been reported in the literature.[19–24] The present work reports the findings of an experimental investigation on the change of electrical resistivity of styrene–butadiene rubber (SBR) filled with conductive carbon black when subjected to variations of temperature and pressure. The effect of temperature, pressure, and constant compressive stress over a time period on the electrical resistivity of conductive carbon black–filled SBR composites constituted the focus of this investigation. The mechanical properties of these composites are also reported to confirm their suitability for practical applications.

EXPERIMENTAL

Materials

The polymer, styrene–butadiene rubber (SBR-1502, styrene content 23.5%, ML_{1+4} 100°C, 51), was supplied

TABLE I
Physical Characteristics of Vulcan XC-72 Carbon Black

Property	Value
Nitrogen surface area (m/g)	180
DBP[a] absorption number (mL/100 g)	178
Particle diameter (nm)	29
Electron microscopic surface area (m/g)	86
CTAB[b] surface area (m/g)	86

[a] Dibutyl phthalate.
[b] Cetyl trimethyl ammonium bromide.

by Synthetic and Chemicals Ltd. (Barielley, India). The filler, conductive carbon black (CCB; Vulcan XC-72), was procured from Cabot India Ltd. The physical characteristics of the carbon black are presented in Table I. Zinc oxide was procured from Merck Ltd., Mumbai, India. Antioxidant TQ was procured from Bayer India Ltd., Mumbai, India. MBTS was procured from ICI India Ltd. Sulfur was procured from s.d.fine-CHEM Ltd., Mumbai, India. Other compounding ingredients were of chemically pure grade and procured from standard suppliers.

Sample preparation

The materials used and compound formulations are given in Table II. The rubber and the various ingredients were mixed in a two-roll mill. The composites were cured at 150°C in an electrically heated press to the optimum cure time, as obtained from a Monsanto R100 rheometer (Monsanto, St. Louis, MO). These vulcanized sheets were allowed to mature at room temperature for 24 h before testing.

Sample testing

Measurement of effect of temperature on volume resistivity

The volume resistivity of the composites in the insulating range ($\rho \geq 10^8$ ohm cm) was measured using an HP 16008A resistivity cell (Hewlett–Packard, Palo Alto, CA) coupled with an HP 4329A resistance meter. The volume resistivity of the composites in the conducting range ($\rho \leq 10^4$ ohm cm) was measured using a four-probe technique.[25] The instruments used for this purpose were a programmable dc voltage/current generator (Advantest TR 6142) and a Digital Multimeter (Schlumberger, UK 7071).

The effect of temperature on the SBR–CCB composites was studied only for the samples that fell in the conducting range with the use of the four-probe technique and instruments stated earlier. The volume resistivity was measured in the temperature range of 25 to 125°C. To estimate the volume resistivity at these elevated temperatures the entire electrode system was placed in an oven where the temperature could be monitored and controlled over the range 20–125°C. The heating and cooling were done in a discrete manner; that is, the sample was allowed to reach a particular temperature and kept at that temperature for some time and, after steady state was achieved, the measurement was carried out. The temperature effect was studied in both heating and cooling modes in two cycles.

Measurement of effect of pressure on volume resistivity

To study the effect of hydrostatic pressure on the volume resistivity, a two-probe technique was used. A cylindrical molded sample (diameter 30 mm; height 14 mm) was placed between two circular electrodes of a typical cell (homemade) where variable pressure could be applied to the sample. The method was based on application of current and voltage measurements. The cell was connected with the Digital Multimeter to give the resistance values directly. The measurements were made after 8 min of the application of each individual pressure.

Measurement of effect of pressure and time on volume resistivity

To study the effect of constant compressive stress (pressure) over a time period on the resistivity of the samples, the same set space mentioned above was used. A constant pressure of 6940.3 Pa was applied on all the samples and the variations in resistivity over a period of 10,800 s (3 h) were recorded.

Measurement of mechanical performance

Mechanical properties, such as tensile strength, modulus, and elongation at break, of different samples were measured using a Zwick universal testing machine (UTM) Model 1445 (Germany).

In the text these composites are identified by an alphanumeric system. The first three block letters rep-

TABLE II
Formulation Used for the Preparation of the Composites

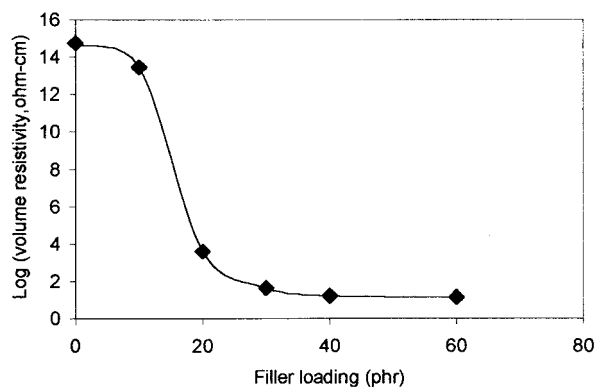| Ingredient	Loading (phr)
SBR 1502	100.0
Zinc oxide	5.0
Stearic acid	1.5
Antioxidant TQ	1.0
Vulcan XC-72	10–60
Process oil	1–7
MBTS	1.0
TMT	0.2
Sulfur	2.0

Figure 1 Effect of filler loading on the electrical resistivity of the SBR–CCB system.

resent the rubber used. The two numbers after these letters represent the loading of the carbon black. The next three block letters represents the conductive carbon black.

RESULTS AND DISCUSSION

Effect of filler loading

The effect of the conductive carbon black (CCB) loading on the volume resistivity of the SBR–CCB composites was investigated and the results are shown in Figure 1. It was found that the resistivity of the composites is almost constant, initially, and then a sharp decrease occurred almost at a particular loading; thereafter, the resistivity again remained almost constant with further addition of carbon black. The loading after which there is no significant change in the resistivity, irrespective of further addition of the filler, is called the percolation limit of the composite system. For the present system, the limit occurred around 30 phr of CCB. Before the percolation limit conductive particles are more isolated from each other, although at the percolation limit a continuous network is formed through the aggregation of conductive particles.

Effect of temperature

The dependency of electrical resistivity on temperature for conductive polymer composites is quite a complex phenomenon. The temperature coefficient of resistance may be positive (PCT), negative (NCT), or zero, depending on the concentration of filler and the nature of the polymers and the filler.[26] The variation of volume resistivity with temperature for the present system, carbon black–filled SBR composites, is shown in Figure 2. The resistivity of carbon black–filled composite progressively increases with the increase in temperature [i.e., a positive coefficient of temperature

(PCT effect) was observed]. This behavior can be explained based on three main theories of conduction as follows: (1) conductive network rearrangement, (2) tunneling effect, and (3) electric field radiation.[27,28] In fact, the actual conduction seems to be the net result of the combined effect of different mechanisms described in these theories. The appreciable PCT effect can be explained by the predominant breakdown of the conducting network structure attributed to differential thermal expansion of rubber matrix compared to the filler.[29] Because of an increased gap between conducting elements with the increase in temperature the resistivity increases; also the probability of electron tunneling and electric field radiation is reduced in this condition.

From Figure 2 it also can be seen that the volume resistivity of all the composites linearly varies on an average with temperature. The slopes of all the curves are almost constant and similar to each other except that, for the 30CCB samples, the curve is steeper between about 25 and 50°C. The rapid change in resistivity here is presumably because it is just at the percolation limit. Even small changes in the network structure would have the greatest effect on resistivity in the percolation region. All five plots corresponding to the respective filler loadings were fitted into linear equations that describe their variance with temperature. The concentrations investigated are slightly less than the exact percolation limit as well as above the percolation limit. The equations of best fit are shown in Table III. It can be inferred that the general behavior of the composites with temperature can be expressed as

$$\log \rho = AT + B \qquad (1)$$

where ρ is the volume resistivity (in ohm cm), A and B are numerical parameters, and T is the temperature (in °C).
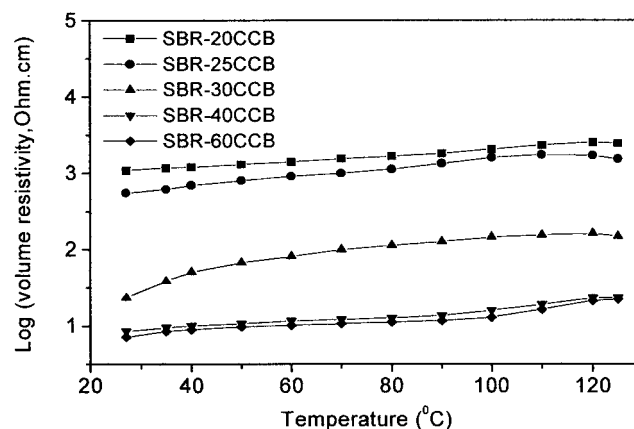

Figure 2 Effect of temperature on the volume resistivity of the composite system based on SBR–CCB.

Filler loading (phr)	Equation of best fit
20	$\log \rho = 0.0038T + 2.9282$
25	$\log \rho = 0.0051T + 2.6374$
30	$\log \rho = 0.0074T + 1.3798$
40	$\log \rho = 0.0043T + 0.8109$
60	$\log \rho = 0.0042T + 0.7491$

Equation (1) gives rise to two numerical parameters, A and B. After a close observation of Table III, it can be seen that at any given measurement temperature the parameters A and B are dependent on the filler loading. It can be noted that the parameter B decreases continuously with increase in filler loading near the percolation limit. Hence, the parameter B can be correlated to the interfiller aggregate distance in the composites, which continuously decreases with increase in filler loading. It can be seen that the parameter A passes through a maximum at the percolation limit. This is because, at the percolation limit, some continuous conductive network is established in the insulating matrix and the system changes from insulating to conducting, as if a conductive wire was running through the insulating matrix. After that, however, further addition of conductive particles simply establishes a few more conductive networks; this situation may be correlated to the increase in diameter of conductive wire inserted in the insulating matrix, so the effect will be only marginal and the rate of change simply decreases.

The effect of repeated heating and cooling on the resistivity of the composites was also investigated. Figures 3 and Figure 4 depict the above discussed behaviors of the composites. It was found that the behavior of the composites, although similar, were composition dependent. The composites with 20, 25, and 30 phr CCB were unable to regain the original values of resistivities that they had at individual temperatures during the heating part of the cycle when subjected to cooling. It is observed that the resistivity values obtained during the cooling part of the cycle at any particular temperature are always higher than those obtained during the heating cycle. This is attributed to the twin phenomena of electrical hysteresis and electrical set that the composites experienced.[30] Composites with 40 and 60 phr CCB (above the percolation), however, show a different behavior. These composites became more conductive after undergoing one heat treatment. That is, the resistivity values obtained during the cooling process at any particular temperature were lower than those obtained during the heating process, and thus after undergoing one heating–cooling cycle the composites had a lower values of resistivity than their initial ones. This can be explained as follows: composites containing conductive filler loading well above the percolation limit (composites containing 40 and 60 phr black) have a greater number of continuous conductive networks in the insulating polymer matrix compared to that of the composites with lower filler loadings; moreover, the average interparticle gap was also lesser. When such composites are subjected to heating there are quite a few phenomena simultaneously operative in the sys-
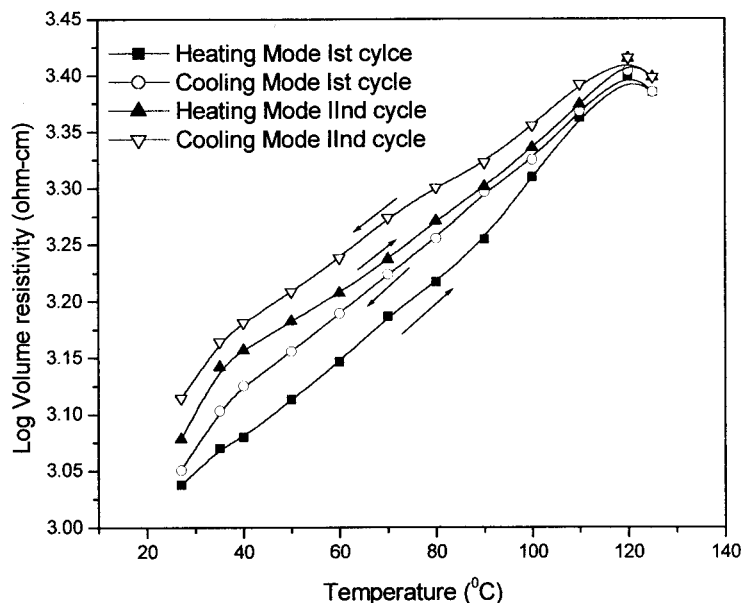

Figure 3 Effect of temperature on volume resistivity during repeated heating–cooling cycles for SBR–20CCB composites.
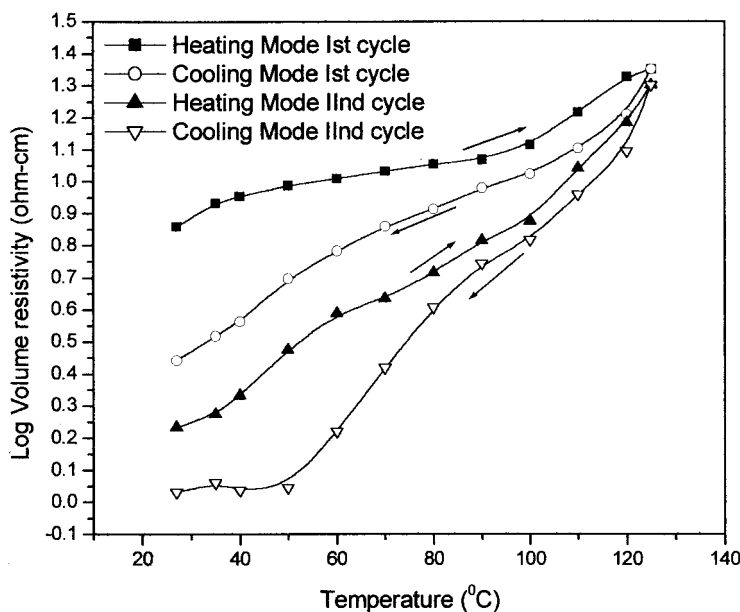
Figure 4 Effect of temperature on volume resistivity during repeated heating–cooling for SBR–60CCB composites.

tem: some that favor the breakdown of networks, like the differential thermal expansion of matrix and conductive network; and some that favor the formation of new networks, like flocculation of conducive particles and electron emission, and so forth. However, the increase or decrease in the conductivity is dependent on the net result of these effects. During cooling, the reformation of networks may occur because of contraction of the matrix. The formation of new networks thus becomes increasingly predominant during cooling, leading to higher conductivity at any specific temperature in the cooling cycle compared to that during the heating cycle, and this is expected to occur only in the case of high filler loadings where the average interparticle distance is less. In this article, two cycles of heating and cooling and their effects on the resistivity are reported. It can be seen that at 125°C, there is a slight decrease in the resistivity for almost all the composites. Aerial oxidation of the composites at this high temperature leads to generation of some polar groups, which decrease the resistivity of the composites.[31]

A plot of relative resistivity (ρ_T/ρ_o, where ρ_T is the resistivity at any temperature and ρ_o is the original resistivity at room temperature at the start of the heating–cooling process) against temperature was drawn for all the composite systems and is presented in Figure 5. It shows that the line representing the 30 phr filler loading alone deviates far apart from all the other lines. This can be explained by the following logic: at 30 phr loading the range of variation in the values of the resistivity is very broad; that is, the ρ_T values change substantially with temperature. However, in the case of other loadings the range of variation of resistivity is considerably less, which means

that the difference between the ρ_o and ρ_T is substantial in the case of 30 phr at any given temperature and this results in the curves as shown in Figure 5. This is because, in the case of loadings below and above the percolation, the change in the magnitude of resistivity is considerably less, whereas only at the percolation is the magnitude change very high. This can be taken as a supporting argument for claiming that the percolation limit is around 30 phr.

It has been observed that during rest between repeated heating–cooling cycles there is only a slight change in the initial resistivity at the start of the experiment of successive cycles. This phenomenon may be explained as follows: during the heating–cooling cycle there is simultaneous operation of two processes in the system: (1) the breakdown of the network structure attributed to uneven thermal expansion between conductive networks consisting of black particles and the matrix rubber; (2) the formation of some new networks attributed to agglomeration of black particles, flocculation, and so forth. The increase or decrease in conductivity is based on the predominance of either one of these processes. However, these processes do not stop in the samples, even during rest, after passing through a heating–cooling cycle. It means that, even during rest, some destruction and formation of the network proceed at a much slower speed, thereby leading to only a slight change in initial value of resistivity at the start of the next cycle.

Effect of pressure

Variation of volume resistivity with the applied pressure of the carbon black–filled systems is shown in Figure 6. The volume resistivity of all carbon black–
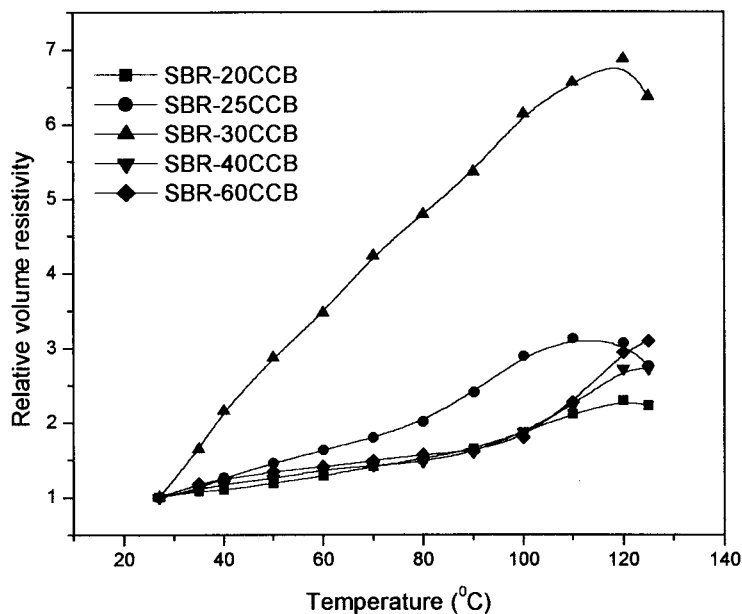
Figure 5 Effect of temperature on relative volume resistivity of SBR–CCB composites.

filled composites decreases sharply as the applied pressure is increased to a certain level, after which further increases in applied pressure exert only a marginal effect. In these composites, carbon black aggregates and polymer chains are interlinked. The application of pressure causes movement of the polymer chains, which affects the movement of the network structure of the conductive carbon black. The change in the resistivity with pressure can be explained by considering two phenomena that occur in the system: (1) the breakdown of existing conductive networks and (2) the formation of additional conductive networks.[32] The formation of this continuous conducting path occurs not only by direct contact between electrically conductive particles dispersed in the rubber matrix, but also when the interparticle distance is only a few nanometers and the electrons can easily jump across the gap. Thus, there is a threshold value for the interparticle gap that is electrically equivalent to the occurrence of interparticle contact. The formation of a continuous path at high applied pressure is facilitated by a decrease in interparticle gap in the discontinuous region. Also this enhances the significant contribution of the electron tunneling effect, thereby causing the
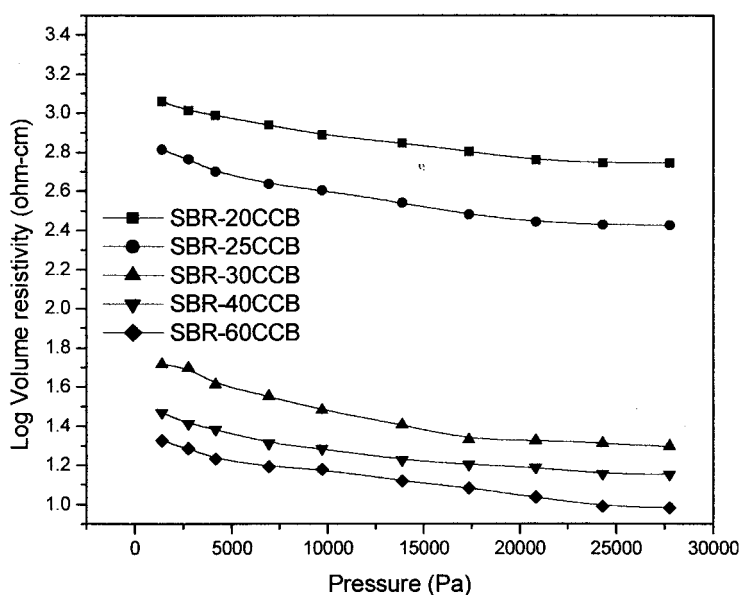

Figure 6 Effect of pressure on the volume resistivity of SBR–CCB composites.

TABLE IV
Equations of Best Fit for Variation of Volume Resistivity with Pressure for the SBR–CCB Composites

Filler loading (phr)	Equation of best fit
20	$\log \rho = 3.0387e^{-0.000004P}$
25	$\log \rho = 2.775e^{-0.000006P}$
30	$\log \rho = 1.6933e^{-0.00001P}$
40	$\log \rho = 1.4307e^{-0.000009P}$
60	$\log \rho = 1.3132e^{0.000009p}$

composites to exhibit high conductivity at high applied pressures.

From Figure 6 it may be observed that the volume resistivity of all the composites decreases exponentially with the applied pressure. All five plots corresponding to the respective filler loadings have been fitted into equations that describe their variation with pressure. The equations of best fit are shown in Table IV. It can be inferred that the general behavior of the composites with pressure can be expressed as

$$\log \rho = Ce^{-DP} \qquad (2)$$

where ρ is the volume resistivity (in ohm cm), C and D are numerical parameters, and P is the pressure (in Pa).

Equation (2) gives rise to two numerical parameters, C and D. From observation of Table IV, it can be seen that at any given pressure the parameters C and D are dependent mainly on the filler loading. The parameters in the case of pressure effect behave in a manner similar to that in the case of temperature. The parameter C behaves similar to parameter B in eq. (1) and

thus can be correlated similarly to the interfiller aggregate distance. The parameter D behaves similar to parameter A in eq. (1) and thus can be correlated similarly to the conductive network formation.

Effect of time under constant compression

Variations of volume resistivity against time for all the composites under a constant compression were studied and results are shown in Figure 7. The semilog plot of volume resistivity against time gives an overall quantitative picture of the variation of resistivity for all the composites. When the samples are held under compression, the volume resistivity of all the composites was found to decrease with time. However, the rate of decrease slows down with time and becomes marginal and assumes a constant value after some time. It was found that this phenomenon is strongly observed in lower filler (percolation < 30 phr) loaded composites than the higher filled (percolation > 30 phr) ones. This is because the systems having lower filler concentration contain only a fewer conductive networks and any small change in the networks affects more significantly the magnitude of resistivity, whereas the systems having higher filler concentration contain a large number of conductive networks and also the average distance between filler aggregates is small and thus the effect of compression is marginal, given that the destruction and formation process has only a marginal effect. The polymer chain mobility at lower filler loadings is expected to be greater than that at higher filler loadings. Also, the increase in conductivity with time for compressed samples, under constant strain, shows that a few new conductive net-
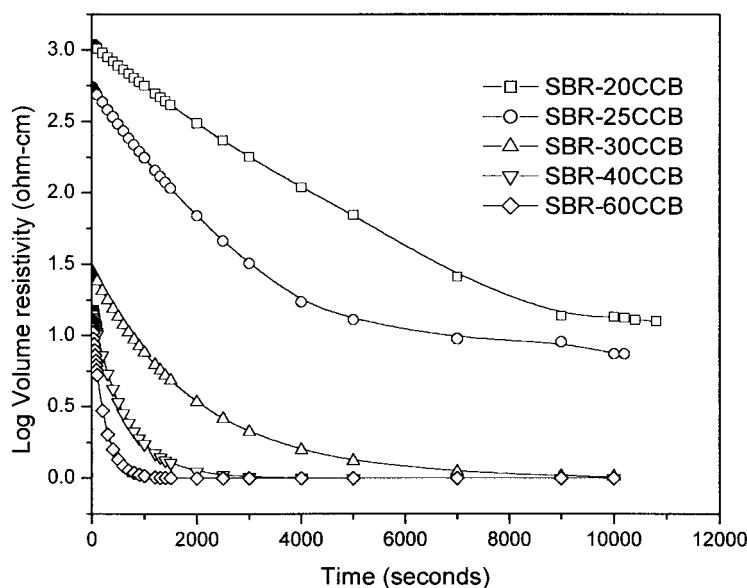

Figure 7 Effect of constant compressive stress on the volume resistivity of SBR–CCB composites.

TABLE V
Equations of Best Fit for Variation of Volume Resistivity with Time Under Constant Compression for the SBR–CCB Composites

Filler loading (phr)	Equation of best fit	Relaxation time (s)
20	$\log \rho = 3.0361e^{-0.0001t}$	4343
25	$\log \rho = 2.6318e^{-0.0002t}$	2172
30	$\log \rho = 1.4496e^{-0.0005t}$	869
40	$\log \rho = 1.1839e^{-0.0016t}$	271
60	$\log \rho = 1.1116e^{-0.0043t}$	101

works are also formed in the system, which may be attributed to the slow chain mobility of polymer chains under compression.[32]

The relaxation characteristics of the conductive samples were studied in terms of their electrical resistivity. From Figure 7 it can been seen that the volume resistivity of all the composites decreases exponentially with time. All five plots corresponding to the respective filler loadings have been fitted into equations that describes in general their variation with time. The equations of best fit are shown in Table V. It can be inferred that the general behavior of the composites with time can be expressed as

$$\log \rho = Xe^{-Yt} \qquad (3)$$

where ρ is the volume resistivity (in ohm cm), X and Y are numerical parameters, and t is time (in s).

From Table V it can be found that the parameter X in the equation decreases continuously with the increase in filler concentration and this may be correlated to the polymer chain mobility that occurs in the samples when subjected to a constant pressure. This is because the degree of chain mobility is expected to decrease continuously when filler loading is increased. It is believed that polymer chains are interlinked with filler aggregates and form some kind of physical crosslinks that restrict the chain mobility.[32] The parameter Y increases with the increase in filler loading and this shows that it can be correlated to the physical restriction exerted on the movement of polymer chains by the carbon black aggregates that act as physical crosslinks in the composites, which naturally increases with increase of filler loading. From this it can be inferred that the inversion or reciprocal of the physical restriction provided by the physical crosslinks is nothing but the degree of polymer chain mobility that is directly related to relaxation of the polymer matrix in the composites under compression. The time at which the volume resistivity curves of the samples becomes asymptotic to the x-axis in Figure 7 is called the relaxation time. When subjected to constant applied pressure, the polymer chains undergo a slow

relaxation process; consequently, during the slow and limited movement of polymer chains, the carbon black aggregates attached to polymer chains also undergo slow and limited movement. This movement of polymer chain and black aggregate facilitates both formation and destruction of the conductive network. Under this condition there is a net increase in the conductive network formation, thereby decreasing the resistivity.

To be more accurate the actual relaxation time is calculated from the simple variation of volume resistivity against time and is presented in Table V. It can be seen that the relaxation time decreases with increase in filler loading at any given instant. This is because the polymer chain mobility caused under a compressive stress is greater in the case of lower filler loadings because of the availability of a large amount of free polymer chains left unsurrounded and unlinked by the filler aggregates. Thus composites with low filler loading require more time to relax, whereas in the case of higher filler concentrations the available free polymer chains for movement are fewer in number, which restricts the polymer chain mobility, and thus these composites require less time for relaxation. Thus with the increase in filler concentration the polymer chain mobility decreases and thus the relaxation time decreases. This is reflected in the values of relaxation times as presented in Table V.

Mechanical properties

The mechanical performance of the SBR–CCB composites was studied in terms of tensile strength, 200% modulus, and elongation at break. SBR is considered to be a non–self-reinforcing rubber and conductive carbon black is regarded to be a semireinforcing filler. The variation of tensile strength is shown in Figure 8. Tensile strength is related to the energy required to deform and fracture the rubber chains. The effective crosslink density of the composites increases with increase in filler loading attributed to the increase of polymer–filler interaction with addition of the filler. Thus, at low filler loading, the energy required to deform and fracture the rubber chains is less because of lower crosslink density, whereas at high filler loading, attributed to the higher crosslink density, the energy required to deform and fracture the chains is high. This results in the increase of tensile strength with increase in filler loading. The variation of 200% modulus with filler loading is shown in Figure 9. It is observed that the modulus increases with increase in filler loading. This can be explained by the following mechanisms: at low filler loading the rubber chains are entrapped in the pores of the carbon black, a phenomenon termed rubber occlusion, which increases the effec-
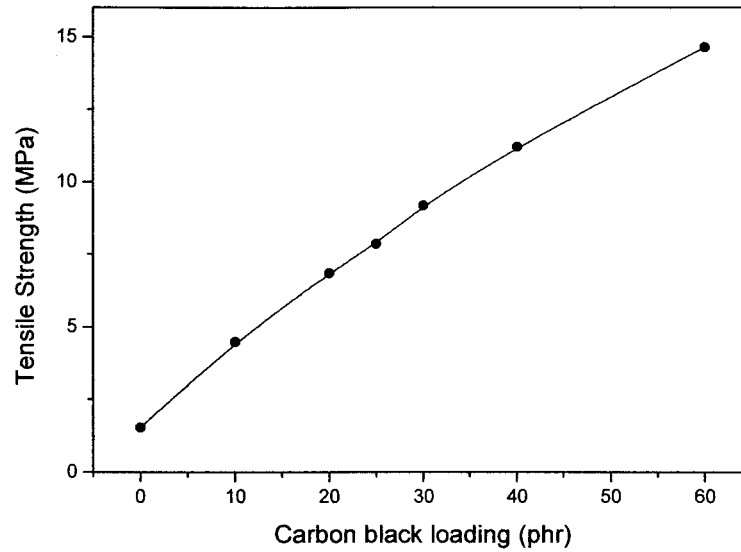
Figure 8 Effect of filler loading on tensile strength of SBR–CCB composites.

tive volume fraction of the filler. With further increase in filler loading other than occlusion, the rubber chains form a thin immobilized layer, called rubber shell, around the filler aggregates, which is responsible for the further increase in the modulus. Furthermore, the simple hydrodynamic effect also influences the increase in modulus. The variation of elongation at break with filler loading is presented in Figure 10. It can be seen that elongation at break increases with increase in filler loading to a maximum and then shows a decrease with further loading of the filler. This is because at very low filler loading, when the matrix is not sufficiently reinforced, it cannot sustain the load and so failure

occurs at lower elongation. However, with increase in filler loading, the matrix is progressively reinforced and a higher elongation at breaking point is observed. With further increase in filler loading, the molecular segmental motion is restricted because of the physical interaction between the filler aggregates and polymer chains. Consequently, the elongation at breaking point decreases with increasing filler loading.

CONCLUSIONS

SBR–conductive carbon black composites show positive temperature coefficient of volume resistivity. All
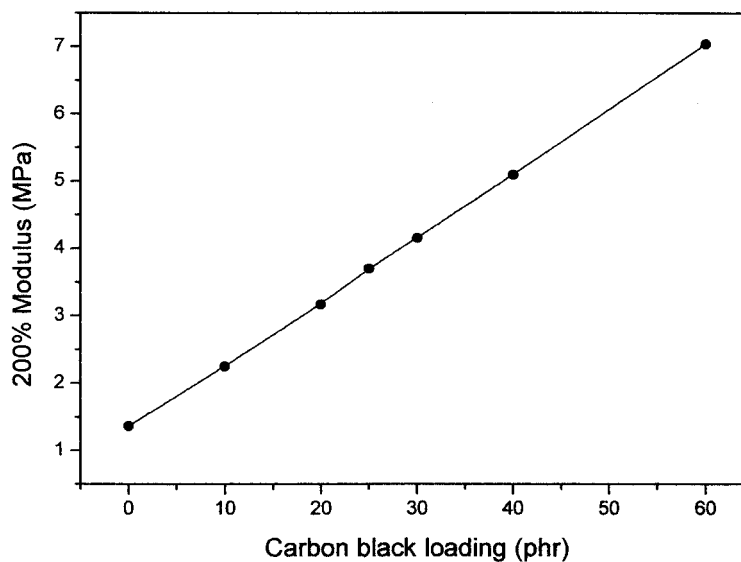

Figure 9 Effect of filler loading on 200% modulus of SBR–CCB composites.
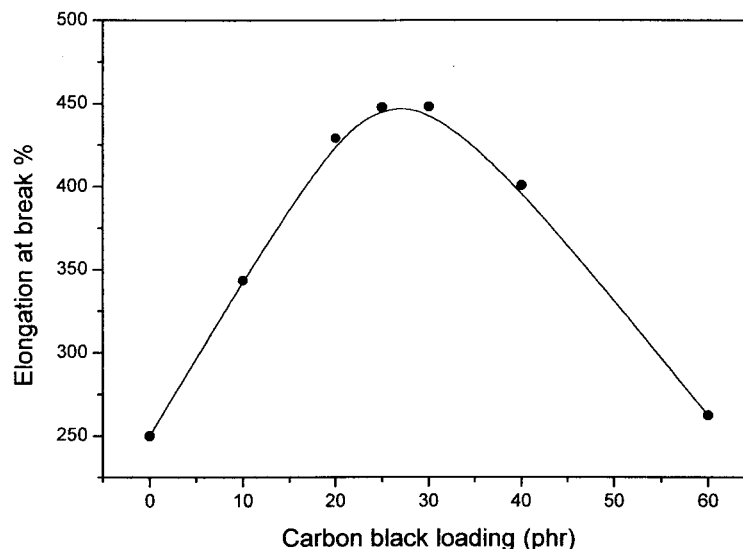
Figure 10   Effect of filler loading on elongation at break of SBR–CCB composites.

these composites exhibited electrical set and electrical hysteresis after the heating–cooling cycle. This reflects that there is a net change in some of conductive networks during the heating–cooling cycle.

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