Measurement of AC Conductivity and Dielectric Properties of Flexible Conductive Styrene–Butadiene Rubber-Carbon Black Composites

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ABSTRACT: Flexible conductive polymer composites were prepared using styrene–butadiene rubber (SBR) as a matrix and conductive carbon black as filler. The filler loading was varied from 10 to 60 phr. The effect of frequency, filler loading, temperature, and applied pressure on the AC conductivity, permittivity, and loss factor of the composites was studied. The AC conductivity of low and high loaded composites was found to be frequency dependent and independent respectively. The permittivity and the loss factor were continuously decreasing with increasing frequency. The increase in filler loading increased the AC conductivity, dielectric constant, and loss factor of the composites. Increase in temperature imposed increase in conductivity and permittivity of the composites. With increasing applied pressure the properties showed exponential increase. The effect of time under a constant compressive stress was studied and dielectric relaxation times were evaluated. © 2007 Wiley Periodicals, Inc. J Appl Polym Sci 104: 986–995, 2007

Key words: AC conductivity; dielectric constant; loss factor

INTRODUCTION

The dispersion of an electrically conductive particulate within an insulating host medium affects the overall performance of the heterogeneous system.[1] Furthermore, if the dispersed filler is in sufficient quantity, a conductive or semi conductive composite may be formed.[2,3] This type of material has been found to possess interesting properties, which are exploited in a variety of applications such as electromagnetic interference (EMI) shielding, electrostatic dissipation (ESD) of charges, electrically conductive adhesives, and circuit components in microelectronics.[4–7] Many empirical relations have been proposed describing their behaviors in terms of the permittivity and conductivity of their constituents.[8,9] Their electrical characteristics are related to the volume fraction of the particulate filler, the size and shape of the particles and also to factors, such as the method of processing and forming, and possible interactions between the conductive and the nonconductive phases.[10,11] The effect of temperature, pressure, and composition on the AC electrical properties has been reported in the literature.[12,13] The dielectric relaxation phenomenon of such composites has also been studied by many researchers.[14,15] Though many research works are available on the electrical properties of styrene–butadiene rubber (SBR)-carbon black composites (CCB),[16–20] no work is available where the effect of these many parameters on all the electrical properties for SBR with CCB as filler is studied.

The present work reports the findings of an experimental investigation on the changes in the alternating current (AC) conductivity, dielectric constant, and loss factor of composites made with SBR as the matrix, and conductive carbon black filler as a function of frequency, composition, temperature, pressure, and time.

EXPERIMENTAL

Materials

Styrene–butadiene rubber (SBR-1502, styrene content 23.5%, ML_{1+4} 100°C, 51) procured from Synthetic and Chemicals, Barielley, India. Conductive Carbon Black (CCB) Vulcan XC-72 was procured from Cabot India Ltd., Mumbai. Zinc oxide was procured from MERCK, Mumbai, India. Antioxidant TQ was procured from BAYER India, Mumbai, India. MBTS was procured from ICI India Ltd., Mumbai, India. Sulfur was procured from s.d.fine-CHEM, Mumbai, India. Other compounding ingredients were of chemically

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Ingredients	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

pure grade procured from standard suppliers. The formulation used for the preparation of the composites is given in Table I.

Sample preparation

The rubber and the various ingredients were mixed in a two roll mill by following the formulation given in Table I. The composites were cured at 150°C in an electrically heated press up to the optimum cure time, as obtained from Monsanto Rheometer R100. The cure cycle graph used for obtaining optimum cure time is shown in Figure 1. Since there is no reversion, the point of meeting of the tangent to the marching line and the tangent to the constant line is taken as optimum cure time. These vulcanized sheets were allowed to mature at room temperature for 24 h before testing.

Sample testing

The composites in the conducting range ($\rho \leq 10^4$ ohm-cm) (in this case SBR with 20 to 60 phr filler loading) were tested for the AC conductivity and permittivity properties. A cylindrically molded sample with a diameter 30 mm and height 12 mm was placed between two circular electrodes of a typical resistivity cell (home made) which is connected to a LCR meter (model 819, Goodwill Instek Co., Taiwan). The resistance, capacitance, and dissipation factor (tan δ) values were directly measured from LCR meter. From the values of capacitance and dissipation factor, the dielectric constant and loss factor of the samples were calculated. The dielectric constant (ε') of the composites were calculated through the capacitance by the fundamental equation,

$$\varepsilon' = ct/0.0885\,A \tag{1}$$

Where c capacitance of the sample in picofarads; t thickness of the sample in centimeter; A area of the sample in cm^2.

The loss factor (ε'') of the composites were calculated by the relation,

$$\varepsilon'' = \tan \delta\, \varepsilon' \tag{2}$$

where tan δ is the dissipation factor of the composites.

The frequency sweep range available with the instrument was 12 Hz to 100 kHz. For studying the effect of applied pressure on different electrical properties of samples, definite loads were applied on the samples placed in the resistivity cell as mentioned earlier and the range of pressure applied was varied from 1.39 to 27.75 kPa. A constant pressure of 6.94 kPa was applied on all the samples and the variation in resistivity over a period of 1800 s was recorded.

To check the effect of temperature on the electrical conductivity a special type of three-probe (positive, negative, and a guard electrode) sample holder fitted in a controlled heating chamber was used. Relatively thinner sample of thickness 2 mm was used for this measurement. The temperature sweep range was from room temperature to 125°C.

RESULTS AND DISCUSSIONS

Effect of frequency

AC conductivity of the composites

The conductance in conductive polymer composites is because of the formation of a continuous network structure by the conductive filler dispersed in the polymer matrix, and this happens only after addition of a certain critical amount of the filler. The loading of the conductive filler after which there is no significant change in resistivity irrespective of further
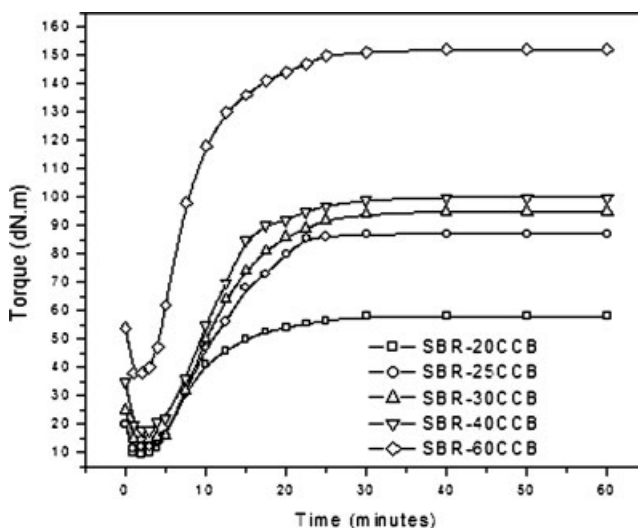
Figure 1 Cure cycle graph of SBR-CCB composites.
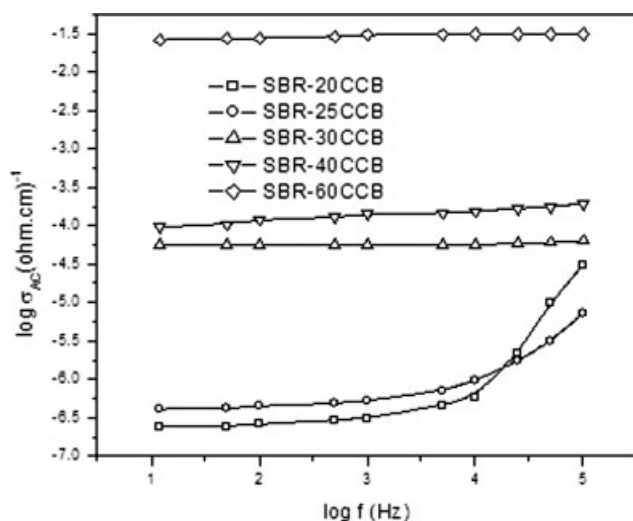
Figure 2 Effect of frequency on AC conductivity of SBR-CCB composites.

addition of the filler is called the percolation limit of the composite system. For the SBR-CCB system, the limit is occurring around 30 phr of carbon black.[21] In general the conduction of current in a conductive polymer composite follows three common mechanisms (1): flow of electrons through the conductive network, (2) electron hopping (jumping of electrons from one conducting particle to the next when the interparticle gap is sufficiently less), and (3) electric field radiation.[22,23]

The variation of AC conductivity (σ_{AC}) of different composites with frequency of applied electric field is shown in Figure 2. It can be observed from the figure that the composites near the percolation limit (containing 20 and 25 phr of filler) show a frequency dependent region and an almost frequency independent region of AC conductivity. For both 20 and 25 phr loaded composites conductivity was found to be marginally dependent on frequency from 12 to 1000 Hz and appreciably over 1000 Hz. The AC conductivity of the composites at and above the percolation (containing 30, 40, and 60 phr) exhibited total frequency independent nature in the measured frequency range. In the composites with 20 and 25 phr of filler, the frequency independent conductivity recorded at low frequencies can be attributed to resistive conduction through the bulk composite. On the other hand, at high frequencies, conductivity appears to be proportional to frequency due to the capacitance of the host medium between the conducting particles or aggregates. Also in these composites the continuous conductive network has just started to form with many conductive filler particles coming close to each other which give rise to appreciable increase in conductivity. This can be explained as follows. At high frequencies, the electrons are sufficiently excited so that they can hop from one con-

ducting cluster to another adding to the conductivity that is already existing because of the smaller interparticle gap. This leads to the increase in conductivity of these composites after a critical frequency (f_c).[24] The f_c for composites containing 20 and 25 phr were found to be $\sim$ 1000 Hz after which there is a marginal increase in conductivity with frequency and then high increase at higher frequencies.

It can be seen that at relatively high frequencies the hopping becomes very dominant so that the conductivity of the composites near percolation approaches conductivity of the composites above percolation. Also it can be interpreted that the formation of a continuous conductive network minimizes the hopping effect and this can be observed from the crossover of the conductivity of the composite containing 20 phr of carbon black to a higher value than that of the composite containing 25 phr of carbon black. The effect of frequency on AC conductivity can be explained with the following relation clearly. The measured AC conductivity of any dielectric system actually composes two components as shown in eq. (3).

$$\sigma_{AC} = \sigma_{DC} + \omega \varepsilon'' \qquad (3)$$

Where σ_{AC} AC conductivity in ohm^{-1} cm^{-1}; σ_{DC} DC conductivity in ohm^{-1} cm^{-1}; ω 2 π f; ω angular frequency and f the measurement frequency in Hz. ε'' dielectric loss factor mainly arising from dipolar and interfacial polarization.

The first component σ_{DC} is truly a DC conductivity component (ionic/electronic conductivity) and the other part arises out of polarization (restricted movement) of permanent dipoles/induced dipoles/accumulated interfacial charges (MWS). This equation reveals that as the frequency increases contribution of the second part due to polarization towards total conductivity increases. However the effect of actual mobility of dipole and induced dipoles mainly depends on the relaxation phenomenon. The effect of interfacial polarization becomes more significant at lower frequency of applied electric field as well as at higher temperatures. However the validity of this equation nullifies if the system becomes conductive not insulative (dielectric). In fact polarization part becomes meaningless in case of conductive system, because the total conductivity will be governed by DC component as it becomes significantly predominant. For samples which contain conductive additives beyond percolation limit (30, 40, and 60 phr) the conduction is due to the free flow of large number of charge particles through continuous conductive network available in the system. This is the reason, why at higher loadings of carbon black (above percolation limit), where system becomes conductive due to the formation of conductive network of car-
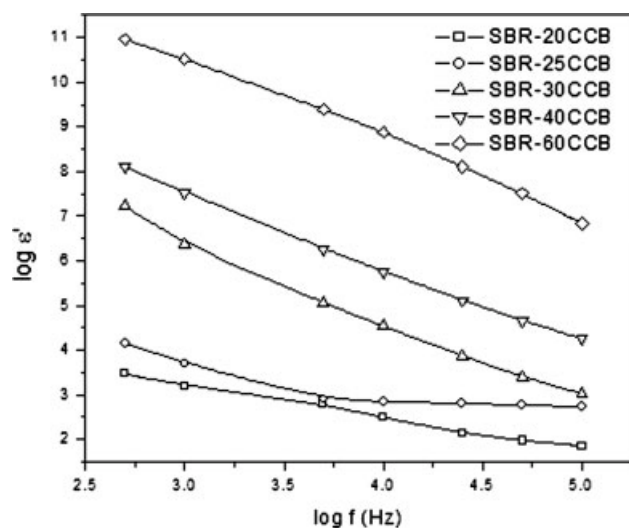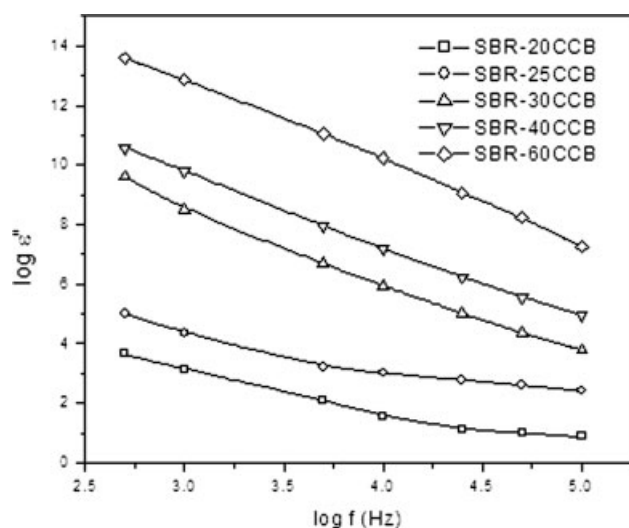
Figure 3 Effect of frequency on permittivity of SBR-CCB composites.

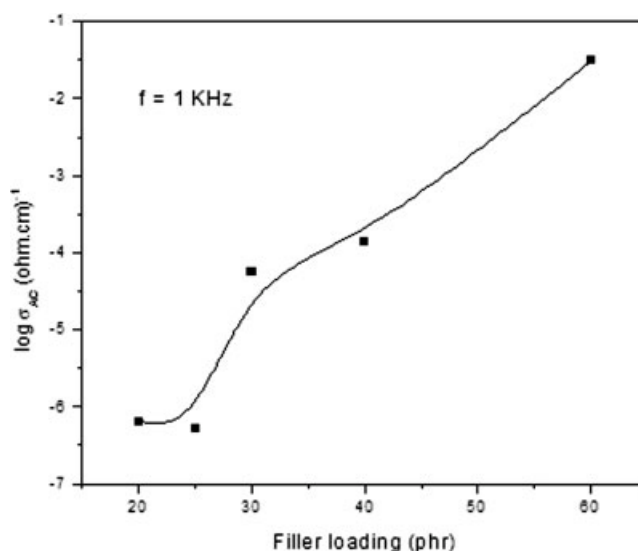**Figure 5** Effect of filler loading on AC conductivity of SBR-CCB composites.

bon black in insulating rubber matrix, the magnitude of conductivity increases several folds and conductivity becomes independent of frequency. At such situation σ_{AC} is purely governed by the value of σ_{DC}, which has grown significantly higher.

Permittivity and loss factor of the composites

The variation of ε' and ε'' with AC frequency of the composites were studied and presented in Figures 3 and 4. It can be observed that both the dielectric constant and loss factor decreases with increase in frequency. At low frequencies, both the ε' and ε'' attains high values and then decreases exponentially with increase in frequency. This behavior clearly indicates that the effect of interfacial polarization,

known as Maxwell-Wagner-Sillars effect (MWS),[25,26] becomes more and more predominant at lower frequency. MWS polarization occurs in dielectrically heterogeneous system because of the accumulation of charges at the interfaces. The present system consists of insulating SBR matrix having conductive filler carbon black. Moreover, the contribution of DC conductivity to dielectric loss factor also increases with decrease in applied electric field frequency. The decrease in permittivity and loss factor with increase in frequency can also be explained by the fact that as the frequency is raised the dipoles get very less time to orient themselves in the direction of the alternating field.[27]

Effect of composition (filler loading)

AC conductivity of the composites

The effect of the conductive carbon black loading on the AC conductivity of the conductive SBR-CCB composites have been studied and shown in Figure 5. The variation in the conductivity of the composites with increase in filler loading (from 20 to 60 phr) at a selected constant frequency of 1 kHz is shown in the plot. Already in a previous work by these authors, it was shown that the SBR-CCB composites become marginally conductive at 20 phr, whereas the percolation limit was 30 phr.[21] It is obvious and also observed that with the increase in the filler loading the conductivity of the composites increases. This is because of the process of formation of a continuous conductive network, which keeps on increasing with increase in filler loading. Since, the magnitude of AC conductivity is dependent on the frequency also, the mechanisms of current flow

Figure 4 Effect of frequency on loss factor of SBR-CCB composites.
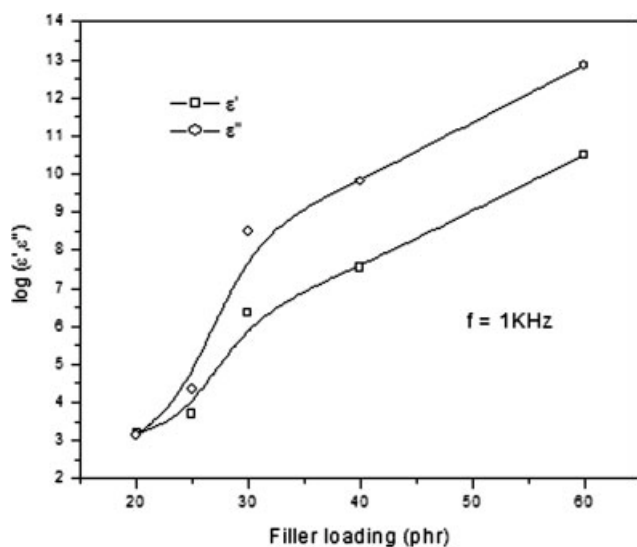
Figure 6 Effect of filler loading on permittivity and loss factor of SBR-CCB composites.

through the continuous network as well as the electron hopping mechanism both operates in the composites, especially at high frequencies which can be explained by the eq. (3) and the subsequent descriptions as above.

Permittivity and loss factor of the composites

The influence of composition of the conductive carbon black on the permittivity and loss factor of the composites were studied and shown in Figure 6. It was found that with the increase in filler loading both ε' (dielectric constant) and ε'' (loss factor) were increasing continuously and this is due to the increased contribution of interfacial polarization associated with the presence of carbon black particles in the insulating rubber matrix. As the conductive filler content is increased there is also increased contribution of conductivity mainly to dielectric loss factor. Thus, the increase in overall interfacial polarization along with increased ionic conductivity due to the increase in filler loading causes the increase in ε' and ε'' of the composites.[27] It should also be noted that the composites with higher filler loadings can reach very high dielectric constant of the order of 10^{10}.
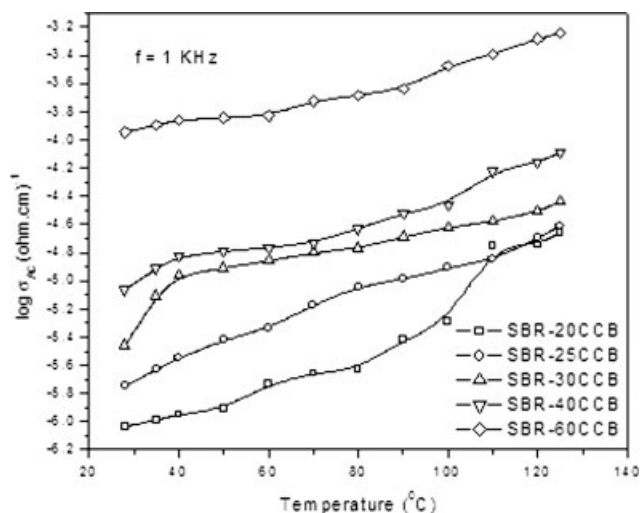
Effect of temperature

AC conductivity of the composites

The effect of temperature on the AC conductivity of the composites was investigated at a constant frequency of 1 kHz and is represented in Figure 7. From the plots it can be seen that for all the composites the conductivity is increasing with the increase

in temperature. At higher temperature and at high frequency of applied electric field, it can be assumed that all the three conducting mechanisms mentioned earlier will be operating and the net result has an enhanced effect, i.e., conduction through electron hopping gets magnified. Also, the increase in temperature perhaps have some positive influence on the flow of current by the enhanced effect of electric field radiation. However, the differential thermal expansion between matrix polymer and conductive filler has negative effect on the change in conductivity against temperature: as the average thermal expansion of rubber matrix (660×10^{-6} K^{-1}) is substantially higher than that of carbon black.[28] It should be noted that unlike the DC conductivity,[21] here the differential thermal expansion between the rubber matrix and the filler is becoming recessive compared to the other mechanisms. The effect of dipoles and interfacial (MWS) polarization gets enhanced at higher temperature and leads to increased conductivity.

Permittivity and loss factor of the composites

The effect of temperature on the permittivity and loss factor of the composites was investigated at a constant frequency of 1 kHz and presented in Figures 8 and 9. From the curves it can be clearly seen that with the increase in temperature both ε' and ε'' are increasing exponentially for all the composites, though the increase becomes weaker with increase in filler loadings. When temperature is increased a gradual increase in dielectric properties is observed. This is mainly because of the fact that both interfacial polarization and ionic conductivity undergoes an increase when there is a rise in temperature and
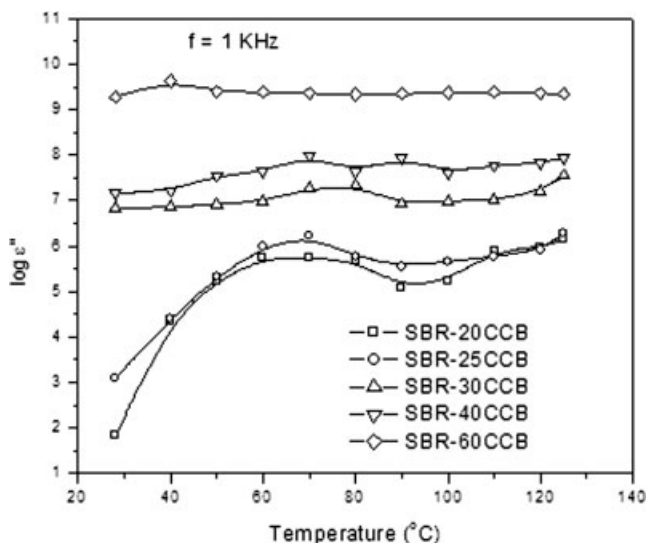

Figure 7 Effect of temperature on AC conductivity of SBR-CCB composites
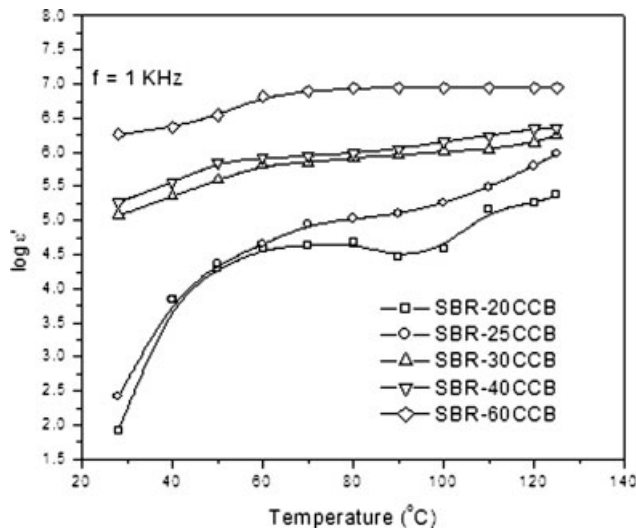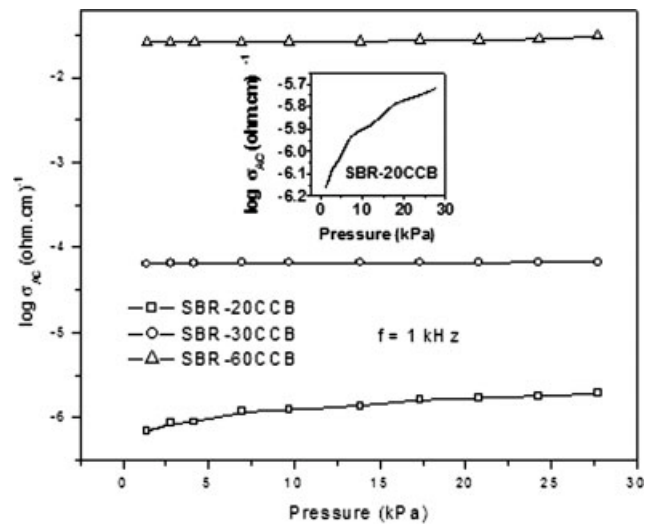
Figure 8 Effect of temperature on permittivity of SBR-CCB composites.

Figure 10 Effect of pressure on AC conductivity of SBR-CCB composites.

this leads to increase in permittivity and loss factor. Also, segmental mobility of polymer chain increases, which favorably affects the conductive network formation and thus affect the dielectric properties. Further the difference in thermal expansion of matrix polymer (which is generally higher) compared to that of conductive filler aggregate is expected to affect dielectric properties adversely, when temperature is increased. So the net effect will be overall effect of both positive and negative contributions.[14] This behavior is observed more in the case of lower filler loaded composites, where the nature of polymer matrix plays a key role whereas in high filler loaded composites the nature of the rigid filler aggregates play a key role than the matrix and hence

the effect of temperature is experienced less in these composites.

Effect of applied pressure

AC conductivity of the composites

The variation of AC conductivity of the composites with applied pressure at a constant frequency of 1 kHz is shown in Figure 10 and inset. The conductivity increases with increase in pressure up to a certain amount of pressure and then becomes constant. In a previous work by the authors[21] DC resistivity was found to decrease with applied pressure and here the same is confirmed through the AC conductivity and the same explanations holds good. This behavior of change in conductivity against applied pressure is found to be true only for composites near percolation (20 and 25 phr) loading of filler whereas in the case of composites containing 30 to 60 phr (at and above percolation) loading of filler the effect of pressure is found to be marginal. Whenever, a polymer composite is subjected to pressure two phenomena takes place simultaneously: (1) the breakdown of the existing conducting networks under the applied pressure and (2) the formation of new conducting paths because of the slow movement of the polymer chains along with the filler aggregates under the applied pressure.[29] In the case of lower filler loadings (20 and 25 phr), the conducting network available for the flow of current itself is very less and hence the breakdown process is relatively slower than the movement of polymer chains with the filler aggregates. This facilitates an appreciable decrease in the interfiller aggregate distance forming new conducting paths thereby giving rise to the chance of more conduction. Also at these small

Figure 9 Effect of temperature on loss factor of SBR-CCB composites.
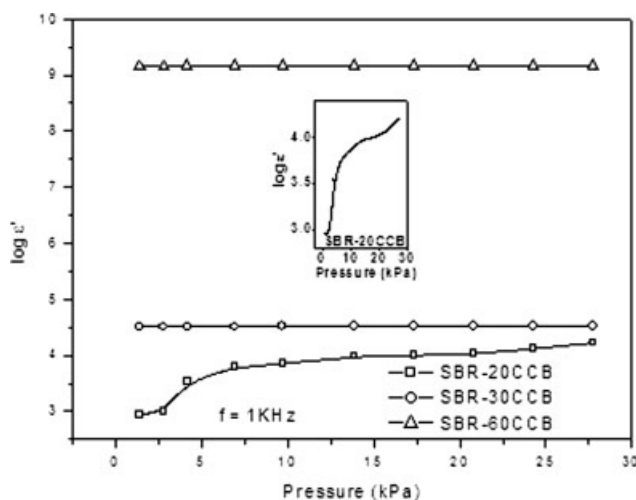
Figure 11 Effect of pressure on permittivity of SBR-CCB composites.

interfiller aggregate gaps the electron tunneling mechanism of conduction also takes place. Thus, there is an increase in conductivity with pressure, but, after a critical pressure the polymer chains are unable to move further making the effect of pressure insignificant. In the case of higher filler loadings (30–60 phr) the movement of polymer chains is hindered by the enormously available rigid filler aggregates thus formation of new paths becomes negligible. Also in these composites there is a complete continuous conductive network and the smaller destructions caused by the applied pressure has lesser effect. Hence, the effect of pressure on the conductivity of these composites is negligible.[21,29]

Permittivity and loss factor of the composites

The variation of permittivity and loss factor of the composites with applied pressure at a constant frequency of 1 kHz is shown in Figures 11 and 12. It can be seen that with increase in pressure both ε' and ε'' increases and then attains a steady state value at and beyond the pressure of 10 kPa for 20 phr sample (please see inset). The dielectric constant and loss factor of all carbon black-filled composites, in general, increases sharply with increase in applied pressure up to a certain level and further increase in applied pressure has only a marginal effect. But for the systems having higher loading of black (at and beyond percolation threshold), the effect of pressure on dielectric constant and loss factor become marginal. For any lossy dielectric having conductive component dispersed in insulating medium, like the present system, its capacitance is directly proportional to the area of the conductor and inversely proportional to the distance between the conductors. In conducting polymer composites, the capacitance

arises from induced double layers on the surfaces of conducting particles, which are separated by non-conducting polymer layers[30] and this capacitance increases with increase in applied pressure due to reduction in the distance between the conducting filler particles with increase in pressure, because of the fact that distance between the conductors is inversely related to capacitance as mentioned earlier.

Thus, when the applied pressure is increased the filler aggregates in the composites come closer together, which decreases the interparticle gap between them and in turn leads to increase in capacitance, and hence the permittivity.[31] Also, it must be noted that when plotted individually all the composites show this pressure effect, but, the effect is pronounced more in composites having lower filler loading than the highly filled ones. This is because whenever a low loaded composite (SBR-20CCB) is subjected to pressure, the polymer chains are more free to move without much restrictions from the filler aggregates and hence, the interfiller aggregate distance decreases to a significant extent leading to increase in capacitance and hence the permittivity. In the case of high loaded composites, the mobility of the polymer chains is restricted by the presence of large amount of filler (reinforcement) and thus the changes in interfiller aggregate distance is much lesser as compared to the low loaded composites and hence the changes in dielectric properties are marginal.
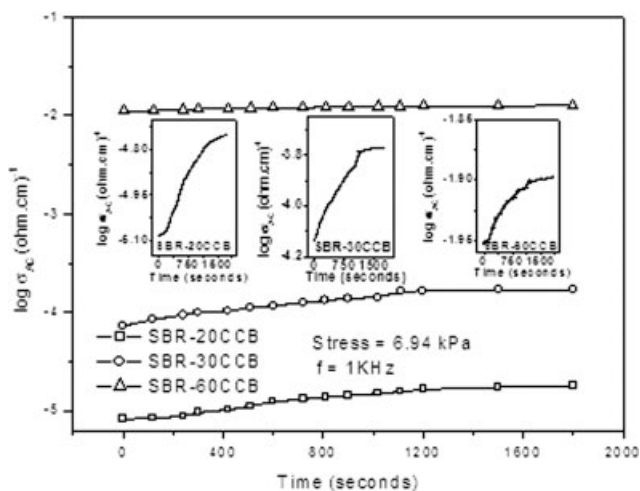
Effect of time under constant compressive stress

AC conductivity of the composites

The effect of a constant compressive stress on the AC conductivity of the composites over a time span have been investigated and presented in Figure 13. It
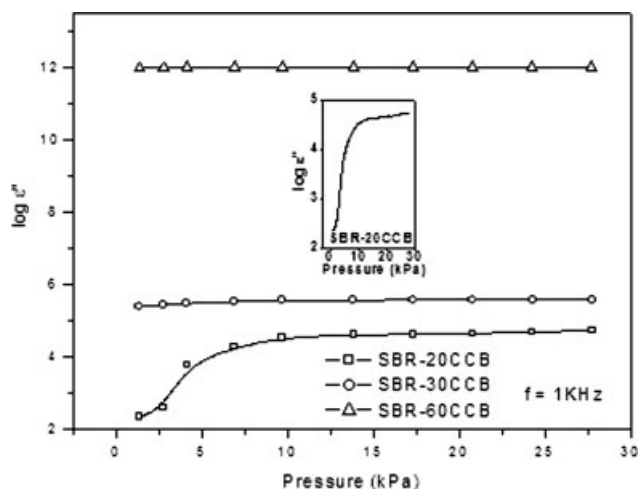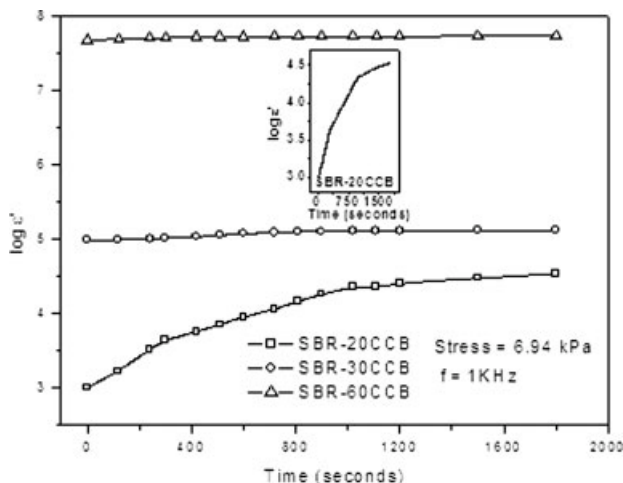

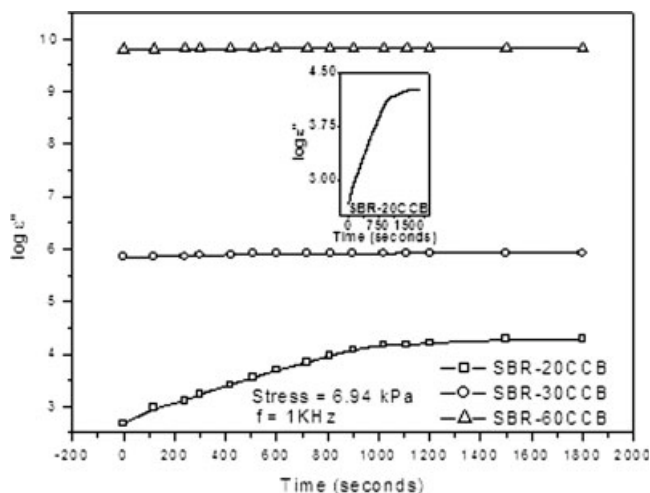**Figure 12** Effect of pressure on loss factor of SBR-CCB composites.

Figure 13 Effect of constant compressive stress on AC conductivity of SBR-CCB composites.

Figure 15 Effect of constant compressive stress on loss factor of SBR-CCB composites.

can be observed from the curve that the AC conductivity of the composites near the percolation (20 and 25 phr) increases with time and then attains a constant value independent of time. In the case of composites at and above percolation (30–60 phr) similar trend is observed when the curves are plotted individually for each system (as shown in inset). However, the effect of time on conductivity for compressed samples are more pronounced in case of samples having lower loading of carbon black (20–30 phr) rather than highly filled systems (40–60 phr). This may be due to the fact that at higher loading interparticle gap is less, so change in conductivity with stress as well as time becomes less significant. Moreover the movement of polymer chains under the stress is hindered by the presence of rigid filler aggregates. In the case of samples with lower loading, as explained earlier, the slow movement of poly-

mer chains along with the filler aggregates plays a key role in forming new conducting paths, thereby increasing the conductivity exponentially with time slowly approaching a constant.

Permittivity and loss factor of the composites

The effect of a constant compressive stress over a time span on the dielectric constant and loss factor of the composites are shown in Figures 14 and 15. It is observed that both dielectric constant and loss factor were increasing exponentially and reaches a constant value after sometime. The increase in permittivity as well as loss factor can be explained in the same way as in the case of their behavior with applied pressure. It can be said that a molecular relaxation takes place with time under a constant compressive stress, which affects the dielectric properties of the systems through formation and destruction of conductive network in insulating matrix. The time after which there is no appreciable change in the value of the dielectric constant is termed as the relaxation time of the composites. The values of relaxation times were calculated based on the change in permittivity against constant applied stress by fitting the plots in Figure 14 to equations. The equations of best fit for all the composites are shown in Table II. The fitted equations obtained can be expressed generally as follows:

$$\log \varepsilon' = X\,e^{Yt} \tag{4}$$

where ε' is the permittivity of the composites, t is the time in seconds, X and Y are the numerical parameters obtained from the equations of best fit. The calculation of relaxation time can be explained in the same fashion as has been done in our previous

Figure 14 Effect of constant compressive stress on Permittivity of SBR-CCB composites.

TABLE II
Equations of Best Fit for Variation of Permittivity with Time under Constant Compressive Stress and Relaxation Time for the SBR-CCB Composites

Filler loading (phr)	Equation of best fit	Relaxation time (s)
20	$\log \varepsilon' = 3.3481\ e^{-0.0002t}$	5000
25	$\log \varepsilon' = 4.7424\ e^{-0.0005t}$	2000
30	$\log \varepsilon' = 5.1496\ e^{-0.00083t}$	1200
40	$\log \varepsilon' = 5.1839\ e^{-0.0016t}$	600
60	$\log \varepsilon' = 5.2116\ e^{-0.0047t}$	211

work, except for the difference that there it was done based on resistivity and here it's done on the basis of permittivity.[21] Out of the two parameters, parameter Y is important for determining the relaxation time as it is the exponent part of the time axis. It can be seen that 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 which acts 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, which is directly related to relaxation of the polymer matrix in the composites under compression. Hence, it may be concluded that the reciprocal value of parameter Y in eq. (4) represents the value of t_R, the relaxation time. The time at which the permittivity curves of the samples becomes asymptotic to the time axis in Figure 14 is called the relaxation time. When subjected to constant applied pressure, the polymer chains undergo 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 formation and destruction of conductive network. Under this condition, there is a net increase in the conductive network formation thereby increasing the permittivity.

From the values of Y the relaxation times for all the composites have been calculated and have been shown in Table II. It can be seen that the relaxation time decreases with increase in filler loading for any given instant. This is because the polymer chain mobility caused under a compressive stress is more 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, and hence the composites with low filler loading requires more time to relax. Whereas, in the case of higher filler concentrations the available free polymer chains for movement are less, which makes the

polymer chain mobility restricted and thus these composites require less time for relaxation. Thus with the increase in filler concentration the polymer chain mobility decreases and hence the relaxation time falls down. This is reflected in the values of relaxation times as presented in Table II.

SUMMARY AND CONCLUSIONS

The AC conductivity of low filler loaded composites exhibits a frequency dependent nature whereas the high filler loaded composites are frequency independent due to the increased contribution of DC conductivity for highly loaded samples. The AC conductivity of the composites increased with increasing temperature and the applied pressure due to the changes in interfacial polarization. While, the dielectric constant and loss factor of the composites decreased with increasing the frequency. The strong effect of interfacial polarization is operative in black filled SBR composites. Ease of polymer chain mobility, which affects the conductive network formation and breakdown in the insulating matrix, in case of systems having low filler loading, is responsible for significant variation of AC conductivity and dielectric properties against applied pressure, temperature, and time. Whereas the effect of these parameters on AC conductivity and dielectric properties is marginal in the case of systems having high filler loading which restricts the polymer chain mobility. Relaxation time for polymer chain movement under constant compressive stress decreases with increasing filler loading.

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